

## Automated measurement of radon daughters Bi-214 and Pb-214 in rainwater

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**Abstract** Since March 1994, the Institut de Tècniques Energètiques (INTE) of the Universitat Politècnica de Catalunya (UPC), has been measuring environmental gamma dose rate and the airborne radon concentration levels outdoors, at the ESCRA Station, which is located in Barcelona. ESCRA simultaneously measures several meteorological parameters as well. As it is well known, the gamma dose rate suffers noticeable increments during rain intervals, which in our case range from 10% up to 40%, mainly caused by the deposition of the radon daughters  $^{214}\text{Bi}$  and  $^{214}\text{Pb}$ . These increments do not seem to be correlated with rainrate, rain interval duration, precipitation volume, or other meteorological parameters. In order to develop models that explain the observed dose enhancement, it is interesting to determine the concentration of the gamma emitters in rainwater and, due to the relative short half life of the two nuclides mentioned above, this must be done as it rains. To this end, we have developed an on-line gamma spectroscopy system for rainwater. The operation of this device, which has been automated by means of a LabView program, consists of the following steps: first, 250 cm<sup>3</sup> of water is collected and, if the rainrate is high enough, is transferred to the measurement tank, a Marinelly-like container located underneath a HPGe solid state detector. All the system is surrounded by a lead shield. Preliminary measurements carried out with this device yielded radon daughter concentrations ranging from 50 Bq/l up to 1600 Bq/l.

**Key words** Bi-214 • Pb-214 • radon • rainwater

### Introduction

It is well known that the environmental gamma dose rate varies with time. Atmospheric pressure, airborne radon concentration, precipitation and snowfall are some of the natural contributors to the temporal variation of the environmental dose rate [2]. During time intervals without precipitation or snowfall, the dependence of environmental gamma dose rate on atmospheric pressure and airborne radon concentration can be employed to know if the dose rate changes are caused by natural or artificial sources. When it is raining, the dose rate may increase considerably due to the presence of radon daughters in rainwater [5], an effect that can mask the presence of artificial gamma emitters. In order to estimate these increments it is necessary to determine the radon daughter concentration in rainwater. Some authors (for example [4] and [1]) have shown some results from indirect measurements.

For over six years, the Institut de Tècniques Energètiques (INTE) of the Universitat Politècnica de Catalunya (UPC) has been measuring environmental gamma dose rate and the outdoors airborne radon concentration levels at the ESCRA station [7], located in Barcelona (Spain). The analysis of the data recorded at the ESCRA showed the increments of the dose rate during rainfall [3] mentioned above. In order to study this phenomenon, an automated device for the measurement of the concentration of gamma emitters in rainwater, which we call SARAC

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(Sistema de Análisis de la Radiactividad del Agua de lluvia en Continuo, on-line measurement of radioactivity in rain-water), was designed and developed.

Other authors, [6] and [8], have developed similar devices for the measurement of radon progeny in rainwater an NaI scintillation detector for gamma spectroscopy. The improvement on the device presented in this work consists in the employment of a High Purity Germanium detector for gamma spectroscopy, which gives better accuracy in the measurements than the other ones.

In this report, we show the components of SARAC, its operation and some results obtained so far.

## Methods

In Fig. 1 a schematic diagram of SARAC is shown. Its components are described below:

- Rainwater collector: A 2 m<sup>2</sup> stainless steel funnel, employed to collect the rainwater. It is located on the roof of the ESCRA Station, 1.5 m from the accumulation container.
- Accumulation container: It consists of a PVC cylinder with a volume of 0.32 litres. It accumulates the collected water before it is transferred in to the next element.
- Counting container: This is a PVC Marinelli-like container that stores the water (transferred from the previous element) during the acquisition of the spectrum by an HPGe detector. Its design was optimised using Monte Carlo simulation of the system.
- Gamma spectroscopy system: The components of this system are:
  - 1) A CANBERRA High Purity Germanium Detector (HPGe) cooled by liquid nitrogen with a thin beryllium window (0.5 mm-thick) at the end cap of the detector. The germanium crystal is 55.5 mm high and has a diameter of 50.5 mm.
  - 2) An EG&G Ortec multichannel analyser (MCA), model DART.
  - 3) The software MAESTRO version 3.2 for Windows95, developed by EG&G Ortec, to control the DART and to acquire and analyse the gamma spectrum.
- Storage tank: Its volume is 80 litres and stores all rain-water collected.
- Shielding: It surrounds the counting container and the end cap of the detector. Its thickness is 5 cm and is made of lead bricks. This shielding is used to reduce the background level.
- Level sensors (S1, S2, S3, S4) and magnetic valves (V1,V2,V3,V4).
- Control system: All the electronic components of the SARAC are controlled by a computer with the help of a program developed under LabVIEW® 5.1. This program controls the magnetic valves and the operation of the MCA using the information delivered by the level sensors. The communication between the sensors, magnetic valves, and the computer is performed by a LabPC 1200 data acquisition interface of National Instruments.

