Validation study for the determination of in-depth contamination
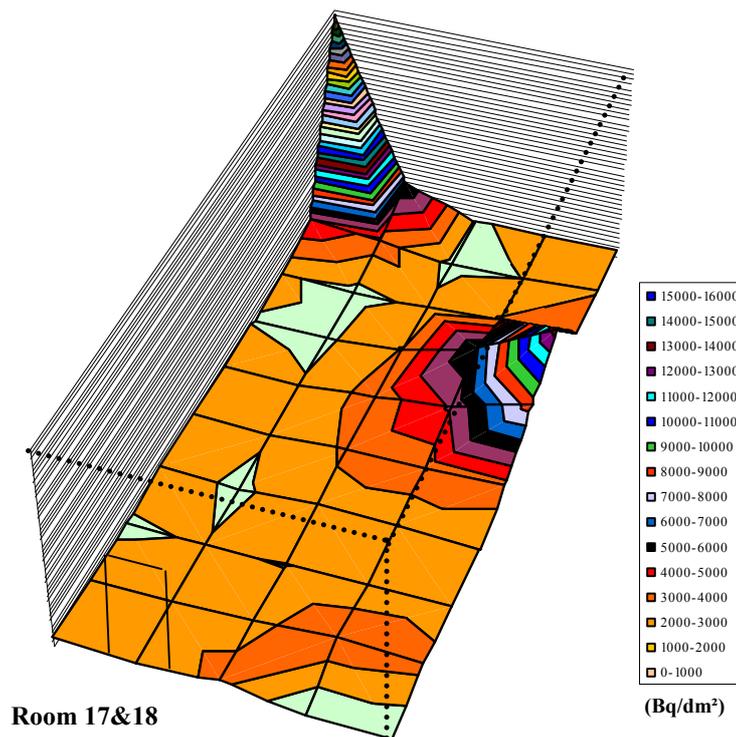
in concrete structures using in-situ gamma spectroscopy

Room 17&18

	(Bq/dm²)
15000 - 16000	
14000 - 15000	
13000 - 14000	
12000 - 13000	
11000 - 12000	
10000 - 11000	
9000 - 10000	
8000 - 9000	
7000 - 8000	
6000 - 7000	
5000 - 6000	
4000 - 5000	
3000 - 4000	
2000 - 3000	
1000 - 2000	
0 - 1000	

**Thesis for Master 2 ITDD - Sylvain LEMIUS –
September, the 24th in 2008 - Valence**

Abstract

The decommissioning of Belgian Reactor 3 (BR3), the first Pressurised Water Reactor (PWR) in Europe, approaches its final phase, in which the building structures are being decontaminated. Certain radionuclides, as Cs-137, might be present due to contamination.

The costs for radwaste are forcing the producers to minimise its amount and to invest in decontamination and characterisation methodologies.

The present document describes a validation study for the determination of in-depth contamination in concrete structures using the ISOCS (In-Situ Object Counting System), a non-destructive measurement technique. The study consisted of three main objectives:

- identification and quantification of the main sources of standard uncertainty,
- optimisation of the main parameters of standard uncertainty towards a decrease in expanded uncertainty,
- verification of this method with a destructive method.

Before this study, the expanded uncertainty seemed to largely exceed 100%. After adjusting several parameters, the expanded uncertainty is estimated at 80%. Optimising still more parameters in the near future, might lead to a reduced expanded uncertainty of about 60%.

Results obtained using the non-destructive method have been compared with the traditional destructive method based on core drilling, sample preparation and standard gamma spectroscopy. For the one spot that has been tested, results obtained from both methods were in agreement.

It might be useful to perform additional measurements using both methods. The use of in-situ gamma spectroscopy to determine the in-depth contamination in concrete structures seems to be a good alternative.

Table of contents

I.   Introduction

A prototype of Pressurised Water Reactor (PWR), the Belgian Reactor 3 (BR3), was ordered in 1957. Becoming critical in October 1962, BR3 was the first PWR in Europe, and it is also the first to be decommissioned.

Indeed according to the present planning, the plant will be completely decommissioned ("green field") by the end of 2020. After many steps, the decommissioning approaches its final phase. Today, the main objective is to deal with concrete. Because it represents the largest amount of radwaste, a good determination of contaminated part is very important.

In view of radwaste minimisation, the improvement of characterisation methods is subject to a special attention. This study is situated in this area.

A possibility to determine the in-depth contamination of Cs-137 could be in-situ gamma spectroscopy, using ISOCS (In-Situ Object Counting System). Before introducing such a method it should be examined if it is "fit for purpose". This can be done by calculation of the expanded uncertainty based on the estimation of the important individual parameters. Optimisation of single parameters leads to a decrease of expanded uncertainty.

