Journal of Environmental Radioactivity 101 (2010) 692–699

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

Multi-group approximation, scattering and calibration coefficients, uncertainty estimates and detection limits of a NaI(Tl)-based gamma spectrometry set-up for low-level activity analysis

Ateba P. Owono*

Laboratoire de Physique Nucléaire, Faculté des Sciences, Université de Yaoundé I, Cameroon

ARTICLE INFO

Article history:
Received 7 May 2009
Received in revised form
19 January 2010
Accepted 5 April 2010
Available online 14 May 2010

Keywords:
Environmental assessment
Gamma spectroscopy
Photon scattering coefficients
Scintillation detector
Calibration coefficients
Uncertainty estimation
Sensitiveness
Detection limits
Negative activity

ABSTRACT

This paper describes a quantitative radioactivity analysis method especially suitable for environmental samples with low-level activity. The method, consisting of a multi-group approximation based on total absorption and Compton spectra of gamma rays, is coherently formalized and a computer algorithm thereof designed to analyze low-level activity NaI(Tl) gamma ray spectra of environmental samples. Milk powder from 1988 was used as the example case. Included is a special analysis on the uncertainty estimation. Gamma sensitiveness is defined and numerically evaluated. The results reproduced the calibration data well, attesting to the reliability of the method. The special analysis shows that the uncertainty of the assessed activity is tied to that of the calibration activity data. More than 77% of measured 1461-keV photons of ^{40}K were counted in the range of clearly lower energies. Pile-up of single line photons (^{137}Cs) looks negligible compared to that of a two-line cascade (^{134}Cs). The detection limit varies with radionuclide and spectrum region and is related to the gamma sensitiveness of the detection system. The best detection limit always lies in a spectrum region holding a line of the radionuclide and the highest sensitiveness. The most radioactive milk powder sample showed a activity concentration of 21 ± 1 Bq g^{-1}for ^{137}Cs, 323 ± 13 Bq g^{-1} for ^{40}K and no ^{134}Cs.

1. Introduction

Usually, the radioactivity of environmental samples of sites and products suspected of contamination must be analyzed before free access is given to the public. Environmental samples generally have low-level activity. Precision and reliability of the results obtained in low-level activity analysis require particular attention. As a matter of fact, subtracting background from gross counts can in some cases either lead to a "negative activity", indicate the absence of a radioisotope that is actually present, or the presence of a radioisotope that is absent. A special analysis of both, uncertainty and detection limit, is therefore necessary and topical in this paper.

Many laboratories use liquid nitrogen cooled semiconductor detectors providing very fine peaks, easy peak identification and precise results. But nitrogen liquefaction is expensive and out of reach of many laboratories. A scintillation detector of NaI(Tl)-type is far cheaper to purchase, it is sturdy, seldom needs servicing and is notably efficient for gamma ray counting. It is used throughout the World for gamma spectroscopy in general, especially to monitor radioactivity of marine (Vlachos, 2005), terrestrial and atmospheric (Tyler, 2004) environment. Further investigations are therefore carried out to improve the ability of a NaI(Tl) spectrometry for quantitative determination of radioelements. But even the method presented by Muminov et al. (2005) still is based on subtraction of spectra and by taking into account mainly the total absorption peak. On the other hand, radioactive decay obeys Poisson statistics stating the relative error for n counts to be $1/\sqrt{n}$. So the relative error for 2 counts is 70.71% and that for 1 count is 100%. The less the counts are, the higher the error. This limits the accuracy in low-level and especially in very low-level activity samples to the extent of wondering if it is really a peak and not an irregularity of the background spectrum. To decide, one can take advantage of the multi-group approximation, known among others in nuclear reactor physics (Owono, 1978), and the scattering coefficients method. Both concepts are based on theory.

