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A prototype radonmeter for seismic surveillance

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Abstract
A new ^{222}Rn monitoring prototype has been designed, assembled and tested at the Istituto Nazionale di Geofisica (ING) specifically addressed to seismic surveillance tasks, exploiting environmental monitoring, etc. It operates with an α scintillation technique (photomultiplier + *Lucas Cell*) coupled with a water input system, that lets continuous dehumidified gas flow, stripped from groundwater under monitoring. Several laboratory tests have been carried out to check the stability and versatility of the system; moreover statistical tests have been accomplished on several data sets obtained with an ^{241}Am radioactive standard source, to check stability of the photomultiplier. A customised *water flow system* has been developed to perform both the highest efficiency and lowest influence of external noise parameters. This new prototype is very cheap and will be integrated within the new multiparametric geochemical monitoring system GMS II, that is currently being developed at ING, specifically designed for geochemical surveillance of seismic events.

Key words *^{222}Rn continuous monitoring – geochemical earthquake prediction*

1. Introduction

The role of ^{222}Rn as a possible earthquake precursor was initially assumed and then attested world-wide from the end of sixties to date (Scholz *et al.*, 1973; Hauksson, 1981; Rikitake, 1987; Thomas, 1988; Wyss, 1991; Zhang *et al.*, 1994; Igarashi *et al.*, 1995).

Following the *Dilatancy Theory* formulation (Scholz *et al.*, 1973), several theoretical models have been developed to explain the occurrence of ^{222}Rn anomalies with earthquakes; however, up to now, none of them completely explain the occurrence of anomalies very far from the earthquake epicentral zone (Fleischer, 1981). Very important enhanced models (Anderson and Grew, 1977) supposed growth and propagation of microfractures at high distances, by mechanisms like «stress-corrosion of crack propagation», that assumes the rock cracking process occurs in different ways, both in dry and wet conditions, where fluids play a fundamental role. Further developments of the previous models tried to explain the ^{222}Rn anomalies in relation to seismic activity like the IRSA Model (Increased Reactive Surface Area) (Hauksson, 1981), or the TMAT (Thermo Molecolar Activation Theory) (Dubinchuk, 1991). Recently, the role of fluids in seismogenic processes as well as the reliability of the geochemical anomalies correlated with earthquakes has been enhanced due to some experimental evidence like earthquake *triggering* (Hills *et al.*, 1993; Linde *et al.*, 1994; Miller *et al.*, 1996; Nur and Walder, 1992; Fournier, 1991).

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This work is a part of a wide research project developed by ING since 1990 on geochemical earthquake precursors within seismic areas (Barbieri *et al.*, 1987). In particular, after the installation of the I prototype of geochemical monitoring system (GMS I) at Pozzo Barozze in Rocca di Papa (Rome) (Quattrocchi and Venanzi, 1989, Dall'Aglio *et al.*, 1990, 1991), potentialities, limits and possible improvements of these automatic systems have been pointed out (Quattrocchi *et al.*, 1992; Quattrocchi and Calcara, 1994; Calcara *et al.*, 1995a,b). The result of these studies is the improved design (Calcara *et al.*, 1995c) of a II prototype of geochemical monitoring system (GMS II), financed since April 1996 by 2 EC contracts (Geochemical Seismic Zonation, No. ENV4-CT96-0291 and Automatic Geochemical Monitoring of Volcanoes, ENV4-CT96-0289).

This radonmeter prototype is one of the GMS II sensors/instruments of the system, improved from the first version (Calcara and Quattrocchi, 1993).

The development of the current version of the radonmeter followed the guidelines of two prototypes built in Japan and in Austria respectively (Noguchi and Wakita, 1977; Friedmann and Honegger, 1978); in particular the Japanese prototype (developed by the Earthquake Chemistry Laboratory – University of Tokyo) developed a network, still on-line.

2. ^{222}Rn continuous monitoring techniques

^{222}Rn is a noble gas that belongs to the ^{238}U radioactive chain as an unstable isotope. It can be detected both through the measurement of the emission due to its own decay process, or through the measurement of the emission due to decay processes of its daughters. ^{222}Rn radioactive decay is of α type, so to make direct measurement, an α-particle detector is needed. This kind of emission is detected by α particle counting, or ionisation current. Other detection techniques use different radioactive emissions (β and γ) due to the decay of ^{214}Pb and ^{214}Bi. Both techniques, direct and indirect have advantages and disadvantages. Let us consider first direct methods: the α scintillation counting method by ZnS(Ag) cells (*Lucas Cells*) was the first developed (1951) and it is the most widely used.

The ionisation chamber method was developed by Friedmann and Honegger (1978) and it is based on the following principle: gas extracted by stripping from groundwater contains several atomic species including ^{222}Rn and its unstable daughters, like ^{218}Po, ^{214}Pb, ^{214}Bi. New atoms produced by radioactive decay are in an ionic state; if there is an electric field around them, an electric charge flux is settled, raising an ionisation current, proportional both to the number of ions, and to the numbers of ^{222}Rn atoms.

This method reveals a few important advantages, if compared to the *Lucas Cells* one: the most important is that it is possible to use larger gas volumes than the other method. On the other hand, the limit of this technique is the high minimum threshold of sensitivity, due to the strength of the electric field needed to move all the charges.