The ultimate goal is to increase the accuracy of the measurement result in view of removing the minimal amount of building structure as radwaste.

Key words

ISOCS, Validation, Radwaste, Concrete, Unconditional release, Clearance level, In-depth contamination, Cs-137, Contamination profile, Relaxation length, Uncertainty.

II. From Belgian nuclear industry to SCK·CEN

This chapter describes the main nuclear scene in Belgium and the role of the SCK·CEN. In view of their waste management Belgium is, as many other European Union countries, applying the concept of unconditional release.

II.1 The main nuclear scene in Belgium

Figure 1 shows a simplified scheme of the main nuclear scene in Belgium in 2005 [1]. The key players in the area of nuclear power production and nuclear engineering in Belgium are **Tractebel** and **Electrabel** (SUEZ). **FBFC** (AREVA NP) makes fuel assemblies for nuclear power plants. **Belgoprocess** is specialised in the processing of radwaste. At the end of the nuclear fuel cycle, the Belgian organisation responsible for the management of interim storage and final disposal of radwaste is **NIRAS-ONDRAF**.
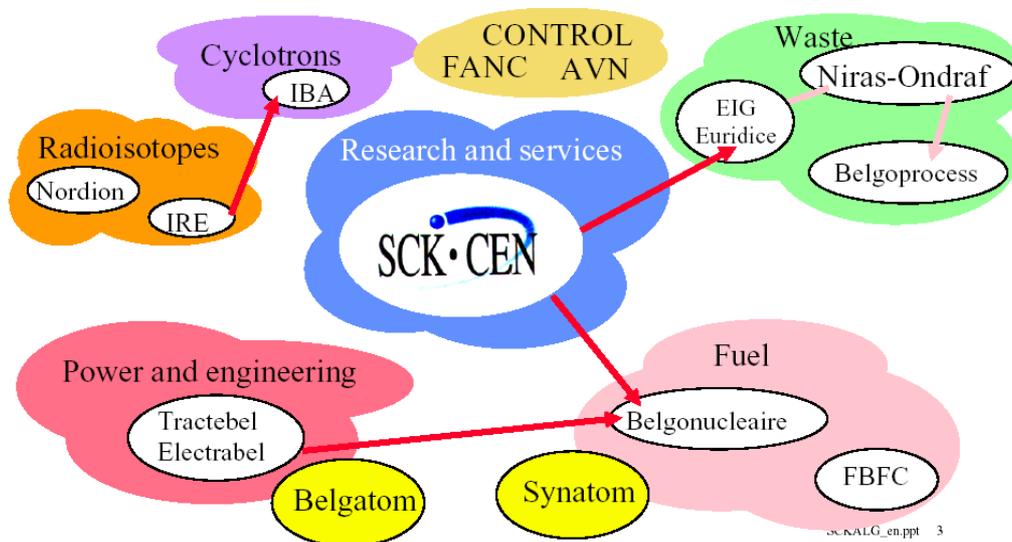

Figure 1: Simplified scheme of the main nuclear scene in Belgium in 2005

Activities of SCK·CEN concern nuclear safety, radiation protection, nuclear fuel cycle and medical and industrial applications.

II.2 Presentation of SCK·CEN

The "Studie Centrum voor Kernergie-Centre d'Etude de l'Energie Nucléaire" (SCK·CEN) in Mol was built in 1954 and 1955, after government's decision to promote on pacific and social interest applications and nuclear studies. The main objective of the SCK·CEN is to maintain a centre of excellence for research on nuclear science and technology and ionising radiation [2].

The main mission of SCK·CEN is to :

- **Conduct research with regard to safety of nuclear installations** (safe treatment and disposal of radwaste, protection of man and environment against radiation)
- **Conduct research with regard to social implications** (in the scope of the pursuit of sustainable development)
- **Develop, gather and spread the necessary knowledge through education and communication**
- **Provide all services asked by nuclear industry, medical sector and government**

Figure 2 shows an areal view of SCK·CEN near the city Mol.

Figure 2: View of SCK•CEN

Today, SCK•CEN employs 600 highly qualified people, and has a total annual budget of 75 M€. It is one of the largest non-university research organisation in Belgium. Since 2005, SCK•CEN has become a foundation of public utility [2].

II.3 Concept of unconditional release

Most countries have similar systems of radwaste classification, usually composed of three main categories, based on activity levels, half-life of radionuclides and thermal power production. In view of decommissioning of nuclear installations, most countries have decided to establish the principle of unconditional release. At present, this is not applied in France for materials originating from nuclear installations.

In 1993 [3], France therefore created a new category, defined as Very Low Level Waste (VLLW). The difference in strategies regarding radwaste categorisation is shown in table 1.

