Applied Radiation and Isotopes 127 (2017) 142–149

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

Uncertainty quantification applied to the radiological characterization of radioactive waste

B. Zaffora[a,*], M. Magistris[a], G. Saporta[b], J.-P. Chevalier[b]

[a] *Radiation Protection Group, European Organization for Nuclear Research, 1211, Geneva 23, Switzerland*
[b] *CNAM, 292 Rue Saint-Martin, 75003 Paris, France*

HIGHLIGHTS

- Uncertainty quantification is essential for radioactive waste acceptance in final repositories.
- Uncertainty can be quantified using classical or intensive numerical methods.
- A procedure to estimate the activity and uncertainty of ETM, DTM and ITM radionuclides is suggested.
- The principal contributor to the hazard factor is the activity of the major gamma-emitters.

ARTICLE INFO

Keywords:
Radioactive waste
Uncertainty quantification
Particle accelerator
Random vector

ABSTRACT

This paper describes the process adopted at the European Organization for Nuclear Research (CERN) to quantify uncertainties affecting the characterization of very-low-level radioactive waste. Radioactive waste is a by-product of the operation of high-energy particle accelerators. Radioactive waste must be characterized to ensure its safe disposal in final repositories. Characterizing radioactive waste means establishing the list of radionuclides together with their activities. The estimated activity levels are compared to the limits given by the national authority of the waste disposal. The quantification of the uncertainty affecting the concentration of the radionuclides is therefore essential to estimate the acceptability of the waste in the final repository but also to control the sorting, volume reduction and packaging phases of the characterization process. The characterization method consists of estimating the activity of produced radionuclides either by experimental methods or statistical approaches. The uncertainties are estimated using classical statistical methods and uncertainty propagation. A mixed multivariate random vector is built to generate random input parameters for the activity calculations. The random vector is a robust tool to account for the unknown radiological history of legacy waste. This analytical technique is also particularly useful to generate random chemical compositions of materials when the trace element concentrations are not available or cannot be measured. The methodology was validated using a waste population of legacy copper activated at CERN. The methodology introduced here represents a first approach for the uncertainty quantification (UQ) of the characterization process of waste produced at particle accelerators.

1. Introduction

The present study introduces an approach to estimate uncertainties in radioactive waste characterization.

The characterization of radioactive waste is a complex task, especially when historical waste is involved. At the European Organization for Nuclear Research (CERN), γ-ray spectrometry is used to estimate the specific activity of easy-to-measure (ETM) radionuclides (IAEA, 2007). Difficult-to-measure (DTM) radionuclides, which are β and low-energy X-ray emitters, are either measured by radiochemical techniques or evaluated by calculations and Monte Carlo simulations (ISO, 2007, 2013). The specific activities are then compared to the acceptance limits of the national agencies for waste management. Waste producers must ensure that these limits are respected and must estimate the distribution of the quantities of interest together with their uncertainties (Classification, 1994; Lowenthal, 1998).

We present three methodologies to estimate the activity of DTM radionuclides and for each method we describe the associated uncertainty quantification (UQ) strategy. We introduce the application of linear regression, geometric mean and the Mean Activity Method for

---

[*] Corresponding author.
E-mail address: biagio.zaffora@cern.ch (B. Zaffora).

http://dx.doi.org/10.1016/j.apradiso.2017.06.001
Received 17 January 2017; Received in revised form 3 May 2017; Accepted 1 June 2017

DTM's activity estimation. Confidence intervals, geometric standard deviation and standard errors are discussed for UQ purposes.

We conclude by presenting an analytical technique to predict the activity of the so-called impossible-to-measure (ITM) radionuclides (ISO, 2013). This technique lies on the construction of a mixed multivariate random variable to stochastically extract activation parameters.

We finally combine the UQ techniques into a global scheme to quantify the uncertainty of the waste characterization process. In the following sections we adopt the terminology presented by the Guide to the expression of uncertainty in measurement (GUM) (JCGM 100, 2008). In this article the term "standard uncertainty" is used to express the standard deviation of a given quantity.

2. Acceptance and hazard factors

CERN eliminates its radioactive waste in the repositories of its two Host States, France and Switzerland, in accordance with a tripartite agreement between the Organization and the latter, signed on 15 November 2010 and entered into force on 16 September 2011.

Very-low-level radioactive waste (VLLW) produced at CERN is disposed of in the French repository in the Aube district. The acceptance criteria are based on the hazard factor called IRAS (Radiological Acceptance Index in Storage):

$$IRAS = \sum_i \frac{a_i}{L_i} \tag{1}$$

where a_i is the specific activity of the radionuclide i (in Bq/g) and L_i is the specific activity limit of the radionuclide i defined as $L_i = 10^{Class_i}$. The class of a radionuclide gives information on its radiotoxicity and varies from 0 to 3 (Critères, 2013).

Waste is accepted at the final repository if the IRAS of each package is below 10 and the weighted IRAS of the batch is below 1:

$$IRAS_{batch} = \frac{\sum_j IRAS_j \times M_j}{\sum_j M_j} < 1 \tag{2}$$

where $IRAS_j$ is the IRAS of the package j as given by Eq. (1) and M_j is its weight.

