

Development of groundwater radon continuous monitors: Comparison between α scintillation and γ spectrometry systems^(*)

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Summary. — Two temporised continuous monitoring systems, designed to measure the radon concentration in natural environments, mainly groundwater, were assembled, tested and cross-check compared, evaluating the background noise, sensitivity, calibration values and soundest application in the Earth Science framework. The two systems have been customised by DINCE Laboratory, based on best-fitting criteria selected according to the ING laboratory, partially in the frame of two EC funded, *Geochemical Seismic Zonation (GSZ)* and *Automatic Geochemical Monitoring of Volcanoes*, addressed to earthquake prediction research and prototype developing, aimed to seismic and volcanic risks surveillance. Following best-fitting criteria of the radon monitoring aimed to natural risk research, both systems are operative by discrete temporised sampling of an aliquot of groundwater, with a minimal interval of six hours. During their functioning at the ENEA Centre of Frascati (Rome), the test-site chosen, both systems provided a continuous and reliable response.

PACS 91.30.Px – Phenomena related to earthquake prediction.

PACS 92.40.Kf – Groundwater.

PACS 29.40.Mc – Scintillation detectors.

PACS 82.80.Ej – X-ray, Mössbauer, and other gamma-ray spectroscopic analysis methods.

1. – Introduction

The development of new radon continuous monitoring instrumentation has been a scientific open field since '60, after the first discovering about the possible use of radon as earthquake forerunner [1-7].

^(*) The authors of this paper have agreed to not receive the proofs for correction.

The *Department of Nuclear Engineering and Conversion of Energy of the University of Rome "La Sapienza"* (DINCE) spent many efforts, since '90, to the design, assembling, testing and certification of different discrete and continuous radio-nuclides monitoring systems, reserving special attention to radon and its daughters [8,9].

The new "radon sensors" developed by DINCE and ING [10-12] have been mainly conceived as part of the *Geochemical Monitoring System* (GMS II) prototype, that is a fully versatile multi-parametric geochemical continuous monitoring system, designed by ING for natural hazard surveillance [13-15].

Very often, in earthquake surveillance studies, the discrete radon measurement has been coupled with other geochemical parameters, mainly gaseous as He, CH₄, Ne, CO₂, H₂, H₂S, etc., while a full multi-parametric approach is lacking in continuous monitoring. Therefore the scientific needs and the experience of ING and DINCE respectively have permitted to conceive the radon monitor prototypes also as part of the GMS II remote station design. The GMS II development is conceived to fill up this lack, using the remote station as "*test-stand*" for new improved geochemical, hydrological, environmental and geophysical sensors, managed separately, as analogic output for a digital serial collector. These are in continuous evolution over the market, linked to the environmental quality control and applied geophysics.

In this paper we will describe the development of two radon prototypes, the first one adopting α scintillation (system A) and the second one adopting γ spectrometry (system G). These two devices have been compared as regards sensitivity and performances.

2. – Two devices for groundwater radon concentration measurement

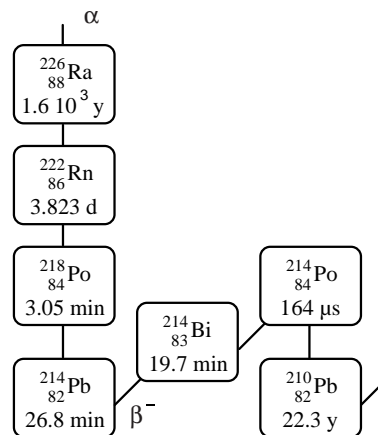
Part of the decay chain related to the ²³⁸U radioactive family from ²²⁶Ra to ²¹⁰Pb is reported in fig. 1. The isotopes of interest for ²²²Rn measurement are, besides radon itself, as α emitter, its four short-lived daughters, whose main emissions are reported in the table annexed to fig. 1. As we can see, ²²²Rn and two of its daughters decay by α emission, while the other two by γ emission. Therefore, the measure of radon concentration in groundwater can be performed either by revealing α -particles emitted by ²²²Rn, ²¹⁴Po and ²¹⁸Po or by revealing γ -rays, following the β^- decay of ²¹⁴Pb and ²¹⁴Bi. In both cases the correlation between counts obtained and radon concentration value is possible only in *secular equilibrium condition*, *i.e.*, after three hours since sampling from source at least.

