Evaluation of Uncertainties of Radiological Characteristics by Means of a Mix of Deterministic and Probabilistic Approaches – 11025

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ABSTRACT

The evaluation of uncertainties of radiological characteristics of low and intermediate level waste is a challenging issue for the global radioactive waste management community. One of the goals of ONDRAF/NIRAS as Belgian waste management agency is to propose a strategy that is applicable for all Belgian radioactive waste producers. In 2008, a statistical methodology was developed and motivated. The basis of the methodology was the lognormal distribution, which is used often in radiation protection. In theory, the proposed distribution function is applicable for low and intermediate level short lived radioactive waste. However, the application in practice was hampered by the use of incompatible characterization methodologies and different terminologies by waste producers. In addition, the method was not compatible with the applicable waste acceptance criteria. The uncertainty approach is in full development. One of the possible scenarios is discussed in this paper. For the evaluation of uncertainties on the scale of a waste package, a stepwise approach is used (process mapping >> evaluation of analysis model >> distribution functions for parameters and activity levels >> distribution functions for related quantities). The goal of waste acceptance is verifying that the distributions of activity levels respect the radiological conformity criteria for the intended waste disposal solution. For surface disposal, small scale intrusion scenarios could be used to determine dose conversion factors to be used in a probabilistic approach. Exploitation of the surface disposal facility includes following up the operations in order to guarantee that the radiological capacity of the disposal facility and its modules are respected. The radiological capacities could be determined by a reference scenario and large scale intrusion scenarios.

INTRODUCTION

Management of uncertainties related to radiological characteristics of low and intermediate level waste is a challenging issue for the global radioactive waste management community. In Belgium, the Royal Decree of November 18, 2002 on the qualification of equipment for storage, processing and conditioning of radioactive waste [1] expresses a legal requirement to determine the uncertainties in a reproducible way. One of the goals of ONDRAF/NIRAS as Belgian waste management agency is to propose a strategy that is applicable for all Belgian radioactive waste producers.

The search for an uncertainty approach was launched at a bilateral meeting between ONDRAF/NIRAS and ANDRA on February 26, 2006, during which ANDRA presented its deterministic uncertainty approach. After this meeting, ONDRAF/NIRAS developed a framework based on the lognormal distribution function [2]. This framework was discussed internally and externally, which lead to a solid proposal in June 2008 [3].

The advantage of a probabilistic approach is that there is no need to pile up conservative hypotheses leading to hyper-conservative declaration values. The inconvenience of a probabilistic approach is that it is generally much more complex than a deterministic approach. This is why ONDRAF/NIRAS searched for a probabilistic framework with a mathematically fully developed work plan [3]. This framework was supported by a number of examples from current practice.

Consultation of some major waste producers brought certain issues to the surface, which hampered the direct application of the lognormal method. In addition, the uncertainty approach for radiological characterization, the waste acceptance criteria and the management system of the waste disposal site have to be compatible. Briefly, the problem of uncertainties needs to be approached in a broader context.

THE 2008 PROPOSAL

In 2008, a probabilistic methodology was developed and motivated. The basis of the methodology was the lognormal distribution function, which is used often in radiation protection. In theory, the proposed distribution function is applicable for low and intermediate level short lived radioactive waste. However, the application in practice is hampered by the use of incompatible characterization methodologies and different terminologies.

The lognormal distribution

The use of the lognormal distribution function is supported by the fact that radionuclide activities are always strictly positive values [3]. The bulk of the values in a batch of similar packages are usually concentrated in the very low values, with long tails (up to multiple decades). As the high values have a strong influence on the mean value, often strong asymmetries are observed as well as discrepancies between mean, median and modus. These observations logically suggest the lognormal distribution as a candidate to represent a population of radioactive waste packages.

Radioactive decay is an intrinsically multiplicative process and therefore the theory of successive random dilution would be applicable, predicting a lognormal distribution function by application of the central limit theorem. Daniels and Higgins [4] show that the lognormal distribution is indeed a natural consequence of the phenomena and processes that lead to radioactive substances in the environment, or at least that the lognormal distribution function gives a good approximation.

The most appropriate representation of the lognormal distribution function in the framework of radioactive waste characterization is based on the median (M) and the geometric standard deviation (σ_G) of the distribution. The probability density function of a lognormal distribution function then takes the form of equation 1.

$$P(y < X/M < y+dy) = \frac{1}{\sqrt{2\pi}\cdot y\cdot \ln(\sigma_G)}\cdot \exp\left(\frac{-\ln(y)^2}{2\cdot \ln(\sigma_G)^2}\right)\cdot dy \qquad \text{(eq. 1)}$$

Envelope values for the geometric standard deviation

Although many different sources of uncertainty can appear, depending on the treatment and conditioning processes, the non destructive measurements, the samples analyzed in laboratories, and the materials used, 3 overarching types of uncertainty have been identified: representativity of a measurement or analysis, confidence in the radionuclide vector and measurement uncertainty.