France			Belgium		
Category	**Indication of activity concentration (Bq/g)**	**Order of magnitude**	**Category**	**Indication of activity concentration (Bq/g)**	**Order of magnitude**
Trés **F**aible Activité	0 to 100	500 €/ton*	Unconditional release	0.1	<< 50 €/ton*
Faible Activité	100 to 100 000	7500 €/m³*	**L**aag **A**ctieve **V**ast **A**fval	0.1 to 32 000 (< 2mSv/h)	25 000 €/m³*

*Estimation, including conditioning and final disposal

Table 1: Waste categorisation and costs in France and Belgium

The use of unconditional release instead of radwaste category "Trés Faible Activité" (TFA) forces the producer to minimise the amount of radwaste and to invest in decontamination and characterisation methodologies. Actually in Belgium, the cost of radwaste is such that it is economically very penalising to remove big quantities of waste as Laag Actieve Vast Afval (LAVA). The system of unconditional release is quite comparable with radwaste acceptance criteria, defining radionuclide specific clearance levels. Table 2 shows the clearance levels in Belgium for Cs-137 and Co-60 according to the Belgian law, described in the royal decree of July, the 27th, in 2001 [4].

Radionuclide	**Clearance level (Cli)**
Cs-137	1 Bq/g
Co-60	0,1 Bq/g

Table 2: Some values of clearance levels.

In order to calculate an unconditional release limit, we need to know the typical spectrum of the waste and the level of activity of each radionuclide. If the sum of each nuclide ratio, between nuclide activity and clearance level of the nuclide is inferior to one, we can release. The following expression illustrates this definition:

(1)
$$\sum \frac{Ci}{Cli} \leq 1$$

Ci   is specific activity of radionuclide
Cli is clearance level of radionuclide

III. Context and purpose of the study

This study has been conducted in view of the decommissioning of Belgian Reactor 3. The main goals were to validate a non-destructive method, to determine the in-depth Cs-137 contamination in concrete and to economically optimise the results.

III.1 Belgian Reactor 3

In 1957, SCK•CEN decided to build the first Pressurised Water Reactor (PWR) in Europe, Belgian Reactor 3, as shown in figure 3. This reactor was built by the company Westinghouse, on the SCK•CEN site in Mol. For an electrical power of 10 MW, BR3 started October, the 10th in 1962 and shut down in 1987. As being a pilot reactor, it allowed to train future nuclear power plant operators. After a while, the reactor was a test bench for advanced PWR fuel, such as MOX. Moreover, the effect of different moderators on neutron spectra has been tested. After the decision to shut down BR3, it was selected in 1989 as one of the four European pilot projects of the European Commission for its R&D program on decommissioning of nuclear installations [2].

Figure 3: View of BR3

From the nineties onwards, the new challenge of BR3 was to demonstrate the feasibility of decommissioning operations. This project has allowed, among other things, to test and develop different dismantling and decontamination techniques. Techniques like milling cutter, EDM (Electrical Discharge Machining) cutting and the plasma arc torch have been used in the reactor pool [5].

During the decommissioning steps, and during the operational lifetime of the reactor, the nuclear plant infrastructure suffered from maintenance operation, failure and leakages or incidents occurring in the different circuits of the plant. This resulted in the contamination of building infrastructure at various activity levels with a different depth, up to several centimetres into the concrete [6] [7].

Today the primary loop has been completely dismantled and evacuated, the reactor pressure vessel is removed and all internal and external components like the steam generator, are evacuated [5]. Thereby, once the Neutron Shield Tank will be dismantled, about 99.9% of the total initial activity will be removed from site. One estimates that the remaining materials that still have to be removed as partly low level radwaste will represent still about 80% of the total radwaste volume.

The present study focuses on the contamination of the building infrastructure. Before decontamination, we need to know the exact depth of the contamination in order to remove the amount of material as radwaste. For the radiological characterisation of infrastructure elements, various methods are available.

Figure 4 on the right shows, common non-destructive analyses used in decommissioning that are based on the measurement of beta radiation, using a hand-held plastic scintillator. But this measurement technique can not be used to determine in-depth contamination. The common way to determine in-depth contamination in concrete is based on sampling by core drilling combined with destructive radiological analyses. However, the use of such a destructive method implies disadvantages. It is very time consuming and therefore costly. After the first promising results, non-destructive analyse using ISOCS (In-Situ Object Counting System), appears to be more effective and efficient for the determination of Cs-137 in-depth contamination in concrete [6].

Figure 4: Measurement using a "Seletra", a hand-held plastic scintillator

III.2 Presentation of ISOCS

ISOCS (In-Situ Object Counting System) is a commercial system introduced by the company Canberra. The system consists of:

- **"ISOCS characterised" germanium detector:** Is a Broad Energy Germanium detector (HPGe) with a relative efficiency of 30%.