Two systems for groundwater radon continuous measurement have been customised by DINCE Laboratory, based on best-fitting criteria selected according to the ING laboratory, devoted to earthquake prediction research and prototypes developing, addressed to seismic and volcanic risk surveillance: both systems are operative by discrete temporised sampling of an aliquot of groundwater, with a minimal interval of three hours before counting.

The first system is based upon the α revealing, using an α *scintillation cell* (system A) and the other one is based upon the γ spectrometry adoption (system G).

In system A the measuring device is made up of an α *scintillation chamber*, coupled with a photomultiplier: it is located within a 3.4 litres (net volume of 2.9 litres) PVC cylinder, assigned to contain the degassing air of the groundwater samples contained in a beaker of 0.5 litres (fig. 2) [12].

A measuring cycle consists of the following succeeding operations: groundwater enters the beaker, near the bottom and overflows on top, flowing for a time interval necessary to a complete renewal of the water initially contained within the beaker, measured in the



Radionuclide	Energy of main emitted radiations (MeV)		
	α	β	γ
^{222}Rn	5.49		
^{218}Po	6.00		
^{214}Pb		0.67	0.242
		0.73	0.295
		1.02	0.352
^{214}Bi		1.0	0.609
		1.51	1.120
		3.26	1.764
^{214}Po	7.69		

Fig. 1. – Main characteristics of ^{222}Rn and its progeny.

previous cycle. For this renewal a few litres are needed. At the same time a diaphragm pump performs firstly a cycle of renewal of the air contained within the cylinder, by air drawn off from the atmosphere, and secondly it creates a partial vacuum.

At the end of these two routines, the opening of a needle valve allows the gurgling of the atmospheric air within the water contained in the beaker. Then the enriched air enters the cylinder until the pressure equilibrium is reached. Selecting a partial vacuum of 0.65 bars, a back-flux of around 1.9 litres of air occurs (at the normal environmental conditions); this aliquot is enough to obtain a nearly complete degassing of the radon dissolved in the water sample ($\sim 90\%$).

This vapour saturated air (relative humidity around 100%), mixed within the cylinder with the atmospheric air, results in a non saturated mixture, avoiding water condensation on the α scintillation chamber walls and on the photo-multiplier window.

After around three hours, the scintillation counting—produced by the α -particles from ^{222}Rn , ^{214}Po and ^{218}Po decays—is finally accomplished.

The electronic chain is made up of an amplifier and a discriminator, and it is linked to a counting system, which processes pulses with amplitude larger than the threshold of the discriminator.

In the system G a *Marinelli Beaker* (BM) with volume of 0.69 L is located around a $2'' \times 2''$ NaI(Tl) detector surrounded by a shield made up of lead pellets, enclosed in a cylinder of 30 cm of diameter, in order to facilitate the transport. The shield thickness is $> 55 \text{ g/cm}^2$, equivalent to 5 cm of melted lead.