Table I illustrates envelope values for the geometric standard deviation linked to the representativity of a gamma measurement. These values were developed by means of a numerical experiment simulating variations in density and source distribution [3].

Table I – Envelope values for the geometric standard deviation (GSD) in function of the apparent density

Apparent density (D)	GSD
$D < 1\ g/cm^3$	2.7
$1\ g/cm^3 \geq D > 3\ g/cm^3$	7.4
$D \geq 3\ g/cm^3$	20

Table II illustrates envelope values for the geometric standard deviation linked to the confidence in the radionuclide vector. These values were derived from the statistical analysis of the source data from some major Belgian radioactive waste producers [3].

Table II – Envelope values for the geometric standard deviation (GSD) in function of the key radionuclides

Target radionuclide / Key radionuclide	Co-60	Cs-137	Pu-239/240
Activation products	1.3	2.7	
Fission products		7.4	
Actinides		20	7.4

The measurement uncertainty is linked to the measurement result and declared as such. In general, the measurement uncertainties are smaller than the uncertainties linked to representativity of the measurement and to the confidence in the radionuclide vector. Only near the edge of the measurement range (detection limit) the measurement uncertainty may contribute significantly to the global uncertainty of the declared activity.

What went wrong?

Consultation of the major Belgian waste producers clarified that the practical application of the proposed solution would be hampered by the following issues:

- Different methods and terms for radiological characterization;
- Specific aspects of waste or characterization method not yet taken into account;
- Not compatible with current waste acceptance criteria.

In addition, through discussions with the regulatory body ONDRAF/NIRAS realized that different actors have different views on matters such as uncertainties. While waste producers may be confident in their characterization effort, the regulatory body could take a more skeptical position regarding the same results. As uncertainty margins are subjective estimations of the confidence in the characterization results, it is not possible to prove either one wrong.

The role of ONDRAF/NIRAS is to take a neutral position. From this position, ONDRAF/NIRAS can guide the waste producers in providing the picture as complete as possible and defend this picture and the subsequent results towards the regulatory body.

BROADENING THE PERSPECTIVE

The current strategy of ONDRAF/NIRAS is to study the problem of uncertainties with a broader perspective. This strategy is based on two general observations:

- Different scales (waste packages and waste streams) imply different sources of uncertainty and different approaches;
- Every waste characterization methodology is unique;
- The discussion of uncertainty margins is an integral part of the radiological characterization;
- .Declarations have to be compatible with criteria for waste acceptance an disposal.

Radiological characterization of waste packages is important for the safe processing and storage of these waste packages. A variety of measurement and calculation methods can be applied to estimate the activity levels of radionuclides enclosed in a waste package, such as:

- Gamma spectrometry, neutron counting or dose rate measurement;
- Measurements on samples, non-conditioned waste packages or conditioned waste packages;
- Mathematical modeling of physical, chemical and radiological processes.

A stepwise approach is proposed for assessing the uncertainties related to the radiological characterization of waste packages. This approach enables us:

- To review the entire characterization process with a special focus on the discrepancies between the physical, chemical or radiological processes and the modeling efforts to describe these processes mathematically;
- To identify the major sources of uncertainty and quantify their possible impact on the results;
- To justify the distribution functions used in a probabilistic approach.

Waste acceptance includes verifying if the radiological conformity criteria are respected. These conformity criteria will be drawn up in line with the guidance on human intrusion provided by the Federal Agency for Nuclear Control. The guidance on human intrusions implies two probabilistic dose criteria. A distribution function is determined for the dose impact expected from a small scale intrusion scenario. For all accepted waste packages for surface disposal, the distribution functions of the expected dose must comply with the two probabilistic dose criteria.

During exploitation of the disposal facility, the evolution of the waste volume and the radiological charge are meticulously followed in time. This guarantees that the radiological capacity of the disposal facility is not exceeded and that the activity is evenly distributed over the disposal modules. The follow-up of the radiological charge of the disposal facility and its modules could be done in a deterministic way, based on the expectation values of the activity levels of the waste packages compared to the radiological capacity of the facility, which can be determined by means of a reference scenario and large scale intrusion scenarios.