The activity of ETM, DTM and ITM radionuclides are estimated using different techniques. To make explicit the contribution to the IRAS of each one of these families Eq. (1) can be rewritten as follows:

$$IRAS = \sum_l \frac{a_{ETM,l}}{L_l} + \sum_m \frac{a_{DTM,m}}{L_m} + \sum_n \frac{a_{ITM,n}}{L_n} \tag{3}$$

where the first summation accounts for the specific activity a_{ETM}, the second term includes the measured DTM radionuclides and the third summation evaluates the contribution to the IRAS of ITMs.

The next section presents the uncertainty calculation of the terms given in Eq. (3).

3. Estimation of uncertainties

3.1. Uncertainty on ETM radionuclides

The major contributors to the uncertainty of a_{ETM} are the weight/density of the waste, the activity distribution within a package, the geometry of the waste items, and the relative position detector/package. A reasonable sorting, based for example on dose rate ranges, helps to limit the effects of activity hotspots (Rzyski and Suarez, 1988). Compaction of high volume objects and filling with small-sized items are effective techniques to create more uniform waste packages.

We can write the specific activity of ETM radionuclides as a function of various input parameters including the net area of a peak S_{net} (given by the difference of a gross count and a background count), the weight of the sample or the waste package m, the γ emission probability I_γ, the counting time t, the efficiency calibration ϵ, and the decay correction factor K (Knoll, 2010; Gilmore, 2008):

$$a_{ETM} = f(S_{net}, m, I_\gamma, t, \epsilon, K) = \frac{S_{net}}{m I_\gamma t \epsilon K}. \tag{4}$$

In this article we limit the discussion to the elements that are of interest for VLLW radiological characterization. A rigorous treatment of uncertainties in γ-ray spectrometry can be found in Lépy et al. (2015). Complementary information can be found in Sima and Arnold (2009).

If a_{ETM} is estimated from a single peak without interferences (we use the hat notation for estimated quantities; in this case the estimate of the activity of the ETM is represented by $\hat{a}_{ETM}$), a simplified formulation of the relative combined standard uncertainty of $\hat{a}_{ETM}$ (or relative standard deviation), indicated with $u_c(\hat{a}_{ETM})/\hat{a}_{ETM}$, is given by Canberra (2009):

$$\frac{u_c(\hat{a}_{ETM})}{\hat{a}_{ETM}} \approx \sqrt{\left(\frac{u(S_{net})}{S_{net}}\right)^2 + \left(\frac{u(m)}{m}\right)^2 + \left(\frac{u(I_\gamma)}{I_\gamma}\right)^2 + \left(\frac{u(t)}{t}\right)^2 + \left(\frac{u(\epsilon)}{\epsilon}\right)^2 + \left(\frac{u(K)}{K}\right)^2}. \tag{5}$$

Eq. (5) is simplified because assumes independence between the input variables but also because real spectra often present multiple interfering peaks from which the activity is evaluated using numerical methods. A complete formulation of the uncertainty associated to the activity of γ-emitters, including the evaluation of the covariance terms, can be found in Lépy et al. (2015).

The net counts under the peak, the weight or the counting time in Eq. (5) can be evaluated following classical uncertainty propagation as showed by GUM (JCGM 100, 2008). However, the uncertainty of the efficiency term can be difficult to estimate when characterizing legacy radioactive waste. This is mainly due to the unknown activity and weight distributions within a waste package.

At CERN, in the absence of more precise information, multiple measurements are performed on different sides of the waste package assuming uniform activity/weight distributions. Specific tests would be required to validate the assumption of uniformity and to quantify the extent of the bias affecting the efficiency calibration function. At the same time, operational uniformity checks have been implemented to limit the impact of possible heterogeneities in activity and weight distributions. We can cite for example dose rate screening and sorting of waste, mixing, cutting and compaction of waste items before the packaging and selection of radioactive waste with similar radiological characteristics whenever possible.

When multiple measurements are performed on a waste package, the specific γ activity of an ETM radionuclide is computed as the weighted average activity obtained from multiple measurements. The standard deviation of the average ETM activity $u(\langle a_{ETM} \rangle)$ obtained from n measurements is given by:

$$\frac{1}{u^2(\langle a_{ETM} \rangle)} = \sum_{i=1}^{n} \frac{1}{u_i^2(\hat{a}_{ETM})} \tag{6}$$

where $u_i(\hat{a}_{ETM})$ is the standard deviation of the ith measured activity as calculated from Eq. (5).

It should be noted that stochastic variations of the efficiency calibration function can be generated by the geometrical settings of the system package/detector. The stochastic variations are due to the uncertainties of the package/detector distance, the apparent density of the waste or the height of the waste within its package. To take these effects into account, we generated multiple efficiency calibration functions by making the input parameters vary within given intervals. Nevertheless, such stochastic variations play a minor role compared with the relative uncertainties affecting the total efficiency (5–10% according to Lépy et al., 2015).

The extreme case of a highly heterogeneous waste package would require dedicated simulations. However, such case is highly unlikely for legacy metallic waste at CERN where the application of particularly unfavourable activity and weight distributions would lead to an unjustified overestimation of the real activity of a waste package.