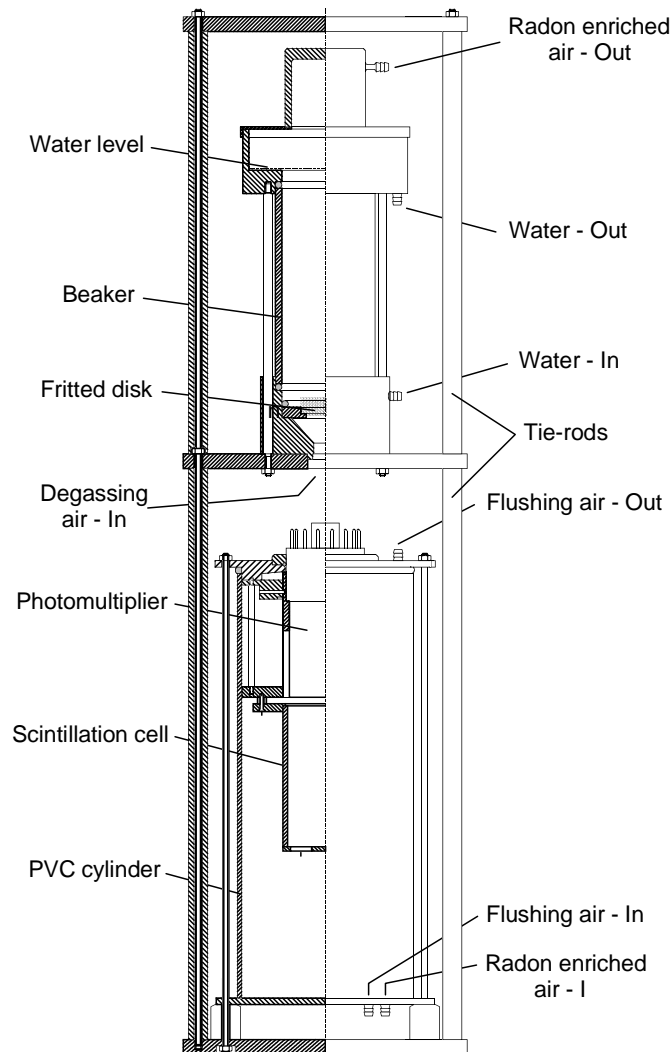


Fig. 2. – Measurement system of the system A. Details of the degassing unit.

The *Region of Interest* (ROI) of the spectrum is that ranging from the γ lines of ^{214}Pb to the 609 keV line of ^{214}Bi ; it is dashed in the spectrum reported in fig. 3, obtained by the electronic operative system. The electronic operative system is made up of a multi-channel card for PC (*EG & G, U.S.A.*™), that makes possible the memorisation of the spectrum related to each measurement.

The spectrum recording and the subsequent possibility to check the entire system represents an advantage with respect to the system A. In fact the α scintillation generates pulses that are not correlated with the energies of the α -particles, subject only to a counting starting from the threshold codified value of the discriminator.

The two systems have been developed and cross-checked at the DINCE Laboratory; in May 1999 they were installed at the ENEA Centre of Frascati, the test-site chosen.

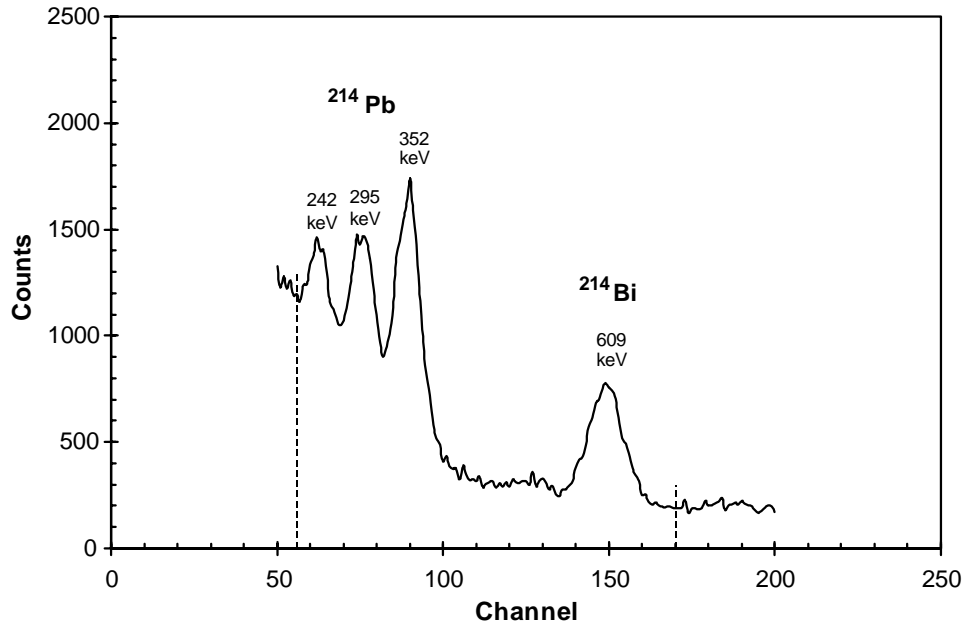


Fig. 3. – γ spectrum obtained with a $2'' \times 2''$ NaI(Tl) scintillator, related to radon daughters. The selected *Region of Interest* (ROI) is delimited by the dashed lines.

