A New Approach to Estimate Uncertainty in Waste Characterization

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INTRODUCTION

The present summary introduces a new 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 [1]. 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 [2] [3]. 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.

We selected weighted linear models for studying the relationship between ETM and measured DTM radionuclides. The use of the so-called bootstrap [4] is described when calculating average specific activities of DTM radionuclides and their distributions. This last technique is also useful when a limited number of samples is available or their collection is made following a non-probabilistic model. Bootstrap can furthermore be used for bias estimation.

We conclude by presenting a scheme to estimate the total uncertainty of the waste characterization process.

ACCEPTANCE AND HAZARD FACTORS

CERN eliminates its radioactive waste in the final repositories made available by the two Host States (France and Switzerland), in compliance with the Tripartite Agreement [5]. Low-level radioactive waste 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:

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

where a_i is the specific activity of the radionuclide i (in Bq/g) and L_i is the limit of the radionuclide i defined as $L_i = 10^{Class_i}$. The class of a radionuclide gives information on its radiotoxicity and varies between 0 and 3 [6].

Waste is accepted at the final repository if the IRAS of each package is below 10 and the 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 and M_j is its weight (in kg).

We introduce here various techniques to calculate the IRAS and its uncertainty. Eq. (1) is 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 calculated DTM radionuclides, defined as Impossible-to-Measure radionuclides (ITM) [1].

The random uncertainty of a_{ETM} is calculated propagating the uncertainties on the net area of the peaks (S_{net}), the weight of the sample or the waste package (m), the counting time (t), the γ emission probability (I_γ) and the efficiency calibration (ϵ). Dedicated simulations must be performed if the activity distribution in a package is not uniform.

The term a_{DTM} can be estimated using linear models, the so-called Scaling Factor (SF) method or the Mean Activity Method [7]. Finally, a_{ITM} is evaluated using calculation and simulation codes.

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

ESTIMATION OF UNCERTAINTIES

Uncertainty on ETM radionuclides

The major contributors to the uncertainty of a_{ETM} are the weight/density of the waste, the activity distribution, the geometry of the waste items and the relative position detector/package. A rational sorting, based for example on dose rate ranges, helps to limit the effects of hotspots.

A simplified formulation of the random uncertainty of a_{ETM} is [8]:

$$\sigma_{a_{ETM}} = a_{ETM}\left[\left(\frac{\sigma_{S_{net}}}{S_{net}}\right)^2 + \left(\frac{\sigma_m}{m}\right)^2 + \left(\frac{\sigma_t}{t}\right)^2 + \left(\frac{\sigma_{I_\gamma}}{I_\gamma}\right)^2 + \left(\frac{\sigma_\epsilon}{\epsilon}\right)^2 + \left(\frac{\sigma_K}{K}\right)^2\right]^{1/2}. \tag{4}$$

In Eq. (4) the term K accounts for nuclide decay.

Multiple efficiency calibration functions can be generated to account for uncertainty on the size of the waste package, its density and the distance package/detector [9].

Uncertainty on DTM radionuclides

The weighted linear model

A weighted linear model can be used to predict the activity of DTMs on waste packages if a correlation exists between the

specific activity of DTM nuclides and a major ETM, called Key Nuclide (KN). The linear model is weighted to account for the uncertainties of the measurements on samples. Only results above the detection limits must be used.

In the present summary we consider only prediction intervals because they are wider and more conservative than confidence intervals.

The prediction interval of a_{DTM} for a given activity a_{KN} of the Key Nuclide is:

$$\sigma_{a_{DTM}} = s_{a_{DTM}} \sqrt{1 + \frac{1}{n} + \frac{(a_{KN} - \overline{a}_{KN})^2}{(n-1)s^2_{a_{KN}}}} \quad (5)$$

where n is the number of samples, $s_{a_{DTM}}$ is the standard deviation of the residuals of the DTMs and $s_{a_{KN}}$ is the standard deviation of the specific activity of the KN [10].

Scaling Factors and geometric mean

The Scaling Factor technique consists of checking for consistent and reproducible correlations between DTM radionuclides and a Key Nuclide. The correlation is used to estimate a_{DTM} by measuring the activity of the KN in each waste package [2] [7].

If activated under similar conditions, the activity distribution of samples is often log-normal. This is commonly observed for both γ and β emitters. The distribution of the Scaling Factors, calculated as the ratios of a_{DTM} and a_{KN}, is also log-normal.

For log-normal distributions a good estimator of central tendency is the geometric mean [2]:

$$\overline{SF} = exp\left(\frac{\sum_{i=1}^{n} \ln(SF_i)}{n}\right). \quad (6)$$

The uncertainty of the geometric mean is calculated from the geometric standard deviation D (called dispersion) [7]:

$$D = exp\left(\sqrt{\frac{\sum_{i=1}^{n} [\ln(SF_i) - \ln(\overline{SF})]^2}{n-1}}\right). \quad (7)$$

Finally, the uncertainty $\sigma_{a_{DTM}}$ of the activity of the DTM in the package i ($a_{DTM,i} = \overline{SF} \times a_{KN,i}$) is calculated propagating $\sigma_{a_{ETM}}$ from Eq. (4) and D from Eq. (7). As for the linear model, only values above the detection limits must be used.