- **ISOCS calibration software**: The detector characterisation is produced by the Monte Carlo N Particle modelling code [8]. The radiation response profile of the detector is determined for a 1000 meter diameter sphere around the detector over 45 keV through 7 MeV energy range. The results of this characterisation are delivered to the user in the ISOCS software. Moreover, one needs to choose between the various mathematical geometry templates, provided by Canberra. Before generating the efficiency curve, measurement parameters, needs to be introduced into the software.

- **ISOCS shielding system**: Composed of lead ring shielding and different collimators allowing focusing of 30°, 90° and 180°. These elements can be placed on a cart.

- **Laptop:** With acquisition software such as GENIE 2K
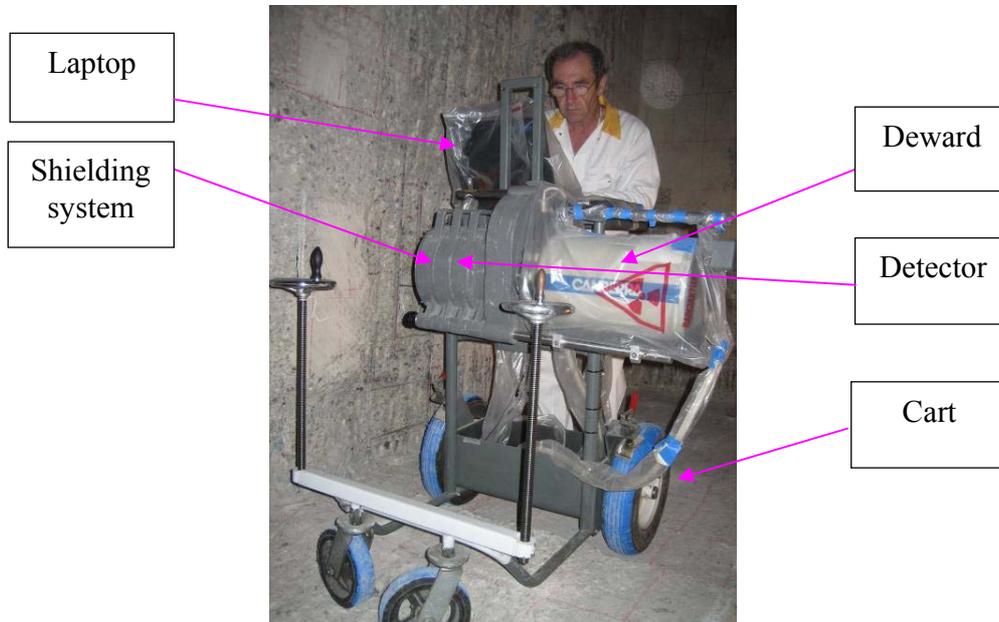
Figure 5 shows the ISOCS system.

Figure 5: Typical ISOCS set-up

III.2.1 Description of the methodology

According to the literature, the contamination profile follows an exponential decreasing, as shown in figure 6. The characteristic of an exponential profile is composed by expression (2).
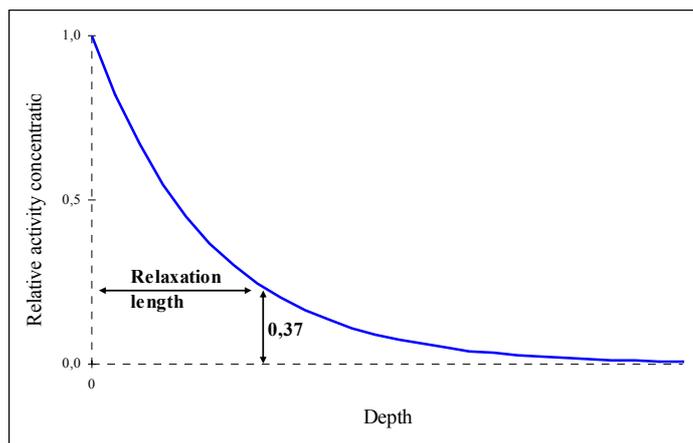

Figure 6: Exponential profile of the contamination

$$(2) \qquad A_x = A_0 \times e^{-\left(\frac{x}{RL}\right)}$$

Where: **Ax** is the activity concentration at depth x (Bq/dm²)
Ao is the activity concentration at the surface (Bq/dm²)
x is the depth (mm)
RL is the relaxation length (mm), corresponding to a distance in the material. Where the activity concentration amounts to 37 % (1/e) of the activity concentration at the surface, Ao.

