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Self-attenuation correction factors for bioindicators measured by γ spectrometry for energies < 100 keV

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ABSTRACT

The mass attenuation coefficients for a number of marine and terrestrial bioindicators were measured using γ spectrometry for energies between 22 and 80 keV.

These values were then used to find the correction factor k for the apparent radioactivity. The experimental results were compared with a Monte Carlo simulation performed using PENELOPE in order to evaluate the reliability of the simplified calculation and to determine the correction factors.

1. Introduction

Radiological monitoring laboratories need to assess the marine and terrestrial environment in order to verify the radioactivity level, especially around sensitive facilities handling nuclear materials.

The radioactivity in environmental samples is measured with various techniques, among them direct γ−ray spectrometry is preferred as it is non-destructive, relatively simple to use and less expensive than Radiochemical Neutron Activation Analysis (RNAA) or Accelerator Mass Spectrometry (AMS).

The activity of the sample is obtained measuring its gamma emission rate with a germanium detector calibrated with a multinuclide standard source to determine the efficiency for various energies in the range of interest. In this respect, improvements in shielding and studies in calibrating functions [1–3] allowed LEPS (Low Energy Photon Spectroscopy) to be applied to environmental applications.

Environmental samples exhibit a low counting rate. Thus, in order to lower the detection limit to few Becquerel per kilogram, the mass of the sample used in the measurements must be substantial. The samples, dried, ground and homogenized, are packed into cylindrical polyethylene containers (matrices). Their height ranges from 2 to 7.2 cm, inducing attenuation at low energies.

As it is well known, self-absorption acquires importance for radiating energies lower than 100 keV. In fact, at these energies the photoelectric interaction predominates over coherent and incoherent scattering, increasing the probability of complete absorption of the gamma rays by the atoms of the material. This can be observed, for example, in the case of ^{129}I in the presence of ^{127}I where, depending on stable iodine concentration, an attenuation up to 30% was reported [4–7].

Since, over the energy range in question, gamma rays may experience total absorption or Compton scattering, the beam arrives at the detector attenuated and faded in energy. Therefore, accurate gamma-assays require correction factors for the measured (apparent) activity in order to account for the large losses in the total number of soft gamma rays in the counting measurements. The attenuation phenomenon depends on various factors: the emission energy, the sample composition and density, the shape and the size of the container, the distance between the sample and the detector and the detector response. It is thus necessary to perform an accurate measurement of the mass energy attenuation coefficient $\mu_m(E)$ for different energies. The correction factors may then be deduced in different ways: either solving the resulting equations by numerical methods [1,8] or using Monte Carlo simulations [9–12]. In particular, the Monte Carlo technique allows the generation of a very large number of events where each individual photon is followed from its

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emission in the source volume to its complete absorption. In this way it is possible to obtain an average behavior at a given energy. However, the use of this technique needs the description of the detector geometry as precisely as possible and requires skills and experience in simulations. This is why laboratories assessing environment usually prefer a straightforward procedure for the correction of environmental analysis.

To this end, a summary of the work accomplished on a large number of bioindicators collected up on the French coasts close to nuclear facilities will be presented. The study will be limited to energies below 100 keV. The correction factors, calculated within an approximated analytical model, will be compared with those obtained by simulation with a Monte Carlo code in order to validate the calculation.

2. Experimental set-up and mass coefficients determination

The marine and terrestrial samples to be assessed for small amounts of ^{129}I (29 keV), ^{241}Am (59 keV), ^{210}Pb (45 keV), were collected up on the coasts of the French towns of Brest, Cherbourg and Toulon, around nuclear activity discharge points. They are listed in Table 1 with their densities (obtained by precision mass measurements). These samples were dried and ground to obtain an homogeneous powder which perfectly filled the geometries of the following containers (according to Ref. [26]): *SG*500 (500 cm^3), Optima Jar (220 cm^3) and plexiglass cylinders (diameter: 8 cm) of two different heights (2 and 5 cm).

To measure the mass energy attenuation coefficient $\mu_m(E)$ with $[\mu_m] = (\mathrm{cm}^2\,\mathrm{g}^{-1})$, for each energy and for each geometry, transmission measurements were performed [16–18,4]. The samples were submitted to irradiation with fluorescence X from Ag, Cd, Sb, I and with X and gamma emission from ^{133}Ba and ^{241}Am. To detect the gamma rays from the matrices, a high purity germanium *HPGe GMX* type detector from *ORTEC* was employed. Its resolution is 1.79 keV at 1.33 MeV and, for the same energy, its relative efficiency is 15.5% [19]. In order to cut all the gamma rays not parallel to the detector axis, two lead collimators (5 mm thick with a hole diameter of 5 mm) were placed between the source and the sample and between the sample and the detector. Their employ allowed to reproduce suitable experimental conditions for the application of the point kernel approximation (see next section). During the measurements, the source, the cylindrical matrix, the detector and the collimators were centered along the same axis.

