Available online at www.sciencedirect.com
ScienceDirect
Applied Radiation and Isotopes 64 (2006) 1329–1332
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Optimization of counting times for short-lived gamma-ray emitters in air filter samples

M. Korun*

Jožef Stefan Institute, Jamova 39, Ljubljana, Slovenia

Abstract

A methodology for the optimization of the counting times in a series of measurements of gamma-ray emitters in air filters is presented. In the optimal measurement regime in measurements of all the filters in a batch, the same minimum detectable activity is attained. It is shown how the number of filters, the properties of the gamma-ray emitter and the equipment influence the measurement time of the batch of filters and the minimum detectable activity attained.

Keywords: Gamma-ray spectrometry; Air filters; Short-lived isotopes; Thoron daughters; Minimum detectable activities

1. Introduction

The presence of radionuclides in the air is usually determined by measuring the radioactivity of air filters that retain the radioactivity present in the air which is passed through them. The sensitivity of a measurement of the air concentration of a nuclide emitting gamma-rays with energy E is governed by its properties, i.e. its emission probability I and decay constant λ, by the conditions of sampling, i.e. the air-flow rate through the filter Φ and the sampling time S and by the conditions of the measurement, i.e. counting efficiency $\varepsilon(E)$, counting time T and measurement background. Assuming constant concentration of a radionuclide in the air, its minimum detectable activity concentration is given by Hurtgen et al. (2000):

$$\varsigma = \frac{4.78\lambda^2\sqrt{B}}{\varepsilon(E)I\Phi}\frac{1}{1-e^{-\lambda S}}\,e^{\lambda\Delta T}\frac{1}{1-e^{-\lambda T}}, \quad (1)$$

where ΔT denotes the time interval between the end of sampling and start of counting and the factor 4.78 expresses the probability that an error of the first or second kind occurs with a probability of 5%. B represents the number of background counts registered in the energy interval of a peak occurring at the energy E. It is assumed that the width of this interval is the spectrometer resolution at the energy E times 2.5, a factor which is usually used in gamma-ray spectrometric analysis (De Geer, 2004). The background in the spectrum where the photons emitted are registered arises due to the spectrometer background, which linearly increases with time, and the background due to the presence of radioactivity in air which is not due to the presence of the emitter to be detected. Usually, this radioactivity belongs to the short-lived radon and thoron daughters ^{214}Pb, ^{214}Bi and ^{212}Pb with half lives of 26.8 min, 19.9 min and 10.64 h, respectively. These radionuclides are attached to aerosols and are easily collected by aerosol filters. At an air-flow rate of 1 L/s and a concentration of radon and thoron daughters in the air of 10 Bq/m^3, as given by UNSCEAR (1993) as typical air concentrations above soil, the saturated activities of ^{214}Bi and ^{212}Pb on the filter are about 40 and 550 Bq, respectively. Usually, thoron daughters on the filter are the major influence on the measurement background since radon daughters may decay substantially between the end of collection and the start of measurement. It should be noted that a few hours are needed to collect the filters and start the measurements. Therefore, only the contribution of thoron daughters to the measurement background will be considered here.

*Tel.: +386 1 4773900; fax: +386 1 4773151.
E-mail address: matjaz.korun@ijs.si.

0969-8043/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2006.02.039

When considering detection limits in air filter measurements, two cases should be considered: radionuclides having much shorter and much larger half lives than ^{212}Pb. For a radionuclide having substantially longer half-live than ^{212}Pb, a suitable cooling time can be applied to reduce the ^{212}Pb contribution below the spectrometer background. Then, the background count rate is constant during a single measurement as well as in a series of consecutive measurements. On the other hand, when the half life of the radionuclide to be measured is shorter from that of ^{212}Pb, the counting time must also be shorter and consequently the background count rate decreases exponentially with time. As a consequence a series of consecutive measurements exhibits decreasing background count rates. It is the purpose of this contribution to present the counting times for a series of measurements of short-lived emitters which ensure a detection limit as low as possible.

2. The counting times

In a sampling campaign a batch of n filters is received which are to be analyzed on a gamma-ray spectrometer for the air concentration of a radionuclide with decay constant λ emitting gamma-rays of energy E. It is assumed that all filters are of the same geometry and were subject to the same air-flow rate during the same sampling periods. In an optimized counting regime the counting times are selected in such a way that the minimum detectable activities are equal in all measurements. Assuming that the sampling time is much longer than the half life of the radionuclide and the background is due to the presence of natural radioactivity in the air, the minimum detectable activity of the kth measurement is

$$\varsigma_k = \frac{4.78\lambda^2\sqrt{B(\Delta T_k, T_k)}}{\varepsilon I \Phi}\frac{\mathrm{e}^{\lambda\Delta T_k}}{1-\mathrm{e}^{-\lambda T_k}}, \tag{2}$$

where $B(\Delta T_k,\ T_k)$ denotes the background counts due to the presence of thoron in the air

$$B(\Delta T_k, T_k) = \dot{B}(0)\int_{\Delta T_k}^{\Delta T_k+T_k}\mathrm{e}^{-\lambda_B t}\,\mathrm{d}t = \frac{\dot{B}(0)}{\lambda_B}\mathrm{e}^{-\lambda_B\Delta T_k}(1-\mathrm{e}^{-\lambda_B T_k}). \tag{3}$$