The operations carried out for the measurement of a sample of water are the following:

- a) Rainwater is collected and led to the accumulation con-

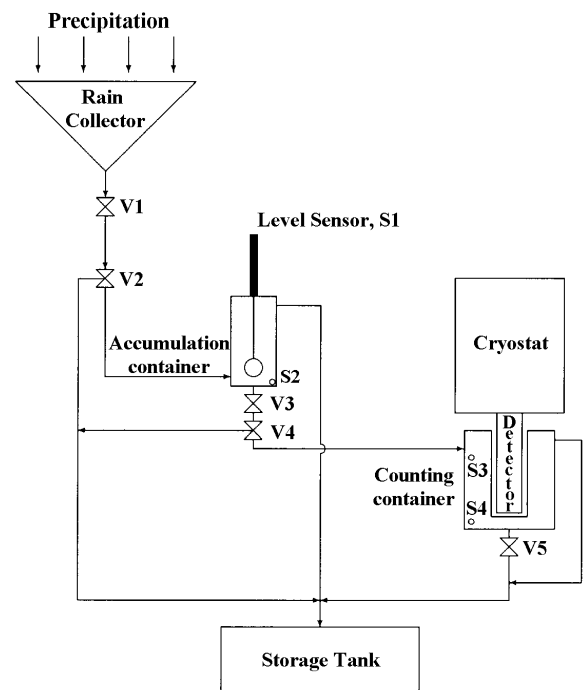


Fig. 1. Schematic diagram of the SARAC device.

tainer. When its volume reaches 250 cm<sup>3</sup>, it is transferred in to the counting container, provided that the sampling time has not exceeded 600 s, otherwise, the water is rejected and transferred in to the storage tank;

- b) The gamma spectroscopy system counts for 600 s. The spectrum is stored on the computer hard disk;
- c) At the end of the counting process, the water is transferred in to the storage tank and the MAESTRO software starts the analysis of the spectrum and writes a report to the disk with the radionuclides identified in the sample. The cycle starts over again once the measurement container has been emptied.

An algorithm to correct for the decay of the short half-life radionuclides detected has also been implemented, which takes into account the sampling and counting times. The final result is thus given in Bq per litre of rainwater.

To perform the efficiency calibration of the system, a cocktail of radionuclides in a water solution, with energies ranging from 88 keV to 1836 keV, was used. The low level detection limit (LLD) was found to be 9 Bq/l  $\pm$  1 Bq/l for energies between 351 keV and 1260 keV.

## Results

SARAC has been in operation since December 1998. In this section the concentration of the radon daughters <sup>214</sup>Pb and <sup>214</sup>Bi in rainwater during 1999 are presented. During this year only 15 episodes of rain were registered.

A preliminary analysis of the data obtained so far indicates that the concentration of radon daughters varies considerably between rain episodes. As shown in Table 1, the concentration of <sup>214</sup>Pb ranges from 48  $\pm$  12 Bq/l to 435  $\pm$  35 Bq/l, and that of <sup>214</sup>Bi from 94  $\pm$  26 Bq/l to 1561  $\pm$  171 Bq/l. This

**Table 1.** Concentration of radon daughters in rainwater measured in Barcelona during 1999.

Rain episode	Date Year 1999 (dd-mm)	Duration (minutes)	Concentration $^{214}\text{Pb}$ (Bq/l)			Concentration $^{214}\text{Bi}$ (Bq/l)		
			Min.	Max.	Mean	Min.	Max.	Mean
1	01-01	454	129±18	290±34	194±24	351±52	915±99	761±88
2	13-03	76	284±32	435±45	306±34	871±97	1270±143	856±99
3	24-03	156	30±11	74±16	51±13	181±37	309±46	220±40
4	25-03	61	27±12	41±16	34±14	246±37	263±48	350±51
5	26-03	20	51±17	85±21	68±19	246±46	344±59	295±52
6	27-03	68	54±15	153±28	96±20	247±48	357±72	285±55
7	15-04	136	48±12	203±24	111±17	199±40	594±74	406±54
8	16-04	31	106±21	205±28	148±24	441±66	549±83	481±71
9	17-04	35	99±18	212±30	154±25	435±61	537±79	477±70
10	28-04	253	60±14	414±45	231±30	327±44	1561±171	903±106
11	13-09	20	59±11	76±13	68±13	247±33	317±41	282±37
12	14-09	126	78±13	174±25	110±17	279±36	839±97	519±63
13	14-09	42	115±18	320±37	192±25	287±39	1547±167	806±94
14	18-09	60	249±29	335±37	282±32	496±60	933±105	725±85
15	20-10	388	21±9	168±22	56±12	89±17	542±67	252±37