According to the fundamental law of gamma ray attenuation, for each energy E and for a sample of mass m expressed in (g) and surface S in (cm^2), the mass energy attenuation coefficient $\mu_m(E)$, in $[\mu_m] = (\mathrm{cm}^2\,\mathrm{g}^{-1})$, writes:

$$\mu_m(E) = \frac{S}{m} \ln \frac{N_0(E)}{N_{(x)}(E)}. \quad (1)$$

In Eq. (1), $N_0(E)$ represents the incident number of gamma rays, while $N_{(x)}(E)$ is the transmitted number of rays after they traveled a distance x inside the sample matrix. For each empty geometry and for each energy, the initial number N_0 of incident gamma rays over a counting time of 1000 s was measured. The transmission measurements were performed afterwards on a longer acquisition time ($t_{max} = 100\,000$ s) to determine the net counting rate $N_{(x)}$. The mass energy attenuation coefficient was then determined for each sample and geometry, according to the different spectral lines. Finally, for each energy, the $\mu_m(E)$ averaged on the different geometries was computed. Tables 2–4 show the mass energy attenuation coefficients for different samples. The species are listed in the table captions.

The values of the mass energy attenuation coefficient for *Fucus serratus* (in Table 2) were in fair agreement with those found in Refs. [4–6].

Table 1
Marine and terrestrial bioindicators.

Sample matrix	Density (g cm^{-3})	State
Fucus V.	0.87 ± 0.04	Dry
F. serratus	0.99 ± 0.10	Dry
Limpets	0.85 ± 0.04	Dry
Posidonies	0.59 ± 0.04	Dry
Mussels	0.86 ± 0.04	Dry
Pinus	0.31 ± 0.04	Dry
U. europaeus	0.48 ± 0.06	Dry
P. lentiscus	0.42 ± 0.02	Dry

Table 2
Mass energy attenuation factors $\mu_m = \mu_m(E)$ in cm^2 g^{-1} for the brown algae *Fucus* and for the seagrass *Posidonia*.

E (keV)	*F. serratus*	*Fucus V.*	*Posidonia*
22.20	1.30 ± 0.22	1.44 ± 0.08	2.11 ± 0.28
23.20	1.15 ± 0.18	1.26 ± 0.03	1.87 ± 0.23
26.34	0.84 ± 0.11	0.91 ± 0.07	1.32 ± 0.13
28.60	0.69 ± 0.10	0.74 ± 0.02	1.08 ± 0.12
30.90	0.61 ± 0.07	0.61 ± 0.02	0.86 ± 0.08
34.90	0.49 ± 0.04	0.48 ± 0.01	0.66 ± 0.08
59.54	0.22 ± 0.02	0.22 ± 0.01	0.26 ± 0.03
81.00	0.18 ± 0.01	0.18 ± 0.01	0.18 ± 0.01

Table 3
Mass energy attenuation factors $\mu_m = \mu_m(E)$ in cm^2 g^{-1} for the gastropods *Limpets* and the bivalve molluscs *Mussels*.

E (keV)	*Limpets*	*Mussels*
22.20	1.36 ± 0.17	1.14 ± 0.13
23.20	1.24 ± 0.09	1.02 ± 0.08
26.34	0.90 ± 0.12	0.76 ± 0.07
28.60	0.74 ± 0.02	0.63 ± 0.06
30.90	0.63 ± 0.03	0.53 ± 0.02
34.90	0.49 ± 0.02	0.42 ± 0.04
59.54	0.25 ± 0.01	0.23 ± 0.03
81.00	0.21 ± 0.01	0.19 ± 0.03

Table 4
Mass energy attenuation factors $\mu_m = \mu_m(E)$ in cm^2 g^{-1} for the Coniferous tree *Pinus*, the gorse *U. europaeus* and the evergreen shrub *Pistacia lentiscus*.

E (keV)	*Pinus*	*U. europaeus*	*P. lentiscus*
22.20	0.65 ± 0.06	0.69 ± 0.06	0.79 ± 0.06
23.20	0.59 ± 0.05	0.61 ± 0.06	0.70 ± 0.11
26.34	0.46 ± 0.04	0.47 ± 0.05	0.53 ± 0.02
28.60	0.40 ± 0.02	0.42 ± 0.05	0.44 ± 0.02
30.90	0.36 ± 0.02	0.34 ± 0.02	0.36 ± 0.01
34.90	0.28 ± 0.02	0.28 ± 0.02	0.28 ± 0.03
59.54	0.21 ± 0.01	0.20 ± 0.01	0.21 ± 0.01
81.00	0.17 ± 0.01	0.18 ± 0.01	0.17 ± 0.01

3. Determination of the correction factor *k*

As discussed in the previous sections, a multiplicative correction factor is applied to the apparent activity. In the following it will be named *k*. This factor, depending on a given geometry and energy, is formally defined as the ratio of the full energy interaction rate in a non-absorbing medium to the actual rate measured from the sample [16,18,20]. In practice *k* is deduced