Here $\dot{B}(0)$ denotes the background count rate at the end of the sampling campaign, λ_B the decay constant of the background, ΔT_k the time interval between the end of the sampling campaign and start of the measurement of the kth filter and T_k the measurement time of the kth filter. Assuming that the measurement time is much shorter than the background decay time, the minimum detectable activity of the kth measurement can be expressed as

$$\varsigma_k = \frac{4.78\lambda^2\sqrt{\dot{B}(0)/\lambda_B}}{\varepsilon I \Phi}\mathrm{e}^{-\lambda_B\Delta T_k/2}\,\mathrm{e}^{\lambda\Delta T_k}\frac{\sqrt{1-\mathrm{e}^{-\lambda_B T_k}}}{1-\mathrm{e}^{-\lambda T_k}} \approx \varsigma(0)\mathrm{e}^{(\lambda-\lambda_B/2)\Delta T_k}\frac{\sqrt{\lambda_B T_k}}{1-\mathrm{e}^{-\lambda T_k}}, \tag{4}$$

where $\zeta(0)$ stands for

$$\varsigma(0) = \frac{4.78\lambda^2\sqrt{\dot{B}(0)/\lambda_B}}{\varepsilon I \Phi}. \tag{5}$$

It can be observed that the minimum detectable activity can be expressed as a product of three factors, the first being defined by the sampling conditions, the measuring equipment and the properties of the radionuclide to be determined, the second by the time interval between sampling and counting and the third by the counting time. At a fixed ΔT_k the counting time necessary to attain the lowest possible minimum detectable activity is given by $\partial\zeta_k/\partial T_k = 0$ which leads to the equation

$$\frac{\lambda T_k\mathrm{e}^{-\lambda T_k}}{1-\mathrm{e}^{-\lambda T_k}} = \frac{1}{2}. \tag{6}$$

The solution of this equation is $\lambda T_k \cong 1.25$. The actual minimum detectable activity attained, ζ_k, depends on ΔT_k as well. In an optimized counting regime, where in measurements of all filters the same minimum detectable activity is attained, Eq. (6) defines the counting time of the last filter. In the previous measurements with a shorter counting time the same minimum detectable activity is attained due to the higher activity in the filter. Since the time interval ΔT_k equals to the sum of the counting times of the first $k-1$ filters, $\Delta T_k = \sum_{j=1}^{k-1} T_j$, and the relation between two successive counting times can be expressed by

$$\varsigma(0)\mathrm{e}^{(\lambda-\lambda_B/2)\sum_{j=1}^{k-1}T_j}\frac{\sqrt{\lambda_B T_k}}{1-\mathrm{e}^{-\lambda T_k}} = \varsigma(0)\mathrm{e}^{(\lambda-\lambda_B/2)\sum_{j=1}^{k-2}T_j}\frac{\sqrt{\lambda_B T_{k-1}}}{1-\mathrm{e}^{-\lambda T_{k-1}}} \tag{7}$$

from which

$$\sqrt{\frac{\lambda_B T_{k-1}}{\lambda_B T_k}}\frac{1-\mathrm{e}^{-\lambda T_k}}{1-\mathrm{e}^{-\lambda T_{k-1}}} = \mathrm{e}^{(\lambda-\lambda_B/2)T_{k-1}} \tag{8}$$

follows. Since the counting time of the last filter is given by $\lambda T_n = 1.25$ the counting time of its predecessor, T_{n-1}, is the solution of the equation

$$\frac{T_{n-1}\mathrm{e}^{-(2-\lambda_B/\lambda)\lambda T_{n-1}}}{(1-\mathrm{e}^{-\lambda T_{n-1}})^2} = \frac{T_n}{(1-\mathrm{e}^{-\lambda T_n})^2}. \tag{9}$$

To calculate the earlier counting times this relation is applied recursively. In an optimized counting regime the products of the second and the third factor in Eq. (4) for all measurements in a batch are equal.

Short-lived radionuclides can be released to the atmosphere during normal operations of nuclear reactors or in event of an accident. The off-site concentrations of radionuclides is determined from the activity of air filters sampled at a system of locations. Because of their radiobiological importance and high abundance in the reactor the concentration of iodine isotopes in the air is of principal concern (Alpert et al., 1986). Therefore, the methodology of optimization and the assessment of the minimum detectable activities is applied to the short-lived