CHARACTERIZATION OF A WASTE PACKAGE

The stepwise approach

A general view on radiological characterization of a waste package is presented in Figure 1. This figure illustrates the concepts of reality and perception, the differentiation of which is essential for the understanding of uncertainties.

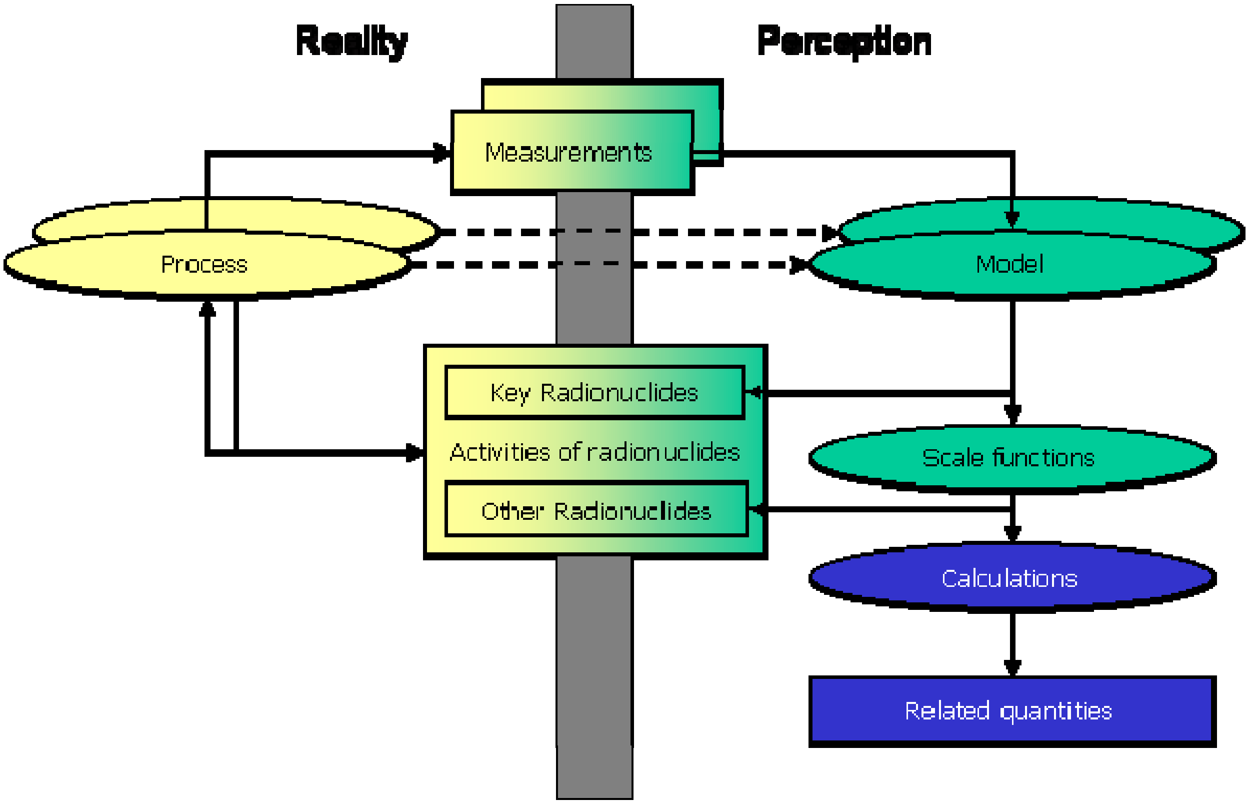

Figure 1 – General view on radiological characterization

A stepwise approach could be used for the evaluation of uncertainties on the scale of a waste package (See Figure 2). The aim of this approach is to review the waste characterization methodology with a particular focus on uncertainty management. First, a mapping of all the processes that are relevant to the waste characterization is done, for example processes related to a measurement technique or processes related to the production and migration of radionuclides. This mapping is fundamental for the assessment of the hypotheses and approximations of the mathematical and computer models and of their possible impact on the characterization results. Subsequently, the probabilistic approach also requires the choice of a suitable probabilistic calculation method and the definition of the relevant distribution functions.