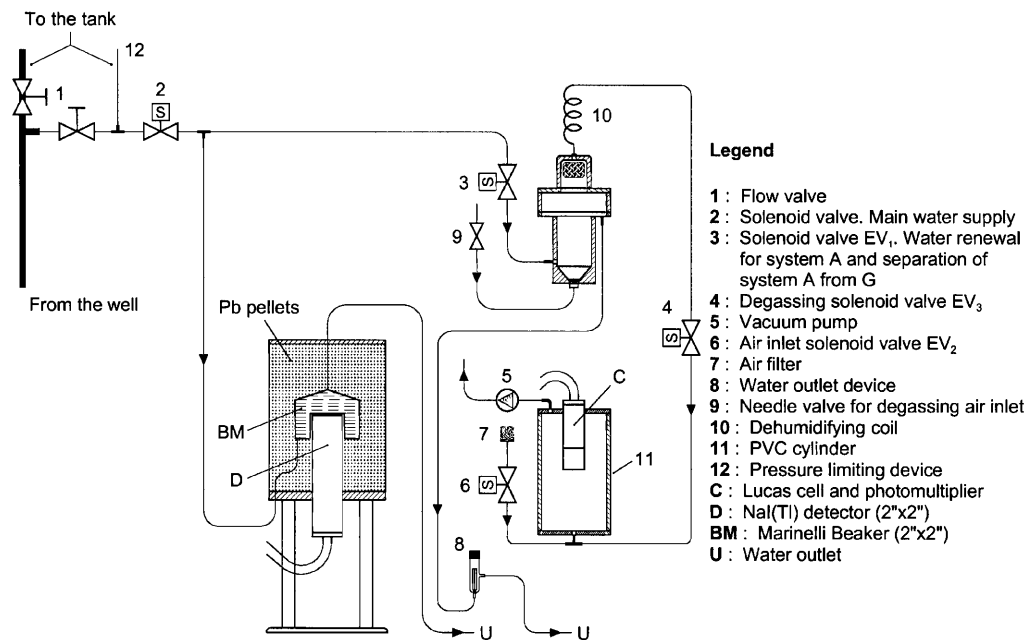


Fig. 4. – Hydraulic scheme of the two systems (A,G) tested together.

