Applied Radiation and Isotopes 87 (2014) 57–60

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Calibration of an air monitor prototype for a radiation surveillance network based on gamma spectrometry

A. Baeza, J.M. Caballero, J.Á. Corbacho, M.Á. Ontalba-Salamanca *, J. Vasco

LARUEX, Department of Applied Physics, University of Extremadura, Avda. Universidad s/n, 10071 Cáceres, Spain

HIGHLIGHTS

- Deficiencies of a commercial air monitoring system are detailed.
- Gamma spectrometry introduction is the basis of the new prototype.
- Efficiency calibration procedure is described for aerosol and gaseous fractions.
- MDA is evaluated for different isotopes and acquisition times.

ARTICLE INFO

Available online 1 December 2013

Keywords:
Radiation surveillance network
Gamma-ray spectrometry
Aerosols
I-131

ABSTRACT

The objective of this work is to present the improvements that have been made in quasi-real-time air radioactivity concentration monitors which were initially based on overall activity determinations, by incorporating gamma spectrometry into the current prototype. To this end it was necessary to develop a careful efficiency calibration procedure for both the particulate and the gaseous fractions of the air being sampled. The work also reports the values of the minimum detectable activity calculated for different isotopes and acquisition times.

1. Introduction

The Extremadura Autonomous Community (SW Spain) has an automatic alert system in quasi-real-time denominated RARE. It began operating in 1992 in the vicinity of the Almaraz Nuclear Power Plant (Baeza et al., 1993). Today, it comprises 10 atmospheric and 2 water permanent radiation monitors plus 1 mobile laboratory (Baeza et al., 2013a). Currently, the 10 RARE atmospheric stations are equipped with Geiger–Müller counters which register the environmental gamma dose rates levels. Four of them have a commercial system that provides the radon decay chain activity, the total alpha and total beta activities of artificial emitters and the activity of the gaseous fraction of I-131. Despite their proven capabilities, these systems can today be considered technologically obsolete due to (i) it is impossible to individually identify and quantify the radionuclides in the aerosols; (ii) gaseous I-131 activity is calculated indirectly; (iii) radiological anomalies cannot be isolated for subsequent analysis in a low-activity laboratory; and (iv) the carbon cartridge used for I-131 collection must be changed manually.

In this work therefore, we shall first describe in greater detail the aforementioned analytical and maintenance shortcomings of the commercial systems currently in operation. We shall then give a brief description of the pilot monitoring station that has been designed, consisting essentially of the replacement of the current detection systems with one based on gamma spectrometry, but also including a number of mechanical modifications and new data acquisition and control software. The changes made to the two analytical modules require the spectrometers to be appropriately efficiency calibrated for the very singular geometry involved in the air measurements. Indeed, the principal objective of the present work is to describe the experimental procedures followed for this calibration and to determine the minimum detectable activity (MDA) values for different anthropogenic radioisotopes that are characteristic of the risk of radioactive contamination. These values allow one to assess the system's capacity to perform analytical radioactive determinations in quasi-real-time.

2. Materials and methods

2.1. The RARE's current atmospheric monitoring stations

Currently, the RARE's aerosol monitoring stations use the BAI-9850-6 (Berthold Technologies, 1997) system. This has two modules to capture and measure atmospheric samples:

2.1.1. Aerosol monitor

A suction pump (flow rate of about 22 $m^3 h^{-1}$) allows the aerosols to be accumulated on a glass-fibre filter which moves continuously under a double scintillation detector, ZnS(Ag)+ plastic, with its electronics. The system provides the total alpha

* Corresponding author. Tel./fax: +34 927257153.
E-mail address: ontalba@unex.es (M.Á. Ontalba-Salamanca).

0969-8043/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.apradiso.2013.11.095

and beta activities of man-made origin, and the radon activity concentration in the particles retained on the filter. These estimates are made on the basis of a temporal pseudo-coincidence circuit: when an alpha particle and a beta particle are detected simultaneously ($\Delta t < 163\ \mu s$), it discards these counts under the assumption that they come from the decays of the natural Pb-214 and Bi-214 isotopes.

The results do not allow the natural and/or artificial radioisotopes captured on the filter to be individually identified and quantified. Also, examination of the provided values leads one to question the reliability of the pseudo-coincidence circuit's operation. Fig. 1 shows the evolution of the total alpha and beta activities of artificial origin, as well as that of radon, for two of the RARE's stations (Saucedilla and Serrejón) during a single day of measurement.

In the case of the Saucedilla station (Fig. 1a), one can see that, as the radon activity concentration increases notably (in the early hours of the day), there is also an increase in the total alpha and beta activity values. This indicates that the pseudo-coincidence method has not eliminated all of the natural component. One has to note that this result is not proportional to the variations in the radon concentration. In the case of the Serrejón station (Fig. 1b), the temporal evolution of the alpha and beta indices is different. Now it increases with the apparent decrease in the radon concentration, indicating that, for this case, the subtraction of the natural contribution is too high. To resolve these discrepancies, daily adjustments should be made to the operation of the pseudo-coincidence circuit, as was proposed in an earlier work (Baeza et al., 1997).