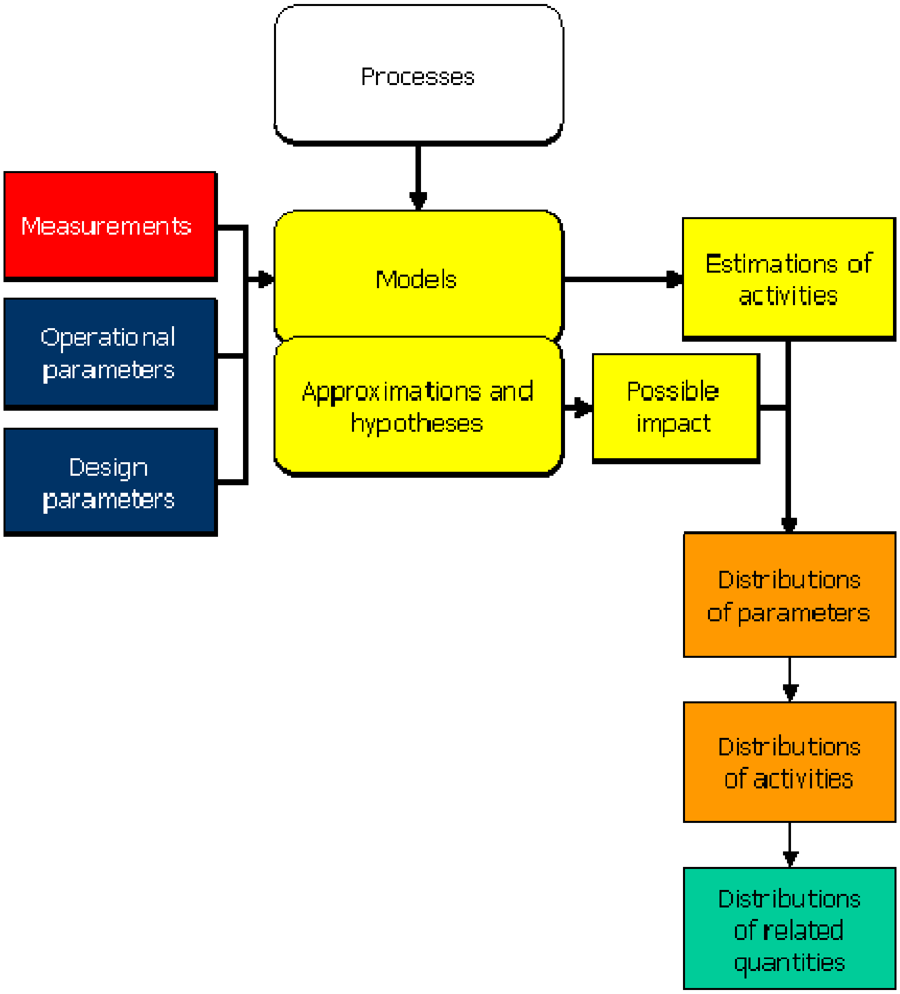

Figure 2 – Stepwise approach for uncertainties

The different steps of the uncertainty approach will be illustrated by means of an example. Gamma spectrometry of a complete waste package has been singled out for this purpose as a widely used measurement technology that often serves as the basis for a radiological characterization method.

Step 1: Process mapping

Measurements give an image of the activity levels of radionuclides present in de waste packages. In order to execute a reliable radiological characterization based on these measurements it is best to map the whole measurement process. This encompasses an accurate description of the physical, chemical, radiological and operational processes involved. It is essential that all events, conditions and parameters that separate the radioactivity and the measurements are treated.

Figure 3 illustrates this process mapping for the selected example of gamma spectrometry of a complete waste package. The identified processes deal with the radionuclides, the materials in the waste package, the measurement geometry and the measurement system. The measurement result of gamma spectrometry is the spectrum.