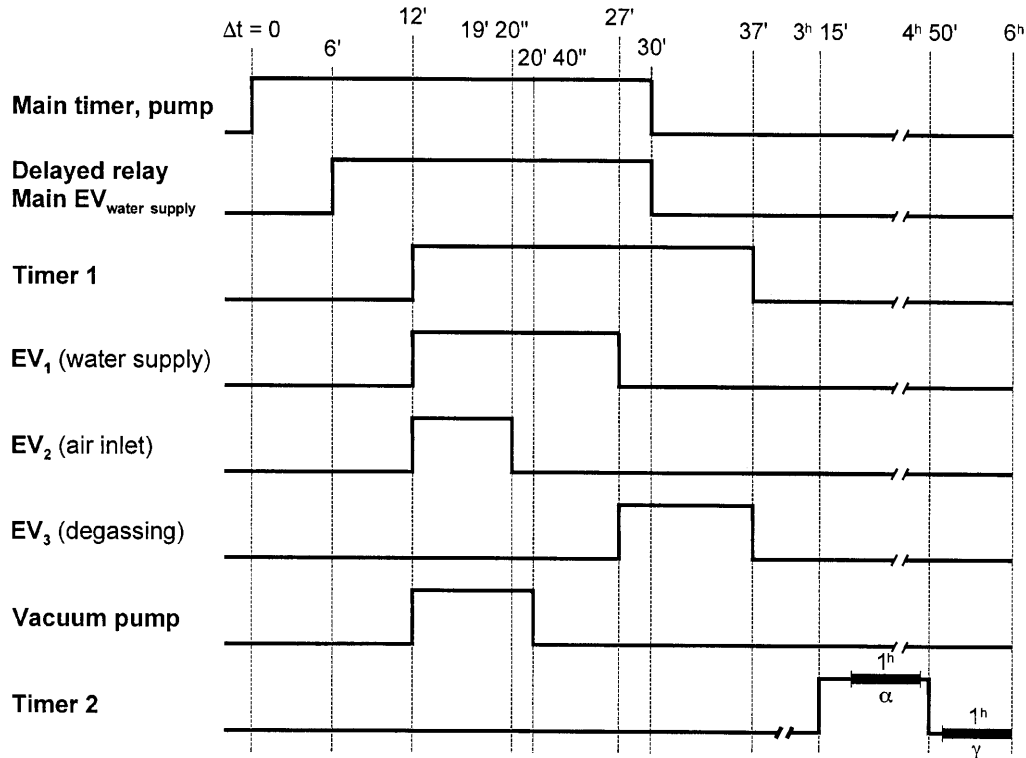


Fig. 5. – Phases of the measuring cycle. Activation logic.

Their layout is shown in fig. 4. Water is pumped out of a deep well at scheduled intervals according to the system requirements; precisely water is supplied only after its flow is stabilised by using a solenoid valve (EV) driven by a delayed relay cascaded to the timer which operates the pump.

The two systems A and G are located in parallel with respect to each other so as to make it possible the exclusion of one of them on demand.

On the one hand, the BM of system G is water supplied directly by the main EV according to the procedure described above; on the other hand, the system A requires further steps: a timer codified by scheduled intervals (timer 1) starts the succession of routines by both opening the EVs and turning ON the diaphragm pump, as shown in fig. 5. The temporised running of either the EVs or the pump is driven by relays. At the time $t_0 = 12'$ the EVs 1 and 2 are opened and the diaphragm pump is turned ON. EV₁ allows the water flux through the sampling beaker of the system A up to the time $t_3 = 27'$: a complete renewal of the water analysed during the previous cycle of measurement occurs. Similarly EV₂ allows the external air, pushed by the diaphragm pump, to enter the cylinder so as to change the air related to the previous cycle of measurement.

Between t_0 and t_3 the following steps occur: at $t_1 = 19'20''$ EV₂ is closed allowing the diaphragm pump, running up to the time $t_2 = 20'40''$, to create a partial vacuum of 0.65 bars. At the time t_3 valve EV₁ is closed and at the same time EV₃ is opened, allowing the air re-entering within the cylinder from the environment through a needle

valve; the external air gurgles in the water contained in the beaker, stripping out the radon gas until the pressure equilibrium is reached. At the time $t_4 = 37'$, after around 25' since t_0 , valve EV_3 is closed and a stand-by step starts, lasting around three hours, before the counting step.

Another timer (timer 2) switches electric supply to the signal processing devices connected to the detectors. The same counting system for the two devices has been used: the two measurements, both from A and G systems, are not accomplished contemporary, but with an interval among them: during the first measuring period (1 h) a PC multi-channel card, used as *scaler*, meaning that it uses a unique ROI comprising the spectrum generated by the α -particles as a whole, allows to memorise the integral α counting; during the subsequent measurement period, the same card allows to record the γ spectrum, which is successively memorised together with the counts related to the selected ROI.

This temporal sequence, the storage of the data to submit to further statistical elaboration and radon concentration calculation are managed by a command file processed by the MCA card software, thus permitting the automatic execution of measurement surveys. A customised software, written in Power-Basic (*Power Basic*TM), fully dedicated to the designed system, is specifically addressed either to the writing of the command file or to the data elaboration; its main subroutines are shown in fig. 6.

3. – Results of comparison between the two systems

From May 1999 to January 2000 both systems have been functioning in parallel; however from December 1998 to March 1999 system G was already working. The radon concentration in the water drawn off the well resulted about 225 Bq/l on average, measured twice a month at the beginning of each cycle with methods described in [16, 17], also used for calibration purposes; the inferred average efficiencies are

$$\begin{aligned} \text{Eff}_G &= 3.22 \pm 0.20 \text{ cpm/Bq l}^{-1}, \\ \text{Eff}_A &= 7.14 \pm 0.14 \text{ cpm/Bq l}^{-1}. \end{aligned}$$

The background measurement of the two adopted systems (F_G and F_A) has been accomplished using “old water”, meaning water kept in a tank without connection with external air for a month period, allowing the dissolved radon to decay completely. In the case of the system A, the “old water” degassing was performed by “old air”, kept in a plastic flask for a monthly period.