The principle of the methodology is to acquire a gamma spectrum and to deduce the contamination depth using the absorption laws in matter. Most of the time, contamination involves many radionuclides occurring in nuclear plants. The easy-to-measure ones are Co-60 and Cs-137. Observed and described in several articles [9] [10], Co-60 precipitates at the concrete surface. One the other hand, Cs-137 is considered as the penalising radionuclide in terms of migration into concrete. That is why, Cs-137 has been chosen for this method. Furthermore, with 32, 36 and 661 keV (via daughter Ba-137m), we are able to use the different behaviour of these emissions. Gamma radiations are attenuated when they go through matter as much as the energy of radiation is low and the thickness of matter is important. The ratio 32/662 or 36/662, enables to assess the contamination depth using spectroscopy, as it is shown in figure 7.
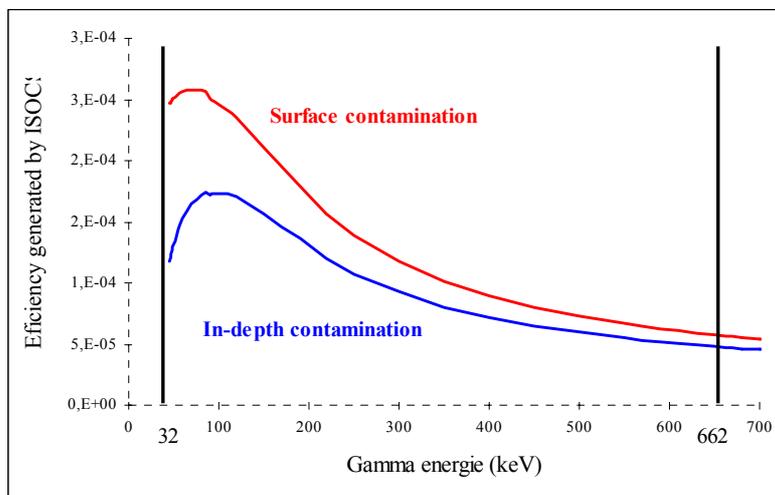

Figure 7: Efficiency curve generated by ISOCS for 2 different contamination depths

III.2.2 ISOCS: An iterative method for efficiency calculation

The first step of the method is to acquire an in-situ spectrum.

During the second step, the geometry is modelled thanks to the ISOCS software which enables to generate an efficiency curve. For this, you have to choose between various models proposed by ISOCS. The models depend on the geometry of the matrix and the profile of the contamination. Once the model is selected, the ISOCS software needs to have input parameters related to the source, measurement condition, matrix data and collimator data. Amongst others, we can quote the distance detector-ground, the chemical composition, the density of concrete and the focusing of the collimator.

According to the iterative plan described in figure 8, modifying one of the parameters leads to a modified relaxation length and therefore different contamination depth.
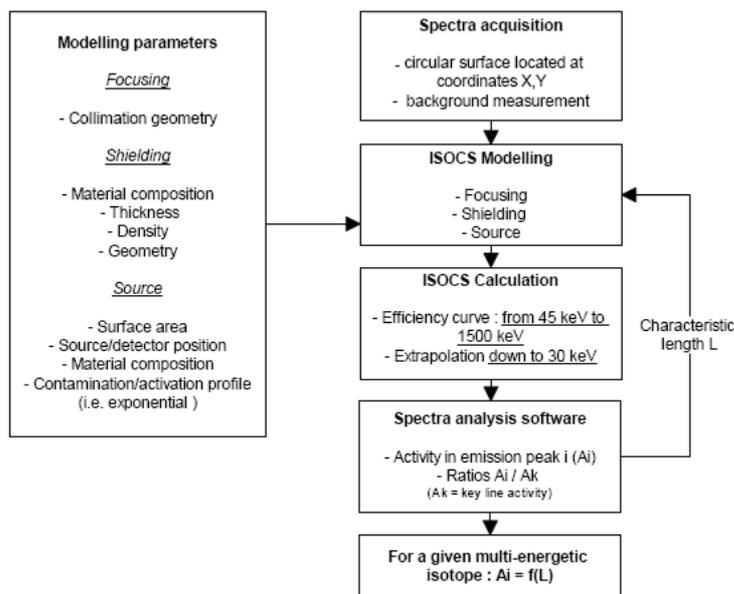

Figure 8: The iterative ISOCS methodology for multi-energetic isotopes

After generating the efficiency curve, the next step is to calculate the activity due to Cs-137, using its 32, and 662 keV emission peaks. For this step, we have taken a hypothetical value for the relaxation length. The iterative calculation changing the relaxation length is conducted until we get the same activity for two emissions peaks, in accordance with the theory.