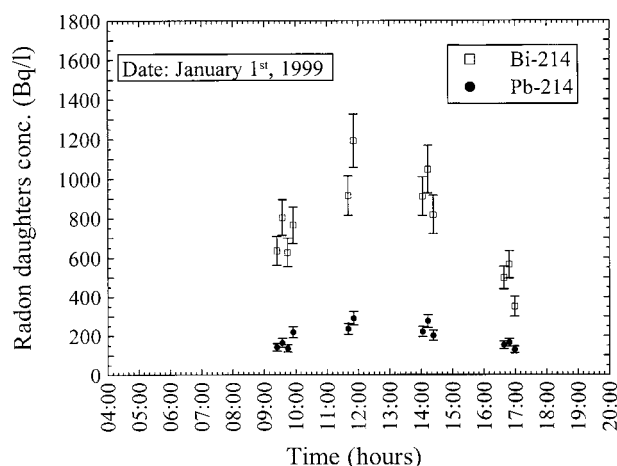
variation can be explained if we consider the path of the clouds and the mechanism involved in the cloud formation. When the storm comes from the sea, the radon and radon daughter concentration in the cloud is less than when the storm comes from the land.

Significant variations of the concentration with time in a single rain episode are also common (see Fig. 2), although in some other cases the measured activity is fairly constant (see Fig. 3). There are several phenomena involved in these variations, and we are still analysing the meteorological data recorded to try to explain it. The rain episodes in Barcelona are very scarce, so we need more data to extract reasonable conclusions.

The disequilibrium observed on the activity of Pb-214 and Bi-214 is due to the time elapsed between the rain drop formation on the cloud and its arrival to the soil. The activity of Bi-214 is always greater than that of Pb-214 because the first radionuclide is the daughter of the second one.

## Conclusions

An automated device for the on-line measurement of the



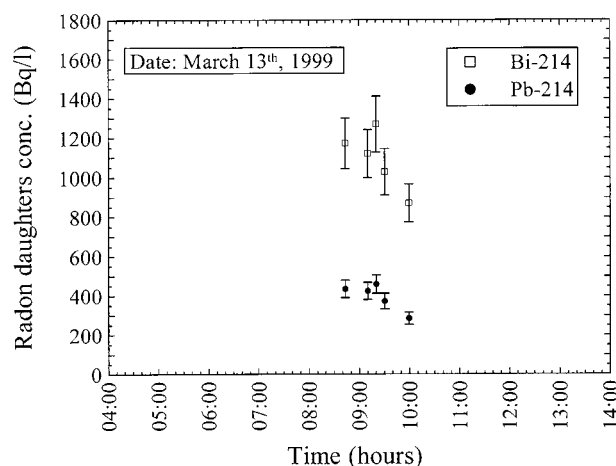
**Fig. 2.** Concentration of the radon daughters Bi-214 and Pb-214 in rainwater measured with the SARAC device in Barcelona on January 1, 1999 from 09:24 h to 16:58 h.

concentration of gamma-ray nuclides in rainwater, named SARAC, has been developed. The system capable of performing a measurement every 10 minutes, is located in the ESCRA station, in Barcelona (Spain), near the Mediterranean coast and has been in operation since December 1998.

Despite the small amount of data recorded due to the scarcity of rain episodes in Barcelona during 1999, considerable temporal variations of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  were observed. They can be explained due to the air masses origin, but is required more measurements to explain the variation of radon daughters during a single rain episode.

Work is in progress to explain gamma dose rate variations in terms of rain rate and radon daughter concentration in the water. The constructed device is also an efficient surveillance instrument capable of detecting small quantities of artificial radionuclides in rainfall.

We are presently planning to improve the performance of SARAC by substituting the currently employed High Purity Ge (HPGe) detector – cooled by liquid nitrogen – by an autonomous HPGe cooled by means of a cryoelectric device.



**Fig. 3.** Concentration of the radon daughters Bi-214 and Pb-214 in rainwater measured with the SARAC device in Barcelona on March 13, 1999 from 08:43 h to 09:59 h.

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