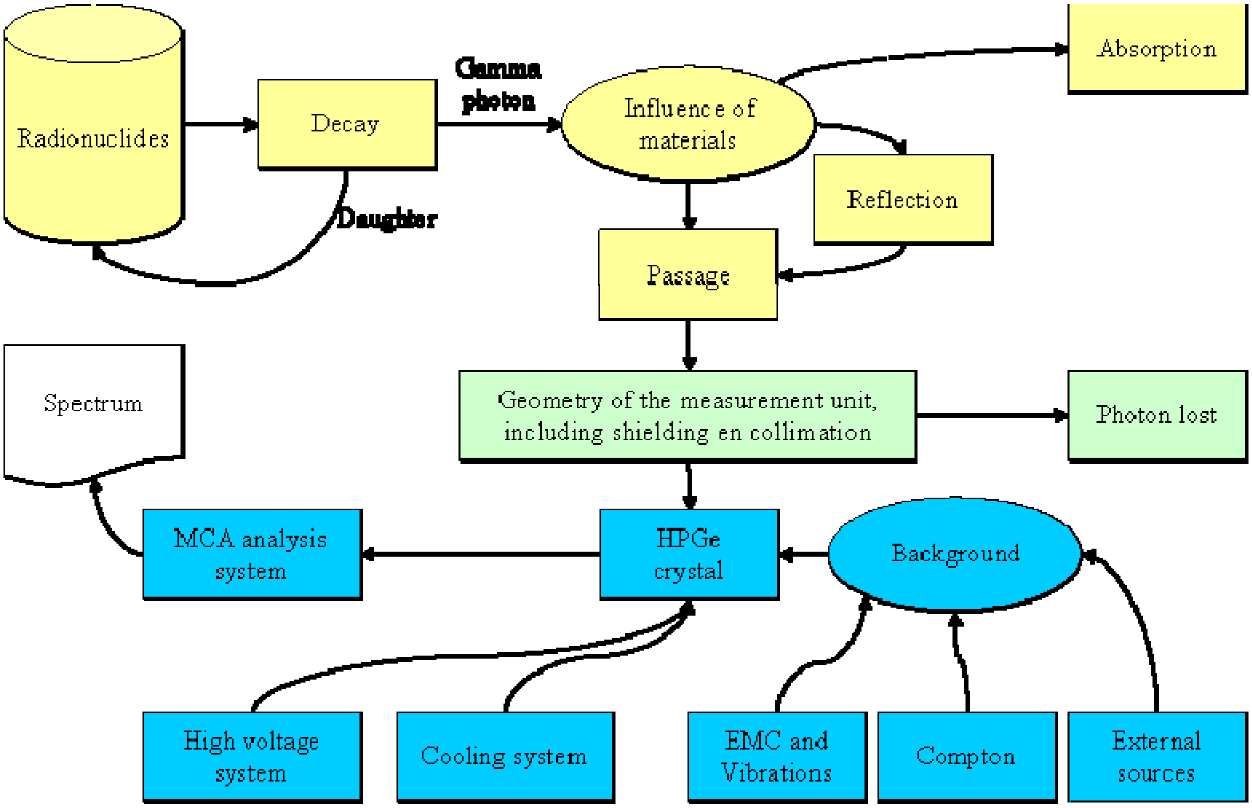

Figure 3 – Process mapping of gamma spectrometry

Sometimes, there are no measurements available, for example if the object of the radiological characterization is future production or if a manual measurement encompasses too large a radiological risk. In that case the radiological characterization is based on the understanding of radionuclide production and migration processes, which are then described as good as possible.

The understanding of radionuclide production and migration processes often also forms the basis for the development of a radionuclide vector that is used to calculate scaling factors to determine radionuclides that are difficult or impossible to measure.

Step 2: Model assessment

In order to transform the processes described in step 1 into a workable mathematical model it is necessary to make some simplifications and hypotheses. Simplifications imply known elements that are too complex to implement or that would lead to specific models on too small a scale, hypotheses indicate unknown or uncertain elements for which a choice has to be made.

Uncertainties related to the radiological characteristics are mainly due to differences between the real processes and the mathematical models that represent these processes. Figure 4 illustrates how a gamma spectrum is analyzed to retrieve the radionuclide activities.