Figure 9 shows an example of the result that we can get. By variation of the relaxation length in the ISOCS modelling parameters, and by calculation of the activity due to Cs-137 for each emission, we can make a representative graph. According to the absorption in matter laws, the point of intersection between the curves corresponds to the real relaxation length and the real activity "Ao" due to Cs-137.
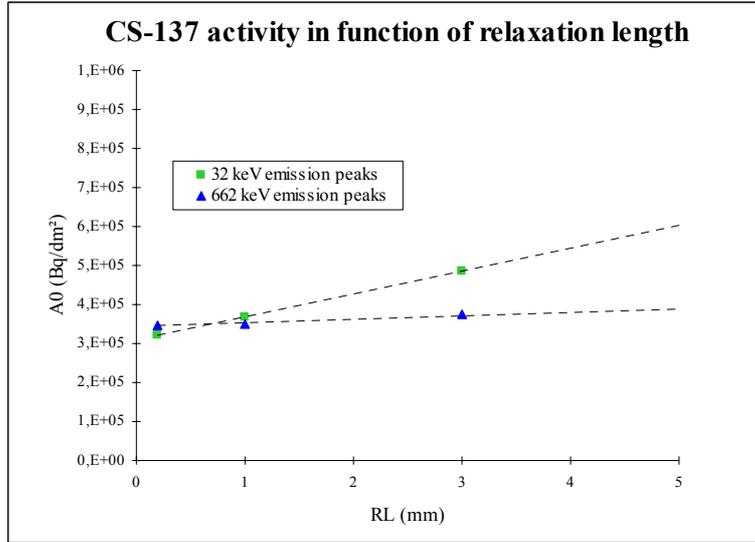
Figure 9: Activity curves for Cs-137 using its 32 keV emission peaks in function of relaxation length

Finally, the contamination depth is determined following expression (2). We used the value 77 Bq/dm² as the surface activity "Ax" corresponding to the Cs-137 clearance level in Bq/dm². Following expression (3), we can calculate the contamination depth namely:

$$(2) \quad \rightarrow \quad \frac{A_x}{A_0} = e^{-\left(\frac{x}{RL}\right)} \quad \rightarrow \quad Ln\left(\frac{A_x}{A_0}\right) = -\frac{x}{RL}$$

$$\rightarrow \quad (3) \quad x = -Ln\left(\frac{A_x}{A_0}\right) \times RL$$

III.3 Objectives of work: Validation and optimisation

During the decontamination process, a certain amount of radioactive waste will be produced. In order to minimize this amount of waste, it is important to know as accurate as possible, the in-depth contamination profile. It is necessary to define the most cost effective way to clean a concrete element down to the unconditional release limit. This study will focus on the use of ISOCS, and consists of three main objectives.

I. Identify and evaluate the main sources of uncertainty and calculate an expanded uncertainty for a fixed routine scanning set-up. Moreover propose a way to reduce the main sources of uncertainty.

II. Adjust the different parameters in order to increase the effectiveness for the fixed routine scanning set-up, and define an alternative measurement set-up for the measurement of specific spots.

III. In order to verify the validity of the calculation using ISOCS, a comparison with the traditional method, using core drilling, should be made.

IV. Uncertainty calculation

In this chapter, the sources of uncertainty generated using ISOCS are identified, and the way to calculate them is described. The expanded uncertainty is calculated for a fixed routine scanning set-up. Finally, the way to reduce the main sources of uncertainty is presented.