Mean Activity Method and the bootstrap

The so called Mean Activity Method is a technique to calculate the specific activity of Difficult-to-Measure radionuclides if DTMs and the Key Nuclide are not correlated. This technique consists of calculating the arithmetic average of sample's activity for each DTM. The calculation includes values below the detection limit.

Many algorithms exists when dealing with values below the detection limit [11]. In this summary we introduce the use of the bootstrap [4] to calculate the standard error of the mean activity. The bootstrap, which is a re-sampling technique with replacement, is also used to estimate bias.

To show this method we consider a sample of 87 items of activated Copper. The Ni-63 was measured via radiochemical methods. 64 values are above the detection limit. Tab. I shows summary statistics of the sample.

Statistics	Value (Bq/g)
Minimum	0.08
1^{st} quartile	0.27
Median	0.50
Geometric Mean	0.42
Dispersion	1.99
Mean	0.53
Standard deviation	0.37
3^{rd} quartile	0.68
Maximum	2.31
Bootstrapped mean	0.53
Standard error of bootstrap	0.04
Bias	$6{\cdot}10^{-4}$

TABLE I. Summary statistics of Ni-63 in a sample of Copper activated at CERN.

We re-sampled 999 times the 87 values of Ni-63 activity. The result obtained can be used either to estimate the population mean or to estimate the bias of the sample mean. If the bootstraped mean is used, the average activity of Ni-63 in the waste population is 0.53 Bq/g. The standard error of the mean is 0.04 Bq/g. As can be inferred from Tab. I, this result is identical to the sample standard error. If we want to use the experimental mean, the calculation of the bootstrap gives an estimation of the bias, which is $6{\cdot}10^{-4}$.

We can observe that the bias is very low. This was expected because the original sample was in fact a census. The uncertainty of the sampling strategy is not considered in this example. Fig. 1 shows the histogram and the normal q-q plot of the bootstrapped mean.

Uncertainty on ITM radionuclides

The specific activity of ITM radionuclides and their uncertainty are estimated by randomly extracting a subsample from ~2.35 million CERN activation scenarios.

Using the results of the simulations we have identified the significant radionuclides produced by activation of 43 different materials, together with the ranges of the so called Correlation Factors (CF). CFs are similar to Scaling Factors but they are estimated from simulations [3].

The input space of the simulations includes the chemical composition of the materials, the energy of the accelerators (from 160 MeV of Linac 4 up to 7 TeV of the Large Hadron Collider), the location inside the tunnels, the irradiation time and the decay time.

After the random extraction of a subsample from the database, the distribution of the correlation between Difficult-to-Measure and Key Nuclides is checked and Correlation Factors are calculated. Depending on the distribution found the appropriate central tendency estimator is calculated. Linear models can also be used and, as for the Scaling Factors, the activity uncertainty can be estimated using prediction intervals.