One of the newest methods for continuous monitoring of ^{222}Rn uses *solid state photodiodes*, exploiting the property of silica photodiodes (*i.e., s.s. photodiode* made by *Hamamatsu Photonics K.K.*) to reveal α particles. This kind of photodiode can be used for Rn monitoring both in gaseous phase and liquid phase, overcoming the problem of stripping Rn from water cutting a systematic error in the measuring process as a whole.

One of these systems was developed by Varhegyi *et al.* (1992), while another one, specifically addressed to the gaseous phase, was made recently in Japan by Igarashi *et al.* (1995) for continuous monitoring tasks: it recorded a ^{222}Rn anomaly just before the Kobe earthquake (17/1/1995, $M = 7.2$).

The liquid scintillation technique is used only for discrete samples of water.

Another method is based upon γ spectrometry, revealing γ rays, *i.e.*, due to ^{214}Pb and ^{214}Bi decay processes. It employs a NaI(Tl) sensor, detecting Rn content directly in water using a *Beaker Marinelli* (Belloni *et al.*, 1995), or Rn stripped from water and then adsorbed on active carbons (Mancini and Giannelli, 1995).

3. The GMS II ^{222}Rn continuous monitoring prototype

The requirements for sound continuous monitors of radon in groundwater are summarised as follows:

i) Techniques for low-level radiation measurement are required, because radon concentrations in groundwater are often in the order of $10\text{-}10^{-10}$ Ci/l.

ii) The configuration of both the detector and water drainage system is to be as simple as possible and should not be easily affected by chemical reactions due to groundwater flow.

iii) The entire measuring system must be stable for long periods and insensitive to variations of external meteorological conditions, such as temperature and humidity.

iv) Instrument assemblage should be relatively maintenance free and easy to operate because observation wells are usually located in remote areas; facilities at the stations are lower than in the laboratory.

Following these guidelines, we describe the assembled prototype: a customised *Lucas Cell* continuous monitoring device, specifically designed for ^{222}Rn groundwater monitoring. In the impending future an improved version of the described prototype will be ready which differs in the electronics and dehumidification process.

The current version of our system measures the amount of radon dissolved in groundwater, using a stripping method to extract gases continuously from water.

It can be divided into two main parts: the first has the function of extracting ^{222}Rn from water (by bubbling), and the second of measuring the α scintillation due to the decay of ^{222}Rn and its daughters inside the *Lucas Cell.* Figure 1a shows the block diagram of the device, and the assembling (fig. 1b). In the case of discrete measurements of Rn concentration, a mixing between a fixed amount of water as well as a fixed amount of air is necessary to accomplish an efficient radon extraction, while during continuous monitoring both groundwater and air are under flux. It must be kept constant using both flux rate controller and peristaltic pump, followed by glass devices to separate the two phases. Through this method, most of the ^{222}Rn dissolved in water could be extracted. *Stripping efficiency* (%) is a decisive factor to improve the sensitivity of the system as a whole: it is clear that the lower the stripping efficiency, the lower the amount of particles, the α-detector can reveal. It is possible to increase the yield by regulating stripping efficiency. Friedmann and Honegger (1978) showed that by decreasing the ratio between air and water volumes (with respect to an optimised value – around 4 – used in the discrete mode; C. Mancini, personal communication, 1996), a higher concentration of ^{222}Rn in a smaller volume of air is obtained. Thus, with a lower stripping efficiency an increase in sensitivity of the measurement device is achieved.

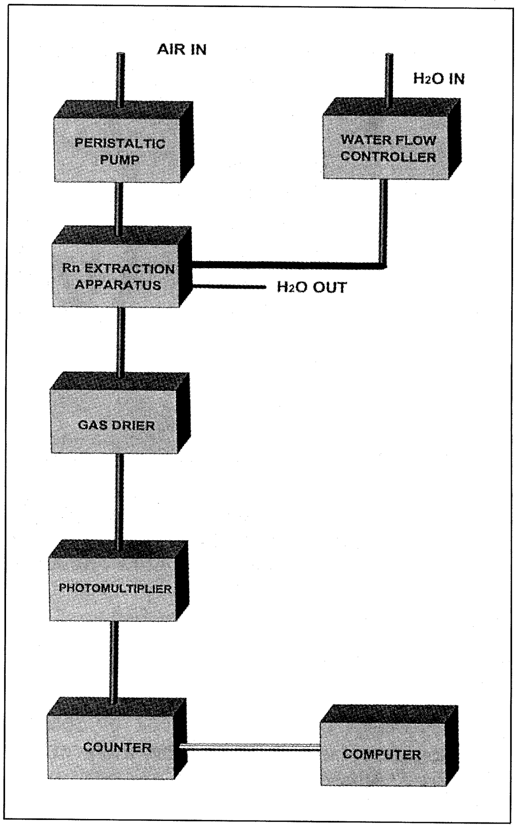

Fig. 1a. Block diagram of the ING radonmeter prototype.