The following values have been obtained during an hourly background counting (expressed as counts/hour = cph):

$$\begin{aligned} \text{System G} : F_G &= 14947 \pm 47 \text{ cph}, \\ \text{System A} : F_A &= 69 \pm 4 \text{ cph}, \end{aligned}$$

as a consequence, for the Lower Limit of Detection (LLD) and the Minimum Detectable Activity (MDA) defined as

$$\begin{aligned} \text{LLD} &= \frac{3\sqrt{F}}{t_{\text{acquisition}}} \text{ (cpm)}, \\ \text{MDA} &= \frac{\text{LLD}}{\text{Eff}} \text{ (Bq/l)}, \end{aligned}$$

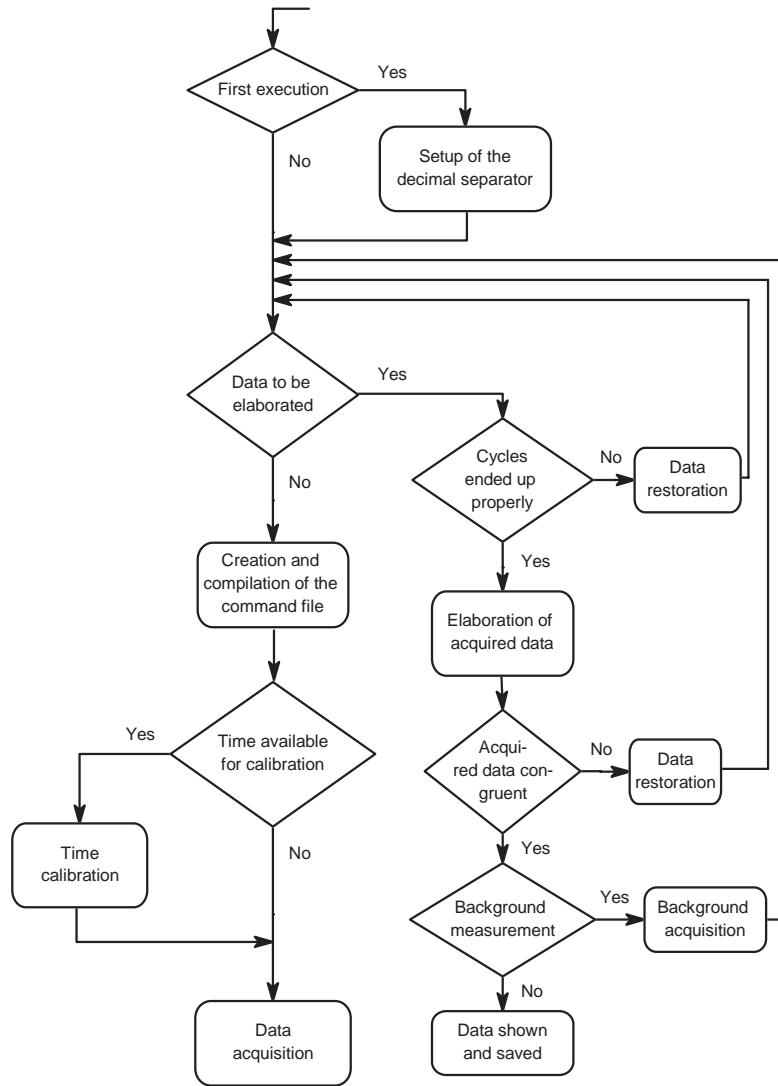


Fig. 6. – Block diagram of the software.

