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Calibration with MCNP of NaI detector for the determination of natural radioactivity levels in the field

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ABSTRACT

In view of assessing natural radioactivity with on-site quantitative gamma spectrometry, efficiency calibration of NaI(Tl) detectors is investigated. A calibration based on Monte Carlo simulation of detector response is proposed, to render reliable quantitative analysis practicable in field campaigns. The method is developed with reference to contact geometry, in which measurements are taken placing the NaI(Tl) probe directly against the solid source to be analyzed. The Monte Carlo code used for the simulations was MCNP. Experimental verification of the calibration goodness is obtained by comparison with appropriate standards, as reported. On-site measurements yield a quick quantitative assessment of natural radioactivity levels present (^{40}K, ^{238}U and ^{232}Th). On-site gamma spectrometry can prove particularly useful insofar as it provides information on materials from which samples cannot be taken.

1. Introduction

The detection and measurement of natural radioactivity is a consolidated field of investigation, playing a fundamental role in many aspects of environmental and health sciences. The latter, in particular, has become of ever more special concern after the release of the 2013 Euratom Directive stressing radon and radioactivity of building materials as prominent concerns in radiation protection (EC, 2013). Often, work in this research area requires collecting large sets of data to produce the desired spatial resolution: hence the need for experimental methods yielding the needed data quality and reliability while keeping field and laboratory activity – and the associated costs – at a reasonable level. Many techniques are available, ranging from laboratory gamma spectrometry, mostly with high purity germanium (HPGe) detectors, to field gamma ray spectrometry with NaI(Tl), HPGe and other detectors, e.g., BGO, CZT or LaBr3. However, *in situ* measurements are often required for a number of possible reasons. For instance: having to characterize a wide area, which would require a very large number of samples to be collected and brought to the laboratory (Miller and Shebell, 1993); or having to investigate objects that cannot be sampled or removed, such as archaeological sites or buildings (Nucciatelli, 2008); and so forth.

HPGe detectors provide very accurate quantitative results thanks to their high spectral resolution. This technique is very powerful when applied to radioactive disequilibrium caused by environmental/geochemical behavior affecting partitioning of elements within natural radioactive families (Gilmour, 2008; Ivanovich and Harmon, 1982). On the other hand, HPGe detectors require long counting times and sophisticated cooling systems: these two requirements pose no major problem in laboratory measurements but render HPGe detectors quite awkward for field measurements, when not downright unfit. Furthermore, NaI detectors have much higher detection efficiencies than HPGe ones, producing a significant reduction in measurement time, and making NaI detectors a favorite for *in situ* measurements.

Once their dependence on temperature is suitably accounted for (Ianakiev, 2009; Kempa, 2013), utilization of NaI(Tl) detectors is easy and practicable in most environmental conditions, as witnessed by the extensive literature covering land application as well as aircraft borne and even underwater ones (Jigiri and Farai, 2005; Baré and Tondeur, 2010; Povinec et al., 1996, 2008; Strati et al., 2014; Van Put et al., 2004; Vlastou et al., 2006; Wedekind, 1999;

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Tsabaris et al., 2008, 2012; Zhang et al., 2015; Androulakaki et al., 2015).

In spite of the wide utilization of NaI detectors over many decades, there is still a lack of concurrence on protocols for the efficiency calibration for quantitative analysis. In view of on-site gamma spectrometry to estimate natural radioactivity levels in the field - specifically ^{40}K, ^{238}U and ^{232}Th contents - NaI(Tl) detectors are usually calibrated by means of standard surfaces, mostly concrete pads containing a known concentration of radionuclides. However, these calibration pads are not available everywhere; and on the other hand, they are not simple to build with the needed homogeneity, may be expensive and, furthermore, may prove complicated to manage insofar as radiation protection (Chiozzi et al., 2000).

The present work focuses on the setup of a method for the efficiency calibration of NaI detectors for on-site gamma spectrometry, based on Monte Carlo techniques. The method is developed for contact geometry, i.e., measurements obtained with the NaI(Tl) probe positioned directly against the solid source under investigation. The goodness of the calibration is verified experimentally, with measurements on standards of known radioactivity content. It affords a suitable method for quick quantitative analysis of natural radioactivity levels in the field.

2. Materials and method

A 3″ × 3″ NaI(Tl) detector, model 905-4 (Ortec-Ametek) was used for on-site gamma-spectrometry. The photomultiplier tube (PMT) was interfaced with a 12" netbook (Samsung) via a digiBASE (Ortec) (PMT base). The spectra were acquired and elaborated with Scintivision MCA (Ortec). The FWHM was equal to 46 keV at 662 keV and 65 keV at 1330 keV.