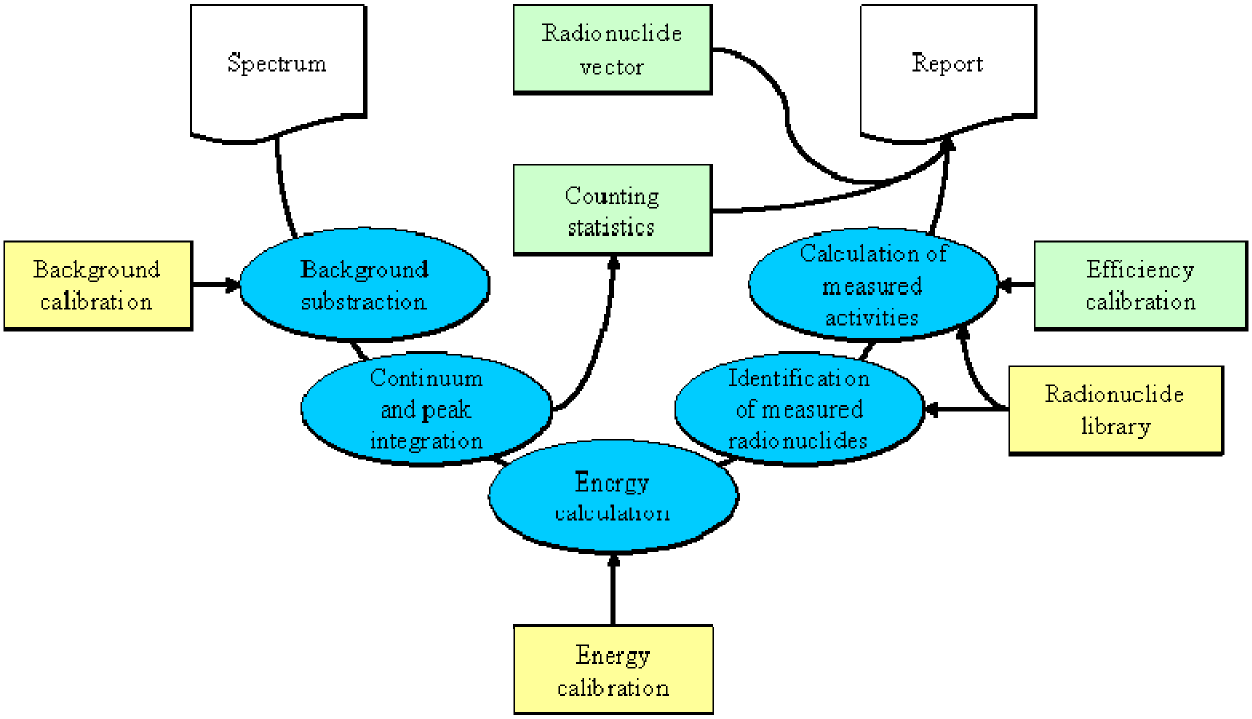

Figure 4 – Gamma spectrometry analysis

Table III shows for a characterization method based on gamma spectrometry how the different processes influence the analysis model. The column headings nominate the different elements of the analysis model. The row titles represent the different processes linked to gamma spectrometry. If a process influences an analysis model element, the corresponding field is marked. If in addition this process encompasses a major source of uncertainty the corresponding field is numbered in Table III.

1. Measurement results:

   The uncertainty margins of the measurement results are derived from the interpretation of counting statistics. These uncertainty margins are based on the Poisson distribution and propagate to the integration of the spectrum. In general, this is done by the analysis software that is delivered with the measurement equipment.

2. Efficiency calibration:

   Discrepancy between reality (heterogeneous density distribution, heterogeneous source distribution) and the analysis model (homogeneous density distribution, homogeneous source distribution) lead to the uncertainties margins of the efficiency calibration. Usually, the analysis software does not consider uncertainty margins for the efficiency of the measurement system.

3. Difficult to measure radionuclides:

   The major source of uncertainty for difficult to measure radionuclides is the representativity of the radionuclide vector. In addition, the method to determine the scaling factors that form the radionuclide vector is subject of a proper uncertainty analysis.

Table III – Mapping processes on analysis model elements for gamma spectrometry

Elements of the analysis model	Measurement result (spectrum)	Background	Integration of the spectrum	Energy calibration	Radionuclide library	Efficiency calibration			Difficult to measure radionuclides
Options						Std configs	Transmission	Model	
The package contains radionuclides					X				X
Radionuclides decay to daughters					X				X
The decay of radionuclides generates gamma radiation (or not)					X				3
Gamma radiation is influenced by the presence of materials						2	2	2	
Absorption						2	2	2	
Reflection						2	2	2	
Geometry of the package to be measured						2	2	2	

Elements of the analysis model	Measurement result (spectrum)	Background	Integration of the spectrum	Energy calibration	Radionuclide library	Efficiency calibration			Difficult to measure radionuclides
Options						Std configs	Transmission	Model	
Geometry van the HPGe crystal						X	X	X	
Position of the HPGe crystal related to the package to be measured						X	X	X	
Dimensions of the HPGe crystal						X	X	X	
Collimator						X	X	X	
Shielding						X	X	X	
Reflection on the walls of the measurement cell						X	X	X	
Properties of the detector			X	X				X	
Energy resolution and range			X	X					
Detection efficiency			X					X	
Dead time			X					X	
Influence of the working voltage									
Influence of the temperature and humidity in the measurement cell									
Influence of the temperature of the crystal		X	X	X					
Background / continuum		X							
Natural background		X							
Compton effect		X							
Other sources in the vicinity		X							
EMC interference		X							
Vibrations		X							
Counting statistics	1		1						
Other elements									X

Step 3: Distributions of parameters and activity levels

Parameters or quantities that are influenced by major sources of uncertainty are attributed a distribution function. The distribution function can be defined in one of the following ways:

- Functional form of the cumulative distribution function or the probability density function and the corresponding parameters;
- A number of discrete percentiles, approximating the cumulative distribution function;
- A histogram, approximating the probability density function;
- A graphical representation of the cumulative distribution function or the probability density function, including the basic data used.