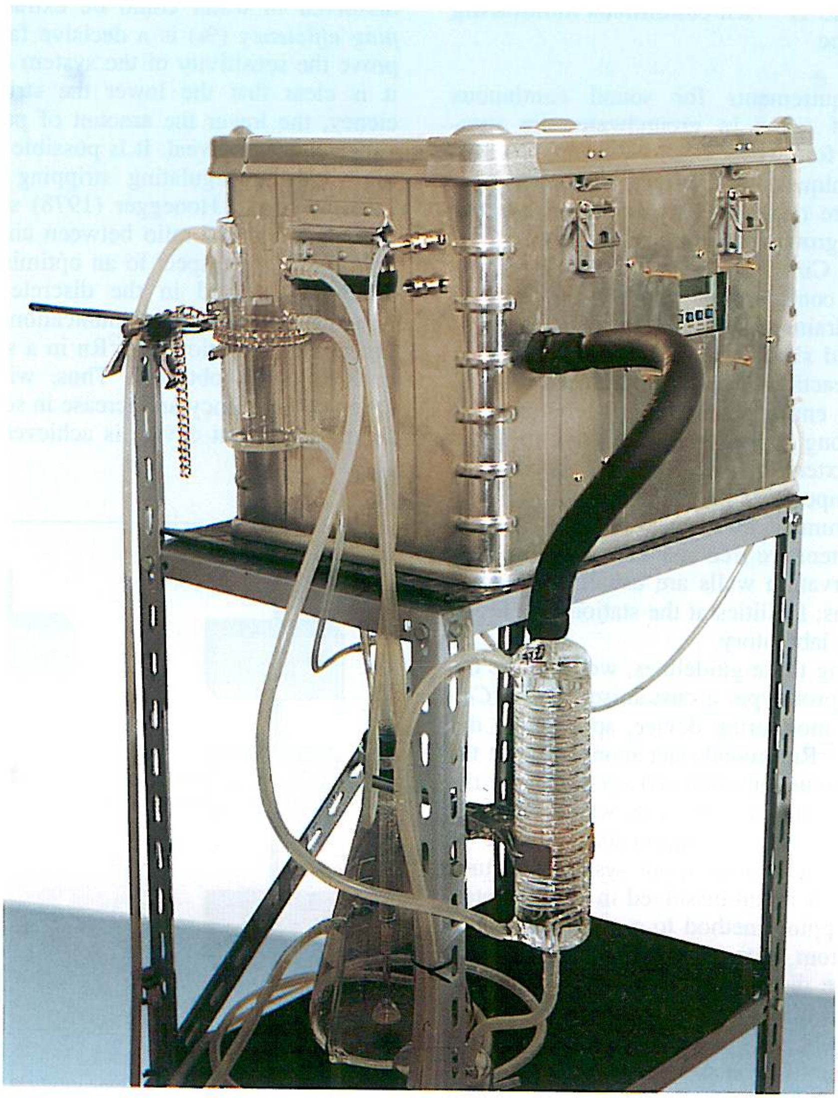

Fig. 1b. Photograph of the final assembly of the ING radonmeter prototype.

The needed optimisation must be reached by the correct balance of the two effects caused by reducing air volume for gas extraction: the smaller gas extraction rate and, on the other hand, the higher concentration of Rn in the same volume of air. After testing, we selected a sound ratio between air and water volumes, that is 1/2 ($V_{air}/V_{H2O} = 0.5$); flow rates of air and water has been fixed at 0.475 cm^3/s and 0.95 cm^3/s respectively.

The second problem to be considered is the long term stability of the stripping apparatus. Since the extraction method is based on the *Henry Law*, a very small variability of temperature is needed to have a constant distribution coefficient K_a (defined as the ratio between radon concentrations in gaseous and water phases).

Considering both the problems we designed the final configuration of a customised *water flow system*, as described below.

A very simple and cheap device for water flow input constancy improvement has been made: a small glass basin with a input water hole shaped to prevent any gas anomalous bubbling from the well and an output hole for discharging water excess (fig. 2). Making reference to the *Bernoulli Law*, whose formula is

$$\delta/\delta s(Z + P/\gamma + v^2/2g) + dv/dt^* \, 1/g = 0$$

the water level in the basin – considering our specific flow conditions – was calculated to be 12.4 cm.

The second glass container (fig. 3) is the device where stripping of radon from groundwater occurs. The size of this bottle is such that the cycle of charge-discharge of water takes approximately the same time as the integration time of α scintillation counting.

Inside the bottle, air/water mixing process occurs below the water surface, since air tube ends at a fixed depth with a removable *porous septum* that allows the mixing process. Both glass basin-stripping bottle assemblage and the variable-rate peristaltic pump assure a very high stability of air and water flows, suggesting that only changes in temperature can be sources of error; to easily correct the time series, the GMS II prototype includes continuous monitoring of water temperature.

The radon extraction system is linked with the α scintillation cell (EDA Instruments, 125 cc volume), coupled with the photomultiplier, passing through a *drier apparatus* (fig. 4), that causes vapour condensation, just before gas flow reaches the *Lucas Cell.* It could be damaged if water wets the ZnS(Ag) film. Condensation is caused by heating and then, cooling the gas flow by two serial coils: the first coil heats, due to the location within the pump motor case and the second coated coil cools by water flux itself. The output of the last coil is connected with a container that collects water due to the condensation. We customised this «condensation system», considering the peculiar conditions of our groundwater under monitoring (around 5-10 °C). Otherwise if the water temperature is higher than 5-10 °C, the system should not cause steam condensation and thus

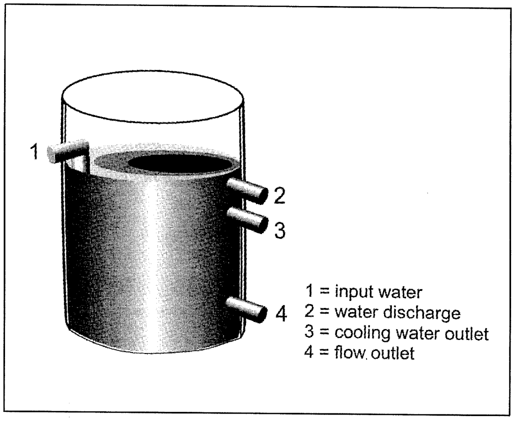

Fig. 2. Water flow controller glass basin.