2. Theoretical concept

2.1. Multi-group approximation

The scattering coefficients method aims at quantitatively mastering the influence of all physical processes at stake in

* Tel.: +237 77 47 91 94/22303278.
E-mail address: aowono1@yahoo.fr

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doi:10.1016/j.jenvrad.2010.04.008

a radiation field. Looking at gamma radiation of a radionuclide, one can neglect the fraction of induced gamma emitting nuclear reactions and take into account only the absorption and the scattering processes. The photon can be absorbed or scattered in the sample itself, or in the lead shield, or even in the scintillation detector; all that makes together the scattering and absorbing environment.

For given materials, sample and geometry of the arrangement of the detection system, let $\rho(E' \rightarrow E)\mathrm{d}E'$ denote the probability of density $\rho(E' \rightarrow E)$ for a photon of energy between E′ and E′+dE′ to be scattered towards energy E and let $\rho(E \rightarrow E'')\mathrm{d}E''$ denote the probability for a photon of energy E to be scattered towards an energy between E'' and $E'' + \mathrm{d}E''$. Then the flux $\Phi(E,t)$ of photons of energy E at given time t is described by the solution $\Phi(E,t)$ of the stationary equation $\mathrm{d}\Phi = 0$ within time interval dt.

The flux variation $\mathrm{d}\Phi$ is a compound of four terms:

- the source term $\dot{Q}(E,t)\mathrm{d}t$ of photons of Energy E produced at time t during dt by a present source,
- the scattering term $\int_0^\infty \Phi(E',t)\rho(E' \rightarrow E;t)\mathrm{d}E'\mathrm{d}t$ into energy E,
- the absorption term $a(E)\Phi(E,t)\mathrm{d}t$ of photons of energy E and absorption probability $a(E)$,
- the scattering term $\int_0^\infty \Phi(E,t)\rho(E \rightarrow E'';t)\mathrm{d}E''\mathrm{d}t$ from energy E to energy E''.

The defined integral $\int_0^\infty \rho(E \rightarrow E'';t)\mathrm{d}E''$ at given time t is a function of E and describes the probability $p(E)$ for a photon of energy E to be scattered at all:

$$p(E) = \int_0^\infty \rho(E \rightarrow E'';t)\mathrm{d}E''. \quad (1)$$

The absorption-to-scattering ratio $r(E) = a(E)/p(E)$ is a well-defined function at every value of energy E. The stationary equation is then

$$\dot{Q}(E,t) + \int_0^\infty \Phi(E',t)\rho(E' \rightarrow E)\mathrm{d}E' - [a(E)+p(E)]\Phi(E,t) = 0. \quad (2)$$

The solution of which has the shape

$$\Phi(E,t) = \frac{\dot{Q}(E,t) + \int_0^\infty \Phi(E',t)\rho(E' \rightarrow E)\mathrm{d}E'}{a(E)+p(E)}. \quad (3)$$

Let

$$S_E = \int_0^\infty \Phi(E',t)\rho(E' \rightarrow E)\mathrm{d}E' \quad (4)$$

denote the total number of photons scattered to E and

$$\dot{Q}_E = \dot{Q}(E,t) \quad (5)$$

the total number of source gamma photons of energy E at given time t, $\Phi(E,t)$ then depicts a photon spectrum $\Phi(E)$ that approaches a hyperbolic function of $p(E)$:

$$\Phi(E) = \frac{\dot{Q}_E + S_E}{[r(E)+1]p(E)}. \quad (6)$$

The quantity $r(E)$ is available through cross section values (Miyasaka and Minami, 1977; Berger et al., 1990, 1998). Thus the scattering probability cannot be 0.

At given time t and energy E, let $\phi_E = \phi(E)$ and $p_E = p(E)$, then for a NaI(Tl) scintillation detector with a gamma ray efficiency of 5–10%, it is $r(E)<0.1$ and $\phi_E \approx \dot{Q}_E + S_E/p_E$.