it follows that

$$\text{System G : } \begin{cases} \text{LLD}_G = \frac{3\sqrt{F_G}}{t_{\text{acquisition}}} \cong 6.11 \text{ cpm,} \\ \text{MDA}_G = \frac{\text{LLD}_G}{\text{Eff}_G} \cong 1.9 \text{ Bq/l,} \end{cases}$$

$$\text{System A : } \begin{cases} \text{LLD}_A = \frac{3\sqrt{F_A}}{t_{\text{acquisition}}} \cong 0.42 \text{ cpm,} \\ \text{MDA}_A = \frac{\text{LLD}_A}{\text{Eff}_A} \cong 0.06 \text{ Bq/l.} \end{cases}$$

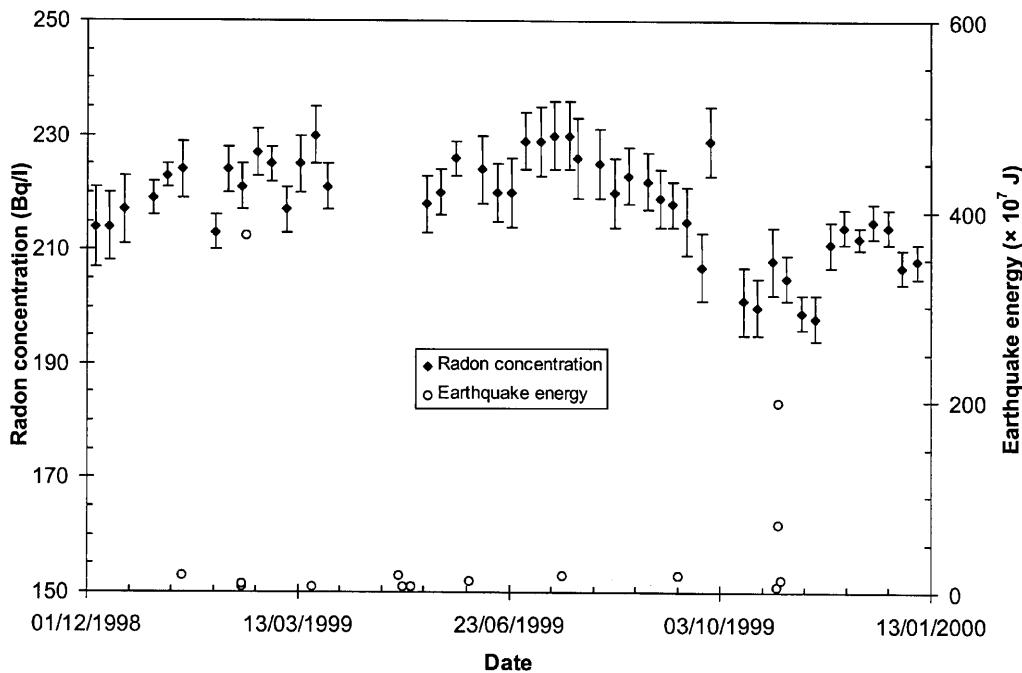


Fig. 7. – Plots of the weekly averaged radon concentration in groundwater obtained with system G (rhombs) and of the energy released by the main seismic events (circles) *vs.* time. Error bars are related to a confidence level of 95%.

Obviously the *scintillation chamber* response to degassed radon is concerned more than its absolute performance, therefore radon contained in the environmental (degassing) air, re-entered within the cylinder, must be considered as part of the *background*. For instance the *background* for system A would result in 560 cph if radon concentration in air were 200 Bq/m³, which corresponds to an MDA_A of 0.17 Bq/l and an LLD_A slightly greater than 1 cpm. This last datum must be compared with the 7 cpm for each Bq/L of radon in water, as the efficiency of the proposed system.

It is evident that the system A is characterised by a higher sensitivity. Anyway the sensitivity could not be the unique choice prerequisite: where radon concentration in water is high enough (*i.e.*, around > 5 Bq/L) it can be convenient the system G. This is due either as a consequence of the possibility to check the measures by the spectrum memorisation, or because the water circulates through a loop (solenoid-valves, tubes, BM), that could be made up of materials not able to be attached by corrosive gases dissolved in groundwater.