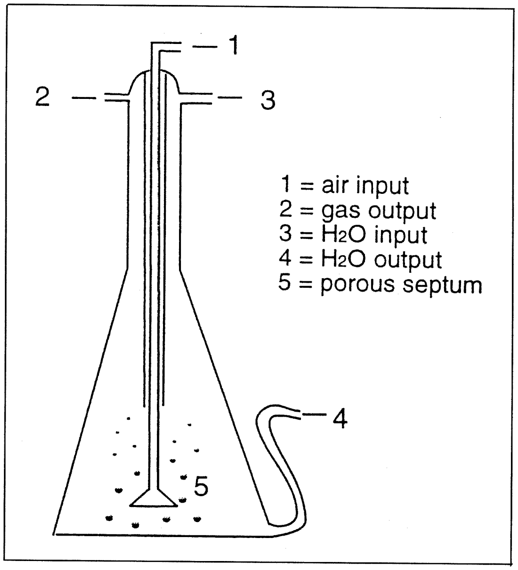

Fig. 3. The Rn extraction glass device.

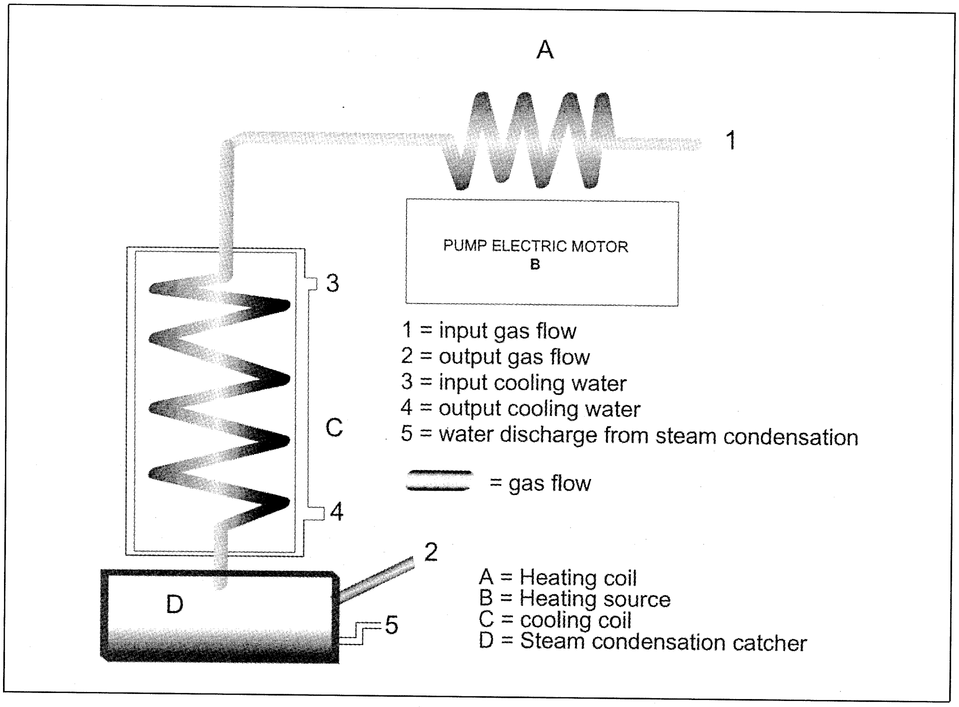

Fig. 4. The drier apparatus.

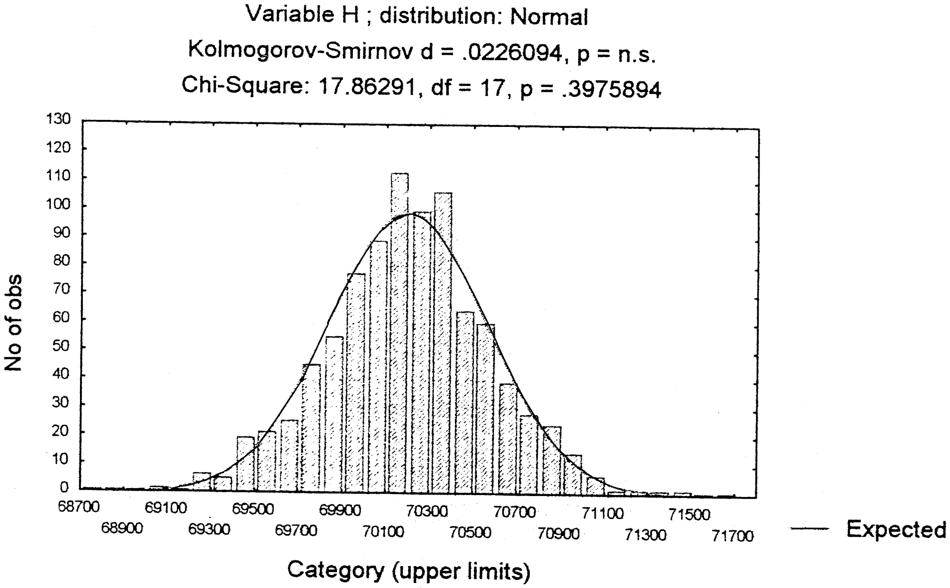

Fig. 5. Histogram of the data set gathered with the ^{241}Am standard cell.