In [3] is shown that by means of numerical and experimental techniques the relevant parameters for gamma spectrometry can be attributed lognormal distributions.

Sources of uncertainty whose impact is minimal compared to major sources of uncertainty can be treated in a deterministic way. This means that the corresponding parameters are attributed a single value without uncertainty margin or distribution function.

In case of very low activity levels, the quantity to be measured is not found in the range of the measurement equipment. A cautious or conservative approach is to determine the declaration values from the detection limit. In general, a detection limit is treated as *unknown value that is lower than or equal to the detection limit*. This means that in a probabilistic analysis there is no information available for the range below the detection limit, and that the probability density function in this region is constant. In this case, the conservative activity limits can be determined by means of a deterministic approach.

The activity levels of the critical radionuclides are calculated using a probabilistic approach. Either they are determined by means of approximate calculation rules if the chosen distributions allow this (e.g. in case of normal distributions, lognormal distributions or constant distributions), or a Monte Carlo simulation is used.

Step 4: Distribution functions of related quantities

For a large number of waste fluxes, the lognormal distribution is considered a candidate for the distribution function of the activity levels of critical radionuclides. The position, shape and width of this distribution function are determined by the median and the geometric standard deviation. A distinguished property of the lognormal distribution is its asymmetric shape in the positive domain. The advantage of using the lognormal distribution for determining the distribution functions of related parameters is the availability of simplified calculation rules [3].

Waste producers may use different distribution functions if this is motivated in a qualitative or quantitative way. However, in this case the probabilistic calculations usually have to be based on Monte Carlo simulation techniques.

UNCERTAINTIES AND SURFACE DISPOSAL OF RADIOACTIVE WASTE

Interpretation of uncertainties of radiological characteristics during acceptance of waste for surface disposal

Small scale intrusion scenarios describe the consequences of inadvertent human intrusions after regulatory control has been lifted, whose implications are determined by the radiological contents of one single waste package. It is impossible to model all future human activities, therefore

- Stylized scenarios with a conservative nature should be proposed;
- Conservative parameters and hypotheses based on current knowledge should be used;
- A deterministic approach would be suitable for the calculations.

The selected small scale intrusion scenarios would lead to the calculation of dose conversion factors (in mSv per MBq/m^3 or in mSv/year per MBq/m^3) for the critical radionuclides.

Radiological conformity criteria based on the guidance on small scale intrusions could be imposed as a prerequisite for acceptance [5]. This would imply that for each waste package the distribution function of dose impact of the small scale intrusion scenarios would be weighted against the probabilistic conformity criteria expressed in equation 2.

$$Z = \frac{P95[Dose]}{3mSv/yr} < 1 \qquad \text{(eq. 2)}$$
$$Z' = \frac{P99.99[Dose]}{100mSv/yr} < 1$$

In equation 2, *P*95[*Dose*] and *P*99.99[*Dose*] represent the 95th and 99.99th percentiles of the distribution function of the dose impact $\mathcal{D}[Dose]$ due to a small scale intrusion scenario, defined by equation 3.

$$\mathcal{D}[Dose] = \sum_{\substack{\text{critical}\\ \text{radionuclides}}} \mathcal{D}[C_i] \cdot DCF_i \qquad \text{(eq. 3)}$$

The distribution function of the dose impact would be determined by the distribution functions of the activity concentrations of the critical radionuclides $\mathcal{D}[C_i]$ and the corresponding dose conversion factors DCF_i for the considered intrusion scenarios. If the activity concentrations of a waste package are declared as lognormal distribution functions, the approximate calculation rules from [3] could be used. Otherwise Monte Carlo simulations seem to be necessary. In both cases it seems useful to consider the correlations between related radionuclides.

Interpretation of uncertainties of radiological characteristics during exploitation of the surface disposal facility

The reference scenario describes the natural evolution of the disposal facility in the case that after the regulatory control is lifted no human intrusions take place. This scenario would be used to evaluate the dose uptake by a critical individual of the general public and hence to limit the radiological capacity (in MBq) of the disposal facility for the critical radionuclides.

Large scale intrusion scenarios describe the radiological consequences of human intrusions after the regulatory control has been lifted, whose impact is determined by many waste packages [5]. These scenarios would be used to determine the radiological capacity (in MBq) of the individual modules for the critical radionuclides, taking into account the maximum allowed ratio capacity module / capacity facility.

A filling strategy is developed according to the current knowledge of the global waste characteristics. This filling strategy includes:

- The expected evolution of the waste volume;
- The expected evolution of the radiological charge compared to the radiological capacity of the facility.