In view of identifying and evaluating the main sources of uncertainty, the first step is to enumerate all potential sources. Every standard uncertainty has been classified into 6 categories as it is shown in figure 10.
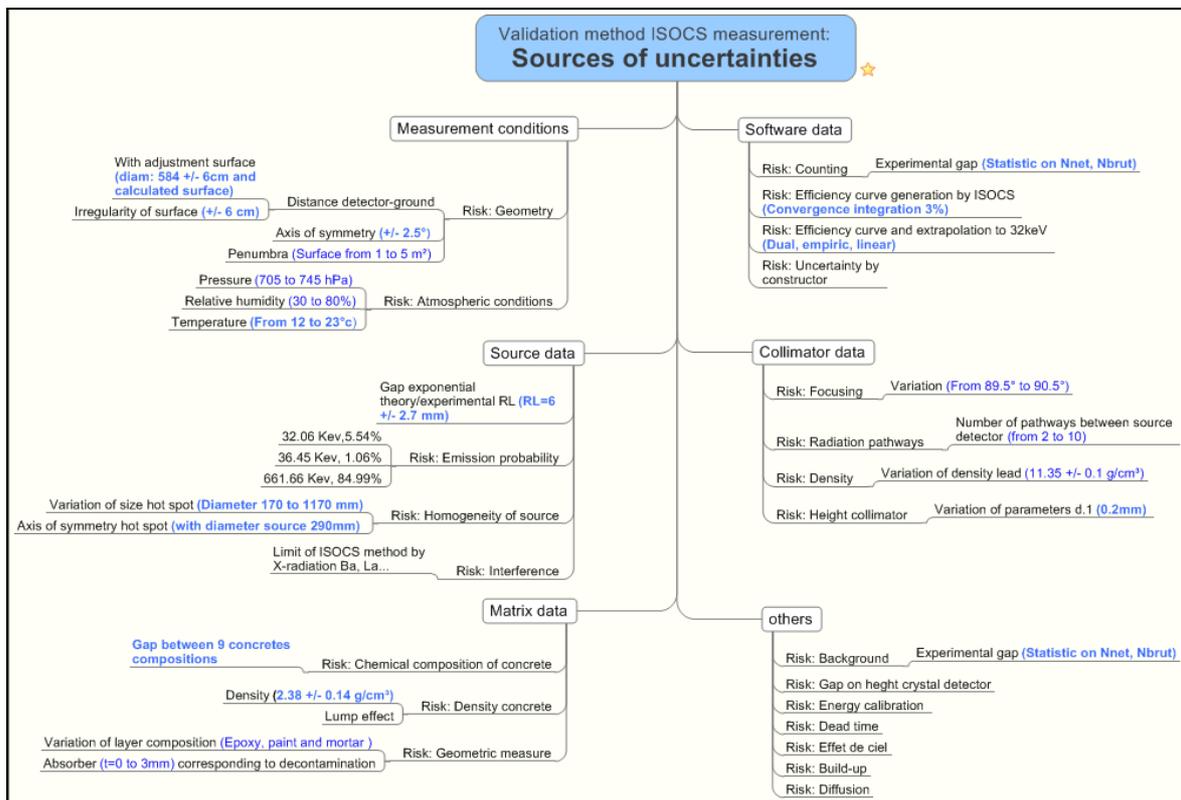

Figure 10: Sources of ISOCS's uncertainties

Following expression (4), uncertainties can be attributed to one of the parameters mentioned in this formula.

$$(4) \qquad x = -Ln\left(\frac{A_x \times \varepsilon \times I}{N_{count} - Bkg}\right) \times RL$$

Where: **Ax** is the surface activity measured (77 Bq/dm³, clearance level of Cs-137)
ε is the efficiency curve generated by ISOCS
I is the emission probability of radionuclide considered
Ncount is the count rate (c/s)
Bkg is the background (c/s)
RL is the relaxation length (mm)

In order to know the expanded uncertainty on the contamination depth (**x**), we need to know the standard uncertainty on every individual parameter. To estimate these standard uncertainties, three different methodologies are used. The first concerning the nuclear data (**I**), consists of a bibliography study, in order to evaluate the standard uncertainties on emission probability. The second concerning the number of counts is based on the analysis of many spectra (**Ncount** and **Bkg**). And the last methodology, concerning the parameters link to the efficiency curve (**ε**), and the relaxation length (**RL**), is based on the calculation of the influence of each parameter on in-depth contamination.

The confidence level used for the standard uncertainties is 68% (K=1). However during the calculation of the expanded uncertainty, the confidence level used is 95% (K=2).

IV.1 Nuclear data uncertainty

Three different sources of nuclear data have been compared. Present in *annex 1*, the nuclear data come from:

- Laboratoire National Henry Becquerel LNHB(CEA- France) [11]
- The lund/LBNL Nuclear Data Search [12]
- Erdmann and Soyka database [13]

Because it contains more recent data, the emission peak energy and the emission probability have been taken from "Laboratoire National Henry Becquerel" data base, moreover, this data base is conform with the IAEA data base [14]. However, the standard uncertainty on emission probability has been calculated, including the values from the other data bases.

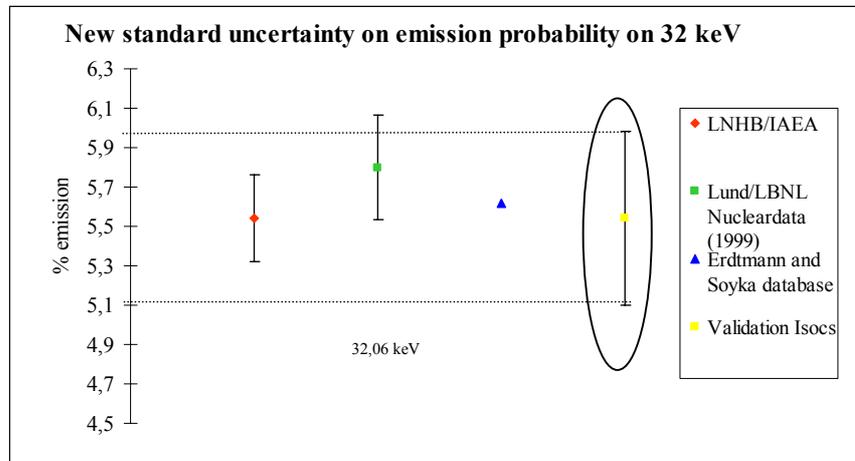A new uncertainty on emission probability calculated according to all databases is illustrated in figure 11.