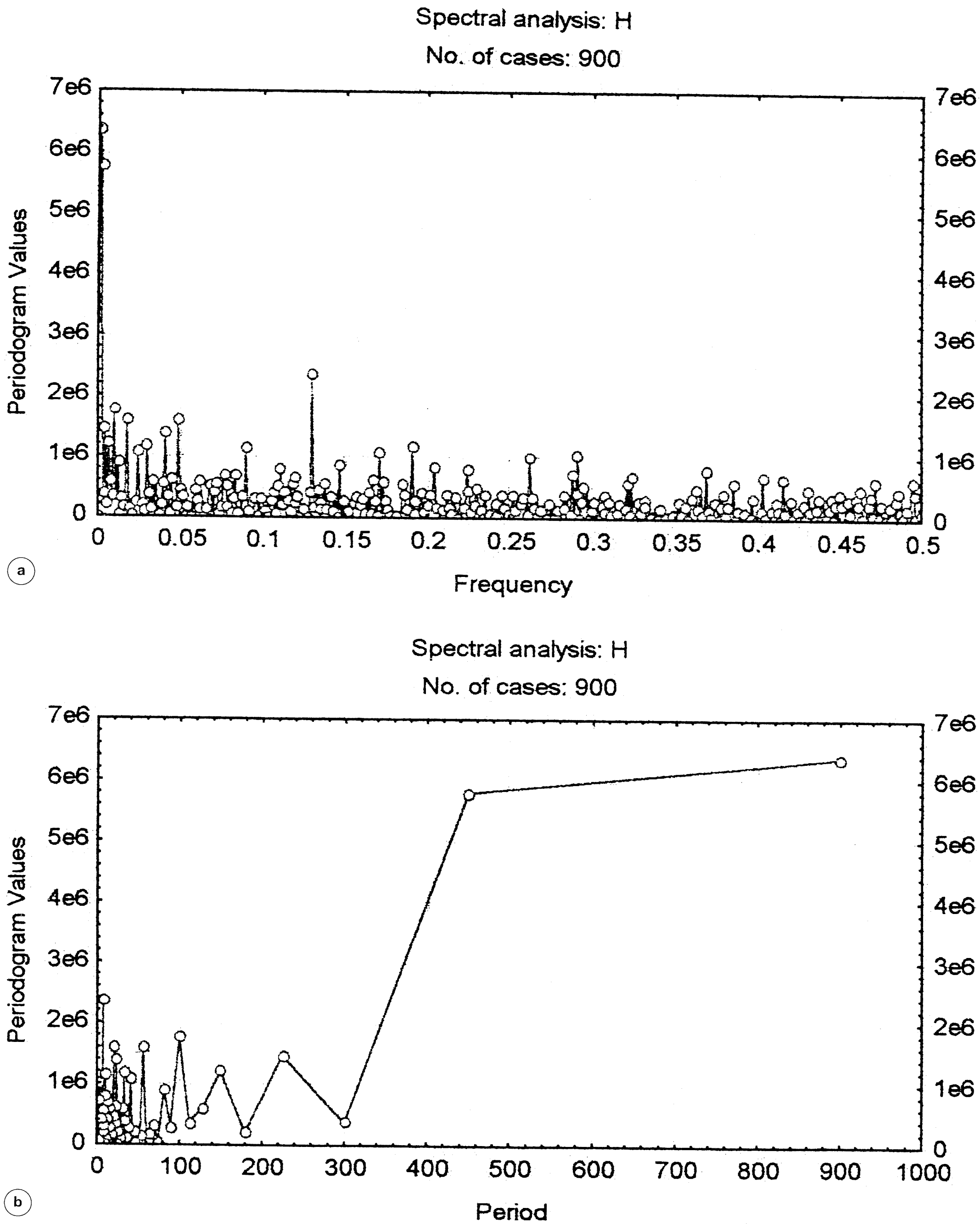

Fig. 6a,b. Spectral analysis of the data set gathered with the ^{241}Am standard cell: a) frequency domain; b) period domain.

another gas drier device must be chosen (Permapure^R exchanger, Peltier cells, silica gel, etc.).

The electronic equipment of this prototype uses a few components with technical specification for long-term performance, like low electrical input, and relatively simple and cheap hardware; the data acquisition device is a GPC 1 made by TESYS-Technology & System S.a.S (Milan, Italy). It is an electronic device that includes a High Voltage device, Preamplifier, Amplifier, Discriminator and Counting Rate meter sections. High voltage regulation has a wide range (500-2500 V) with 10 V steps; the amplifier has a gain control and *background* threshold; the discriminator can easily change the threshold value, allowing high stability in voltage.