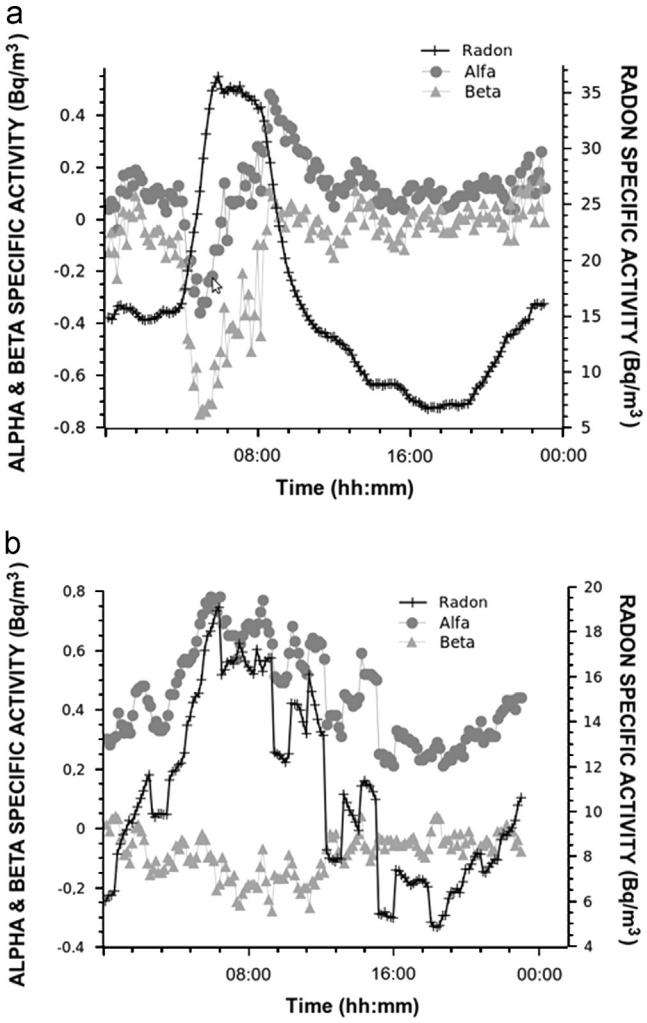

Fig. 1. Total alpha, total beta, and radon specific activities obtained for the currently operating RARE stations located in (a) Saucedilla, and (b) Serrejón (both in Extremadura, Spain).

2.1.2. Gaseous radioiodine monitor

The air from the continuous filter is passed by an additional pump at a flow rate of from 4 to 6 $m^3\ h^{-1}$ through a 5% TEDA (thrithylene di-amine) impregnated carbon cartridge which has a high affinity for the retention of the various species of iodine. Facing the cartridge, there is a NaI(Tl) detector coupled to two single-channel analyser circuits centred on the energy of the 364 keV photopeak to estimate indirectly its net peak area and thence its activity. Moreover, other gaseous radionuclides that might also be retained by the carbon cartridge cannot be identified.

2.2. Prototype of RARE's new atmospheric monitoring station

2.2.1. Brief description of the improvements

The essential modification is the substitution of the currently used detection systems by a gamma spectrometry system, for both the particulate and the gaseous fractions, using $LaBr_3$(Ce) or NaI (Tl) detectors. Both detectors are $2'' \times 2''$ in size, and are coupled to compact digital multichannel electronics, with the Genie2000 software package (Canberra Industries, 2009) being employed for the activity determinations.

Cartridge replacement has been automated by means of a new device which allows the programmable and remote removal of the measured cartridge, and its replacement with a new cartridge in front of the NaI (Tl) detector (Baeza et al., 2013b). The result is a drastic reduction in the frequency of maintenance tasks. Also, new software has been written for the acquisition control, and interpretation of the activities. This sets up the desired data acquisition logic, and allows the quasi-real-time calculation and reporting of the measured activities. It also determines the activities for cumulative spectra over configurable longer time intervals so that the station can also work as surveillance system determining even slight radiological alterations. For that purpose, it is essential to employ a proper background spectrum for activity calculations. The procedure can be summarized as follows: (i) insertion of a clean target (filter or cartridge); (ii) acquisition of short measurement spectra over that target and determination of the corresponding activities in quasi-real-time, subtracting the background spectrum acquired over the clean target (previous step) for the first spectrum, and storing this spectrum to be used as background for the following spectrum to be acquired, and so on; (iii) evaluation of the activities for the cumulative spectrum using the background acquired on the clean target; (iv) position of a new target and start again.

2.2.2. Efficiency calibration of the new detection systems

2.2.2.1. Aerosol monitor. Efficiency calibrations were carried out in order to tune the performance of the gamma spectrometer for the aerosols captured on the continuous filter, and to compare and evaluate the advantages and disadvantages of the use of different scintillation detectors. Two detectors were tested – one of $LaBr_3$(Ce) and one of NaI(Tl), both with a $2'' \times 2''$ crystal, but, because of the nature of these two materials, with different energy resolutions (specifically, 2.6% and 7%, respectively, for the Cs-137 photopeak). For the efficiency calibration, we used an experimental method based on fabricating a series of samples of known activity, with geometry as similar as possible to that of the aerosol deposition on the filter. This geometry is, of course, very different from those considered standard in a low-activity laboratory: a