As Fig. 1a shows: $\phi(E)$ is then minimal at $p(E) = 1$ because all photons produced at energy E by the source $\dot{Q}_E$ and through "inwards" scattering S_E are scattered away from E. So, photons from previous generations are no longer there. For $p(E) < 1$, the incoming photons $\dot{Q}_E + S_E$ are added to non scattered photons from previous generations: $\phi(E) > \dot{Q}_E + S_E$. Fig. 1b shows the general case (Eq. (6)) with gamma ray absorption: at p(E) = 1, φ(E)is minimal but less than $\dot{Q}_E + S_E$ because a fraction of all photons produced at energy E by the source $\dot{Q}_E$ and "inwards" scattering S_E is absorbed and again photons from previous generations are no longer there.

Back to Eq. (3): Heisenberg's uncertainty principle and experimental broadening causes width ΔE of the line of energy E. Considering a gamma spectrum line of mean energy E_k: $\dot{Q}_E = 0$ outside interval $E_k \pm \Delta E_k$ and $\dot{Q}_E \neq 0$ within that interval. One can then substitute:

- to $\Phi(E)$ the true counting rate n_i of photons of energy E between $E_i - \Delta E_i$ and $E_i + \Delta E_i$,
- to $\Phi(E')$ the activity A_k emitting photons of energy E′ between $E_k - \Delta E_k$ and $E_k + \Delta E_k$,
- to $p(E) = \int_0^\infty \rho(E \rightarrow E'';t)\mathrm{d}E''$ the probability p_i for a photon of energy E between $E_i - \Delta E_i$ and $E_i + \Delta E_i$ to be scattered at all.

Since energy intervals $2\Delta E_i$ around E_i and $2\Delta E_k$ around E_k set energy groups, respectively, numbered i and k with corresponding average group energies E_i and E_k, advantage can now be taken of the multi-group approximation to numerically handle the integral Eq. (2).

2.2. Scattering coefficients

To quantitatively describe the contribution of group k to group i, an average probability $\overline{p}_{ki}$ for scattering photons of energy E' between $E_k - \Delta E_k$ and $E_k + \Delta E_k$ from group k to group i of photons of energy E between $E_i - \Delta E_i$ and $E_i + \Delta E_i$, can be defined by

$$A_k \overline{p}_{ik} = \int_{E_k-\Delta E_k}^{E_k+\Delta E_k} \Phi(E')\rho(E' \rightarrow E)\mathrm{d}E' \quad (i \neq k) \quad (7)$$

Then Eq. (3) becomes

$$\frac{1}{\eta_i} n_i = \sum_k \frac{\overline{p}_{ik}}{(1+r_i)p_i} A_k; \quad (8)$$

where η_i denotes the average of the energy dependent efficiency $\eta(E)$ of the scintillation detector for photons of energy E within group i; $\overline{p}_{ik}/(1+r_i)p_i$ is the scattering coefficient, since it is built of scattering probabilities alone. The true counting rate of group i is deduced to

$$n_i = \frac{\eta_i}{(1+r_i)p_i} \sum_k \overline{p}_{ik} A_k. \quad (9)$$

η_i, p_i, $\overline{p}_{ik}$ and r_i are group constants.

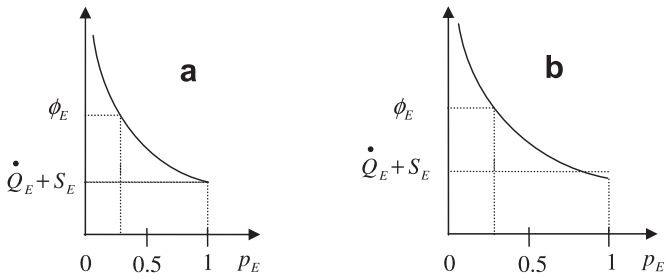

Fig. 1. Photon flux $\varphi(E)$ as a function of scattering probability $p(E)$ at given energy E: (a) for a NaI(Tl) scintillation detector of typical 5 to 10 % efficiency, (b) taking in account gamma ray absorption.