Integration time of measurements can be changed from 1 s to 99 h and the memory capacity is wide up to 1000 data recorded. The GPC 1 output unit will be improved for GMS II monitoring specific purposes.

Photomultiplier calibration tests were accomplished by a ^{241}Am standard cell (1360 ± 40 decays per minute), to check signal constancy and system efficiency. The efficiency of electronic equipment is 86%. Photomultiplier-counter *background* was evaluated by an empty *Lucas Cell* (EDA Instruments, 125 cc volume); at the same time the statistical distribution of results and temperature drift was checked by the ^{241}Am standard (fig. 5). Statistical analyses show that the obtained data set is a sample that belongs to a normal distribution, with an error 5% lower. Spectral analysis (fig. 6a,b) excludes – considering the available data up to date – any cycle in the electronic equipment running, within the photomultiplier coupled with other components.

The last test, performed to check the influence of changes in external temperature, showed that there is no appreciable link between this parameter and the photomultiplier response (figs. 7a-c), remaining negligible if compared with changes due to Rn solubility in water with changes in temperature.

The ^{222}Rn continuous monitoring system was tested in laboratory too, using water flow from the public water supply (ACEA Rome).

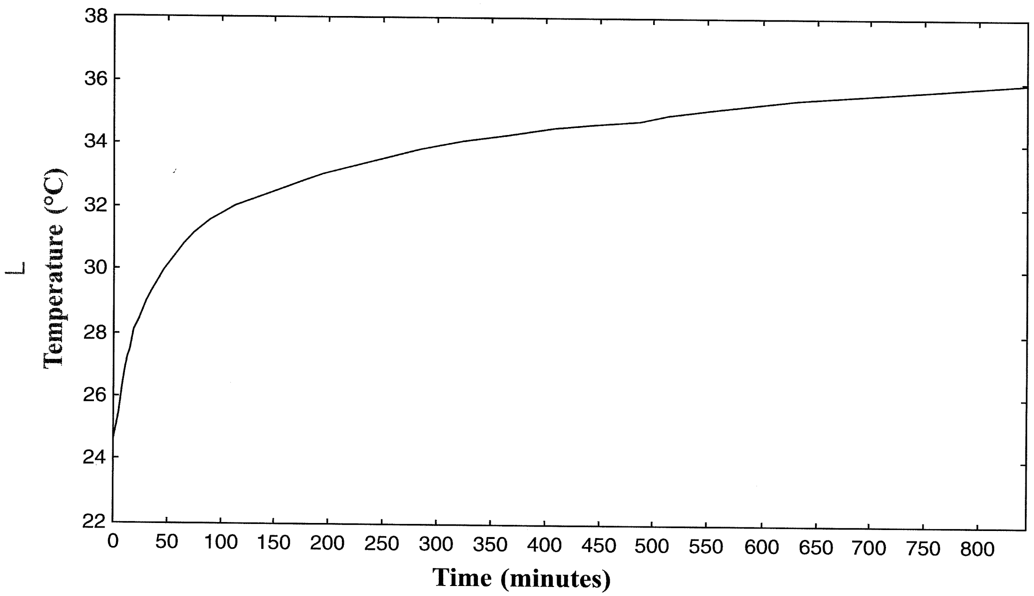

Fig. 7a. Diagram of temperature *versus* time inside the radonmeter device.

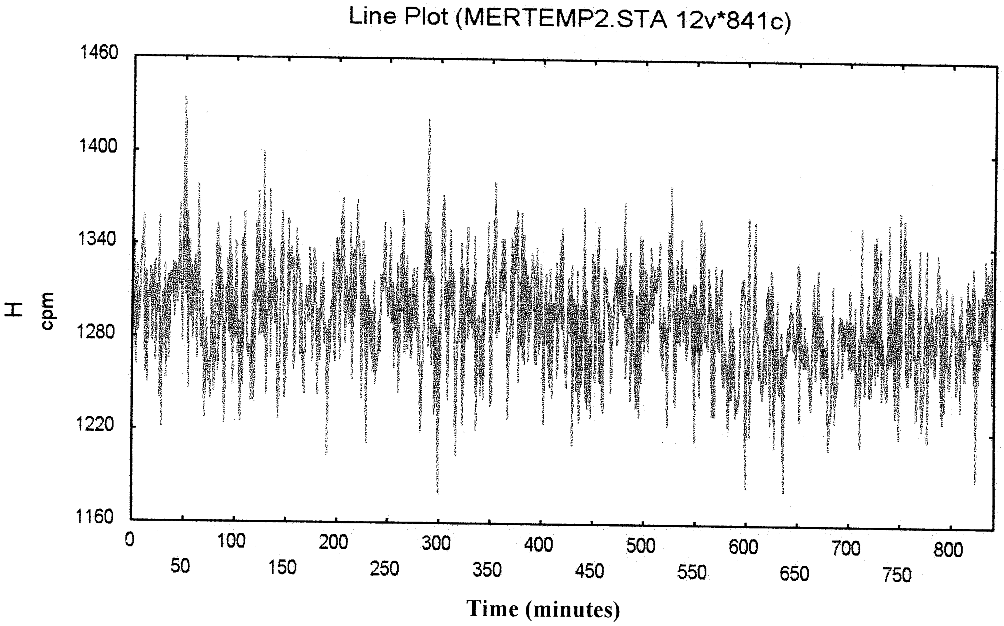

Fig. 7b. Counts per minuted data set gathered with the ^{241}Am standard cell, with variable temperature.