In fig. 7 the radon concentration in groundwater, weekly averaged, measured with the system G from December 1998 to January 2000 has been plot (rhombs) *versus* time together with the energy released by the main seismic events (circles) which took place within 30 km from the test-site during the same period. Because of the small earthquake sizes, they cannot be correlated with fluctuations in radon concentration [18,19].

A demonstration of the good agreement between the two systems is in fig. 8 where all data collected from 20/12/1999 to 10/01/2000 with system A (points) and system G (squares) have been reported.

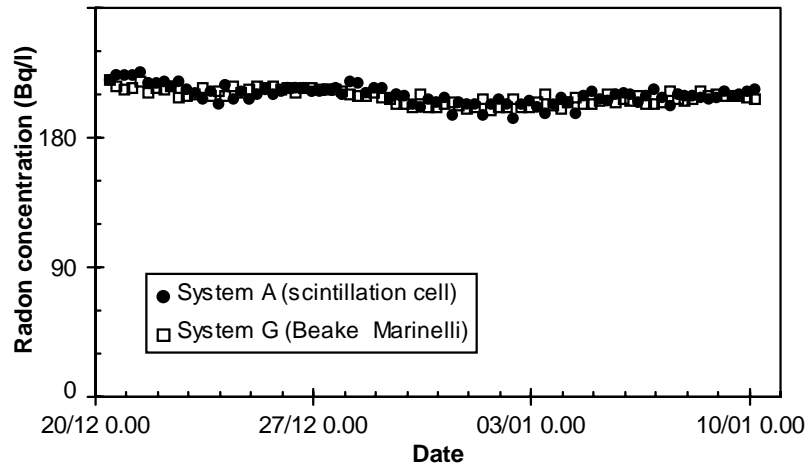


Fig. 8. – Plots of radon concentration in groundwater obtained with systems A (points) and G (squares) related to a typical measurement cycle.

4. – Conclusions

Radon continuous monitor prototypes realised by DINCE, adopting α -scintillation (system A) and γ -spectrometry (system G), arise from the critical overview of the state of art in this research field, from results gathered in the last decades and from researches addressed to earthquake prediction and seismic-volcanic surveillance, enhanced by ING. In the prototype development we have taken in consideration the present and future needs of the impending multi-parametric geochemical networks, addressed to Natural Hazards Assessment.

In particular the realised radon monitor prototypes were conceived as part of a multi-parametric *Geochemical Monitoring Station* (GMS II) prototype, designed and realised as *test-stand* for sensors (chemical, hydrological, geophysical, organic chemistry, etc.) in continuous evolution throughout the international market. This view is strongly suggested to approach the main pre-requisite of a remote station: the maximum sensor/instrument versatility despite the shared software and hardware for the network as a whole.

Low radon concentration, typical of the natural fluids to be monitored in seismically active sites, requires very high sensibility methods: for previous Authors the most sensitive method remains the α -particles determination, that could be accomplished either by the measurement of the counting rate or by the measurement of ionisation current.

We suggest the adoption of α -particles counting by *scintillation cell* in the case of low-medium groundwater radon concentration or for indoor/soil gas radon determination, whereas we prefer the use of the γ -spectrometry coupled with a *Marinelli Beaker* device in case of higher (> 10 Bq/L) radon concentration in groundwater, mostly in the presence of acidic and reducing gaseous input from depth as CO_2 and H_2S and rougher environmental conditions.

Moreover the possibility to memorise the γ -spectrum with the system G is a very powerful tool to check periodically or in real time the reliability of the response of the system.

We confirm that the choice of the structure configuration, *i.e.* the hydraulic scheme from groundwater source to the final output and the extraction method of gaseous phase from the liquid one, strongly affects the reliability of the radon monitor as a whole.

We preferred in any case a customised *temporised flushing device* to a *continuous flushing device*, with minimum rate of radon measure every three hours, to let the radioactive equilibrium between radon and its short-lived progeny be established, with the aim to obtain a true radon concentration measure.

The comparison between the two low-cost devices allowed to infer the respective calibration factors, the stability of the two systems and the expected reliability in different geochemical-geological situations.

Fairly similar systems to system A described in this paper will be installed in two sites in the context of a geochemical and hydrogeological monitoring project (MICRAT) of ARPA Umbria.

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