2.1. High resolution gamma spectrometry

Albeit the spectra of interest were collected with the NaI(Tl) detector, gamma spectra of soil samples were collected and analyzed in the laboratory on high resolution detectors, to characterize radiometrically the extended sources used for Monte Carlo modeling. To this end, a coaxial p-type HPGe detector by Ortec-Ametek (relative efficiency: 32.5%, FWHM equal to 1.8 keV at 1330 keV and energy range 50–2000 keV) was used. Soil samples were dried, homogenized and sieved at 2 mm. The system was calibrated for energy and efficiency using a multiple nuclide source (QCY48, Amersham) in jar geometry (diameter: 56 mm; thickness: 10 mm). Counting time of samples was one day. Spectra were analyzed with the GammaVision-32 software (version 6.07, Ortec). Quantitative analysis on samples was obtained subtracting the spectrum of water in the same geometry, while uncertainty on peaks (k = 1.68% level of confidence) was calculated propagating the combined uncertainty over the efficiency fit previously determined with the counting uncertainty.

Minimum detectable activity was calculated making use of the Traditional ORTEC method with a peak cut-off limit of 40%. ^{232}Th was determined using the emissions of its radioactive descendant ^{228}Ac (911 keV). ^{238}U was determined using the emissions of its radioactive descendant ^{226}Ra (186.2 keV). For the correction of the ^{226}Ra peak at 186 keV secular equilibrium between ^{226}Ra–^{238}U and natural $^{235}U/^{238}U$ isotopic ratio was assumed (Gilmore, 2008). Under these hypotheses the ^{226}Ra peak was corrected dividing by 1.7337.

2.2. MCNP5 – general features

The Monte Carlo method can be used to duplicate theoretically a statistical process, such as the interactions of nuclear particles with materials. This method is particularly useful for complex problems that cannot be modeled accurately by deterministic methods. MCNP - Monte Carlo N-particle - is of widespread use in modeling neutron, electron, photon or coupled neutron/electron/photon transport (Briesmeister, 1993). The code –handles arbitrary three-dimensional configurations of materials in geometric volumes bounded by various types of surfaces. Pointwise cross-section data are used typically, albeit grouped data are also available.

The user can create input files containing data regarding.

- geometry specification;
- description of materials and selection of the cross-section evaluations;
- definition of the radiation source;
- information related to the transport and the theoretical model to be applied;
- type of response - called tallies - desired.

It is to be noted that MCNP takes into account implicitly the self absorption of radiation by the volume under investigation.

2.3. NaI detector simulation

The experimental setup for the calibration of the NaI detector described above was modeled with MCNP5 (LANL, 2003). The photomultiplier (PM) tube is separated from the NaI crystal by a 5 mm thick glass window. The photodetector is protected by a 0.05 mm thick aluminum housing, separated from the crystal by a very thin air gap. The thickness of air between the aluminum and the crystal and between the aluminum and the photomultiplier is 0.25 mm on the sides and 0.2 mm at the top and bottom. The dimensions of the integral unit in its casing are: 8.2 cm of diameter in the crystal part and 5.8 cm in the photomultiplier part, 22.35 cm of length. The PM tube base, digiBASE (Ortec), has dimensions: 6.3 cm of diameter and 8.0 cm of length.

The main features and characteristics included in the Monte Carlo model are:

- The NaI crystal: 3″ × 3″, $\rho = 3.6667$ g/cm$_3$;
- The glass separation between the crystal and the PM tube, $\rho = 2.200$ g/cm$_3$;;
- The PM-base: $\rho = 1.2070$ g/cm$_3$
- The digiBase: $\rho = 1.2070$ g/cm$_3$;
- The aluminum housing of the NaI/PM set: $\rho = 2.7020$ g/cm$_3$.

Both the PM tube and the digiBase have been modeled as volumes of constant composition and mean density. The density of the PM tube was selected as discussed in (Baré, 2011) while the digiBase was assumed entirely made of polystyrene.

2.4. Sources matrices simulation

Two different sets of simulations were run: for loose soil and for solid rock. To model the first matrix the elemental composition of a typical alluvial soil from the eastern Po plain (SiO_2, 45.28; TiO_2, 0.58; Al_2O_3, 14.94; Fe_2O_3, 5.6; MnO, 0.15; MgO, 4.13; CaO, 9.01; Na_2O, 0.88; K_2O, 2.32; P_2O_5, 0.15; LOI, 16.96) was considered; for the second matrix the features were chosen of a volcanic rock from the Vulsini District (Northern Latium, Italy) investigated in a previous work (V01 sample in Capaccioni et al., 2013). The known density of 1.3 g/cm^3 and 2.0 g/cm^3, for soil and rock respectively, was used.

2.5. Simulation parameters

Source volume: simulations require a finite volume to be