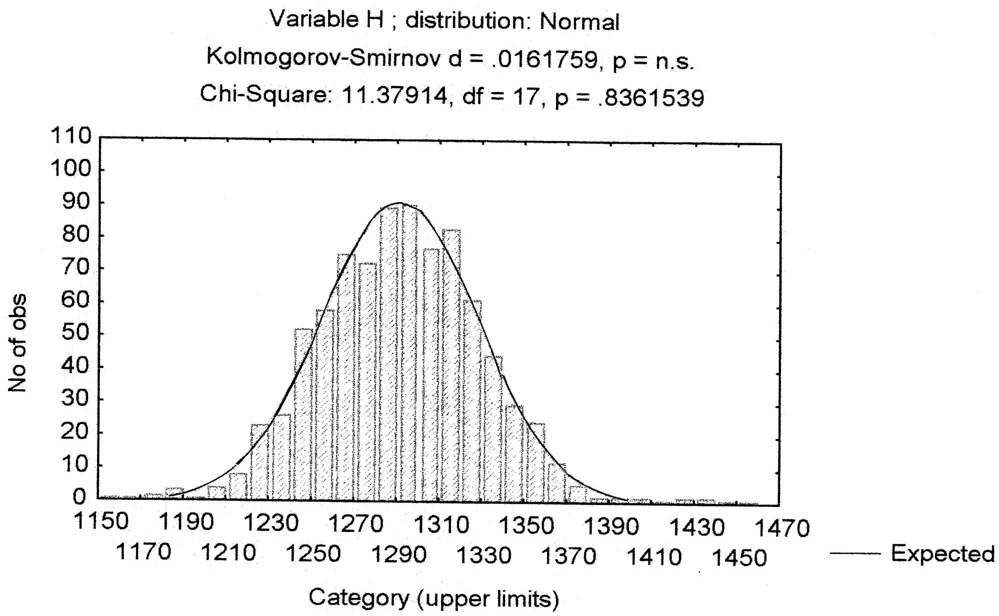

Fig. 7c. Histogram of the data set gathered with the ^{241}Am standard cell, with variable temperature.

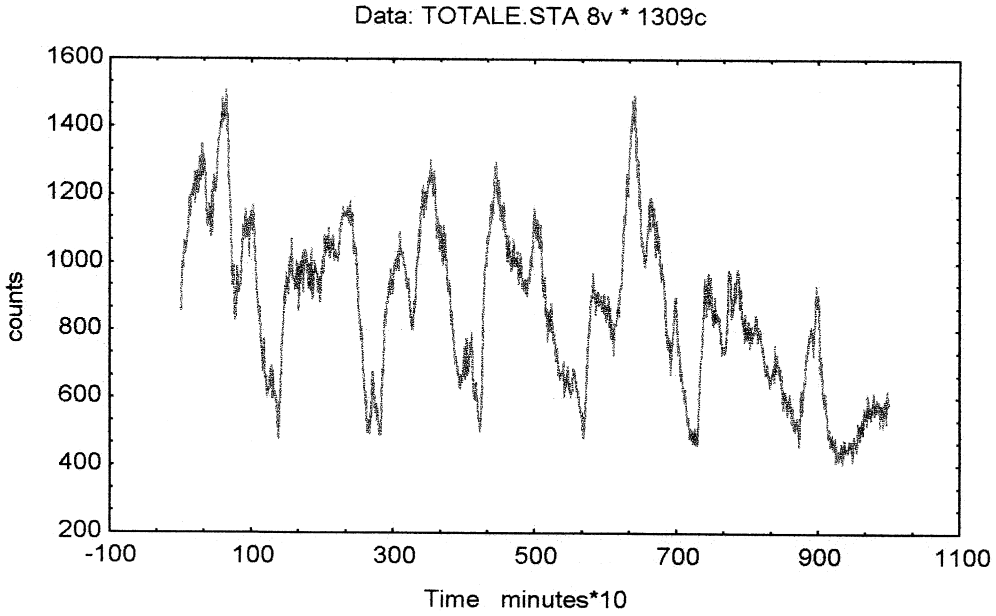

Fig. 8. Counting data set gathered with the ACEA public water supply.

Rn content in this kind of water is very low (about 100 pCi/l). The purpose of this test was to check the stability of the *water flow system*, drier apparatus and electronic equipment. Integration time of measurement during the test was 10 min.

About 45 min after starting the experiment, the whole drainage system ran in a steady condition (temperature stop growing within the first coil in drier apparatus) and steam in gas flow started to condense. A hygrometer must be connected to the gas circuit to check final relative humidity.

A water flow controller system neutralised small changes in the water flow rate, confirming the soundness of this size of the basin for the current configuration working range, ranging from 1.5 to 4.0 l/min; it can be easily improved for each remote site: currently it is online at *Arta Terme* – the Friuli site, before the final installation at the monitoring station of Tivoli *Acque Albule* site (Quattrocchi *et al.*, 1997).