During exploitation of the surface disposal facility, the consumption of radiological capacity of the facility and its modules would be followed in real-time, in order to guarantee that the radiological capacity is respected and that the activity is distributed evenly over the modules. An indicator for consumption of radiological capacity, Y', which could be defined like equation 4, must remain below one at all times.

$$Y' = \sum_{\substack{\text{disposal} \\ \text{facility}}} \sum_{\substack{\text{critical} \\ \text{radionuclides}}} \frac{E[A_{i,j}]}{A_{max,i}} \qquad \text{(eq. 4)}$$

In equation 4, $E[A_{i,j}]$ is the expectation value of the activity level of radionuclide i in waste package j and $A_{max,i}$ is the radiological capacity of the disposal facility for radionuclide i. A similar indicator should be developed for the consumption of radiological capacity of the modules.

This follow-up of the exploitation of the disposal facility can also be used for the re-evaluation of the filling strategy during a future revision of the exploitation license.

Before all waste packages have been emplaced in the disposal modules, probabilistic calculations provide results that can not be interpreted in terms of safety evaluations. Therefore, deterministic estimations seem to be sufficient for good management of the disposal facility. The large quantity of waste packages justifies the use of the expectation value for these calculations.

CONCLUSIONS

The approach of ONDRAF/NIRAS for the determination and interpretation of uncertainties related to the radiological characterization of waste packages is currently in full development. One of the possible scenarios is discussed in the current paper. This scenario requires action on three levels:

- Radiological characterization of waste packages;
- Acceptance of waste packages;
- Exploitation of a waste disposal facility.

For the evaluation of uncertainties on the scale of a waste package, a stepwise approach is used. First a process mapping of the radiological characterization is done. Based on this process mapping the hypotheses and approximations of the mathematical and computer model s are assessed. The discrepancies between the reality and the model, as well ad the possible impact of the major sources of uncertainty is reflected in distribution functions for parameters and activity levels. A probabilistic calculation method is use to determine the distribution functions for related quantities.

The goal of waste acceptance is verifying that the distributions of activity levels respect the radiological conformity criteria for the intended waste disposal solution. For surface disposal, small scale intrusion scenarios could be used to determine dose conversion factors to be used in a probabilistic approach.

Exploitation of the surface disposal facility includes following up the operations in order to guarantee that the radiological capacity of the disposal facility and its modules are respected. The radiological capacities could be determined by a reference scenario and large scale intrusion scenarios. Following-up the consumption of radiological capacity could also be used to re-evaluate the filling strategy when the license is renewed.

The proposed strategy for the evaluation and interpretation of uncertainties related to radiological characterization has the following advantages:

- The stepwise approach guarantees that all sources of uncertainty have been taken into account;
- The probabilistic method for analyzing uncertainties avoids the unnecessary addition of conservative approximations, especially close to activities limits of critical radionuclides;
- The probabilistic method for determining uncertainty margins is compatible with the interpretation by means of probabilistic radiological conformity criteria;
- The large number of involved waste packages justifies the deterministic approach for the interpretation of uncertainties for managing the exploitation of the surface waste disposal facility.

The approach for the determination and interpretation of uncertainties related to radiological characterization of waste packages is an important aspect in the discussions between ONDRAF/NIRAS and the Federal Agency for Nuclear Control on the safety evaluations of the planned surface disposal facility for low and intermediate level short lived radioactive waste. A balance must be found between the feasibility of the approach and the degree of confidence in the results.

REFERENCES

[1] Royal decree of November 18, 2002 on the qualification of equipment for the storage, treatment and conditioning of radioactive waste, Belgisch Staatsblad – Moniteur Belge, pp. 54352-54355, December 3, 2002.

[2] Y. Gigase, Uncertainty Analysis of the Radiological Characteristics of Radioactive Waste Using a Method Based on Log-Normal Distributions – 7285, ICEM07, Bruges, Belgium, September 2-6, 2007.

[3] T. Soetens, Uncertainties Related to Radiological Characteristics of Radioactive Waste Packages, ONDRAF/NIRAS, NIROND-TR 2008-05 E, June 2008.

[4] W.M. Daniels and N.A. Higgins, Environmental Distributions and the Practical Utilisation of Detection Limited Environmental Measurement Data, NRPB-W13, June 2003.

[5] Federal Agency for Nuclear Control, Guide relative à la prise en compte d’intrusion humaine pour les dépôts définitifs en surface de déchets radioactifs, 007-087-F, rév. 1, April 2, 2010.