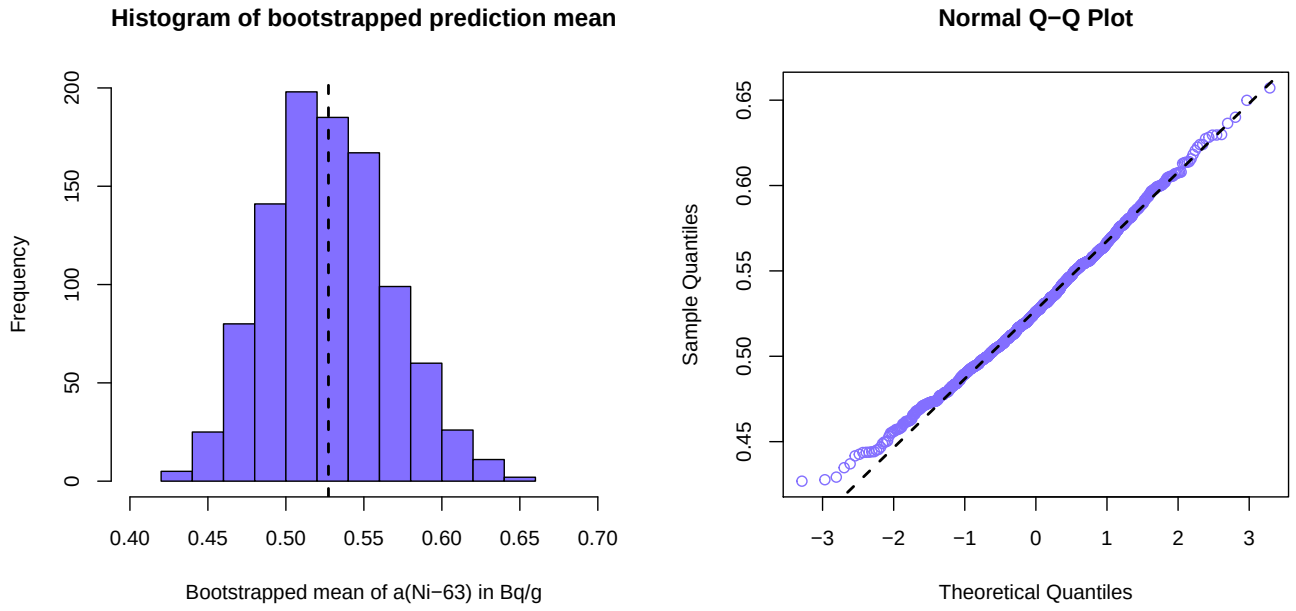

Fig. 1. Distribution of bootstrapped mean (left) and normal q-q plot (right) for a(Ni-63) in the Copper population. The dashed lines indicate the bootstrapped mean (left) and the theoretical normal line (right).

CONCLUSIONS

This summary presents a procedure to calculate the uncertainty of the main terms of the hazard factor called IRAS, which is needed to evaluate the acceptance of radioactive waste by the French repository.

The uncertainties of the activities are calculated either by statistical methods, such as weighted linear models, or by intensive calculation techniques such as the bootstrap.

If $\sigma_{a_{ETM}}$ represents the uncertainty of the γ activity, $\sigma_{a_{DTM}}$ the uncertainty of the Difficult-to-Measure radionuclides and $\sigma_{a_{ITM}}$ the uncertainty of the simulated or calculated radionuclides, we can express the random uncertainty of the IRAS as follows:

$$\sigma_{IRAS} = \left[\sum_l \left(\frac{1}{L_l} \right)^2 \sigma^2_{a_{ETM,l}} + \sum_m \left(\frac{1}{L_m} \right)^2 \sigma^2_{a_{DTM,m}} + \sum_n \left(\frac{1}{L_n} \right)^2 \sigma^2_{a_{ITM,n}} \right]^{1/2}. \quad (8)$$

To calculate the overall uncertainty U of the hazard factor, the bias must be estimated. We show how bootstrap is used to estimate the bias in the case of the Mean Activity Method. If the total bias is evaluated, the overall uncertainty is finally:

$$U = \sigma^2_{IRAS} + \text{bias}^2. \quad (9)$$

The waste producer can use conservative values to estimate specific activities. Such an assumption includes replacing the specific activity (measured or calculated) with its upper bound, the third quartile or collecting samples at the hotspot. The bias term in Eq. (9) increases when such a choice is made and the specific activity is overestimated. High-biased, conservative approaches are useful to identify envelope estimators of the hazard factors and can be employed if the uncertainty of the IRAS cannot be calculated.

In all other situations the uncertainty of the IRAS must be estimated and the present article outlines a new procedure for its evaluation.

REFERENCES

1. IAEA, “Strategy and methodology for radioactive waste characterization,” Tech. Rep. IAEA-TECDOC-1537, Vienna (2007).
2. ISO, *ISO 21238. Nuclear energy - Nuclear fuel technology - Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants*, Geneva (2007).
3. ISO, *ISO 16966. Nuclear energy - Nuclear fuel technology - Theoretical activation calculation method to evaluate the radioactivity of activated waste generated at nuclear reactors*, Geneva (2013).
4. B. EFRON and R. TIBSHIRANI, *An introduction to the bootstrap*, Chapman and Hall Inc., London (1993).
5. CONSEIL-FÉDÉRAL-SUISSE, “Accord entre le Conseil Fédéral Suisse, le Gouvernement de la République Française, et l’Organisation Européenne pour la Recherche Nucléaire relatif à la Protection contre les rayonnements ionisants et à la Sûreté des Installations de l’Organisation Européenne pour la Recherche Nucléaire,”

Tech. Rep. 0.841.592.2 (2011).
6. ANDRA, "Critères radiologiques d'acceptation des déchets TFA," Tech. Rep. SUR.SP.AMES.02.0007, Chatenay-Malabry (2013).
7. IAEA, "Determination and use of scaling factors for waste characterization in nuclear power plants," Tech. Rep. NW-T-1.18, Vienna (2009).
8. CANBERRA, "Customization tools manual," Tech. Rep. Genie 2000 v.3.2, USA (2009).
9. CANBERRA, "Technical advantages of ISOCS/LabSOCS," Tech. rep., USA (2012).
10. S. ROSS, *Introduction to probability and statistics for engineers and scientists*, Elsevier Academic Press, Burlington (2009).
11. C. CROGHAN and P. EGEGHY, "Methods of dealing with values below the limit of detection using SAS," *Presented at the Southeast SAS User Group*, pp. 22–24 (2003).