Data recorded using the public water supply (ACEA Rome) showed two main cycles in the time series, pointed out by spectral analysis (figs. 8 and 9a,b): periods of these cycles are 12 and 24 h, showing no correlation with the results of spectral analysis gathered using the ^{241}Am standard cell. Thus, the two observed spectral peaks are due to changes in temperature and Rn content in the ACEA water, as well as to unknown processes intrinsic in the water flux but external to the apparatus, such as running pumps, degassing, filtering etc.

4. Conclusions

A prototype system for continuous monitoring of ^{222}Rn in water (and gas too) has been developed as part of instruments/sensors of the II prototype of geochemical monitoring system (GMS II). The radonmeter has been designed, developed and tested for seismic and environ-

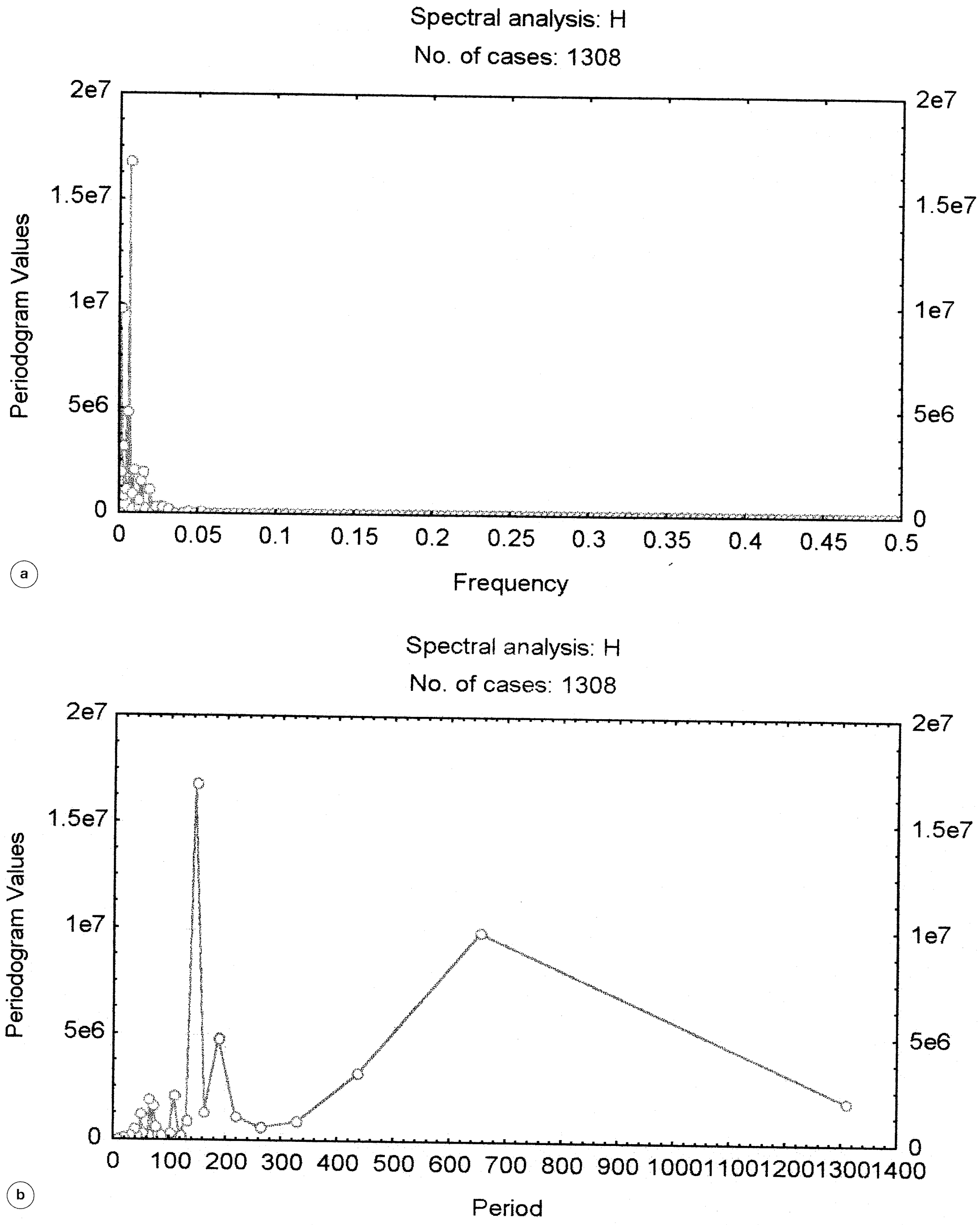

Fig. 9a,b. Spectral analysis of the data set gathered with the ACEA public water supply: a) frequency domain; b) period domain.

mental surveillance purposes. It operates with the α-scintillation technique, coupling *Lucas Cell*-photomultiplier assemblage to a customised *water flow system.* The latest allows a continuous and constant flow of dried gas stripped from water under monitoring. This device can operate continuous measurements (acquisition time is changeable from 1 min to 24 h of very low ^{222}Rn content (about 100 pCi/l), as first tests show. Photomultiplier and electronic equipment stability has been tested; the drainage system has been soundly customised. Stability tests of the drainage system and statistical tests on data sets obtained using both an ^{241}Am radioactive standard source and the ACEA-Rome water supply, are encouraging for long term monitoring use.

The new prototype is very cheap and versatile for continuous field use and it will be included in the GMS II first monitoring station at the Tivoli *Acque Albule* site.

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