New standard uncertainty on emission probability on 32 keV

Figure 11: Standard uncertainty on 32 keV

Cs-137 is emitting two photopeaks very close one another (31.8 and 32.2 keV). As it is difficult to separate these two peaks, we will consider only one peak at 32,06 keV. This standard uncertainty on emission probability is considered with 68% of confidence level (K=1).

Concerning the 662 keV emission peak, we have used the LNHB/IAEA nuclear data. Details of the standard uncertainties used are shown in table 3.

LNHB (2006)			Used for ISOCS validation		
Energy (keV)	Emission (%)	Error (%)	Energy (keV)	Emission (%)	Uncertainties (%)
31,82	1,95	2,00	32,06	5.54	8
32,19	3,59	1,95			
661,66	84,99	0,24	661,66	85,10	0,8

Table 3: Nuclear data used for ISOCS validation

In order to calculate the standard uncertainty on nuclear data, the following expression was used:

$$(5) \qquad \sigma_{NuclearDat} = \sqrt{\left(\frac{\sigma_{32}}{100}\right)^2 + \left(\frac{\sigma_{662}}{100}\right)^2}$$

$$\sigma_{NuclearDat} = \sqrt{\left(\frac{8.0}{100}\right)^2 + \left(\frac{0.8}{100}\right)^2} = 8.1\%$$

We consider the standard uncertainty on nuclear data is 8,1 %.

IV.2 number of counts uncertainty

In order to determine the standard uncertainty on the number of counts, several tests were made. The tests took place in room 143 "Ventillation" in BR3, on the spot N°1 using the routine scanning set-up. The purpose was to study the difference between the uncertainty given by "GENIE 2K" software and the standard deviation calculated on 7 spectra acquired in the same condition. The uncertainty in function of the counting time is shown in figure 12.
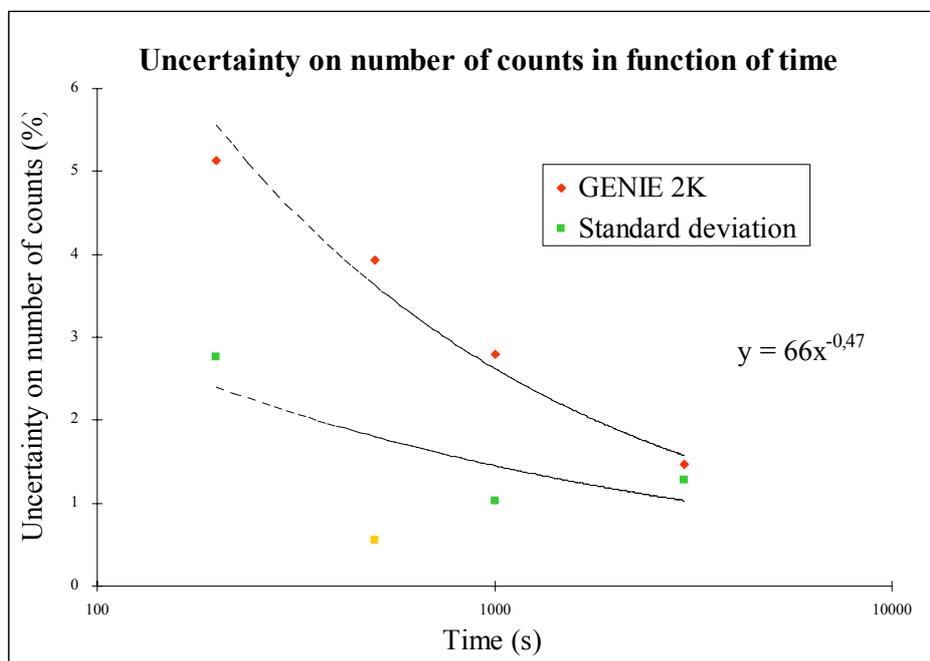

Figure 12: Standard uncertainty on number of counts for 32 keV in function of counting time

In most of the results, the standard uncertainty on the number of counts generated by the "GENIE 2K" software is higher compared to the standard deviation calculation.

In the second step, we used many spectra, in order to determine a standard uncertainty. These spectra were acquired in different measurement conditions and from different premises of BR3, before my internship. The standard uncertainties used are given by the software "GENIE 2K".

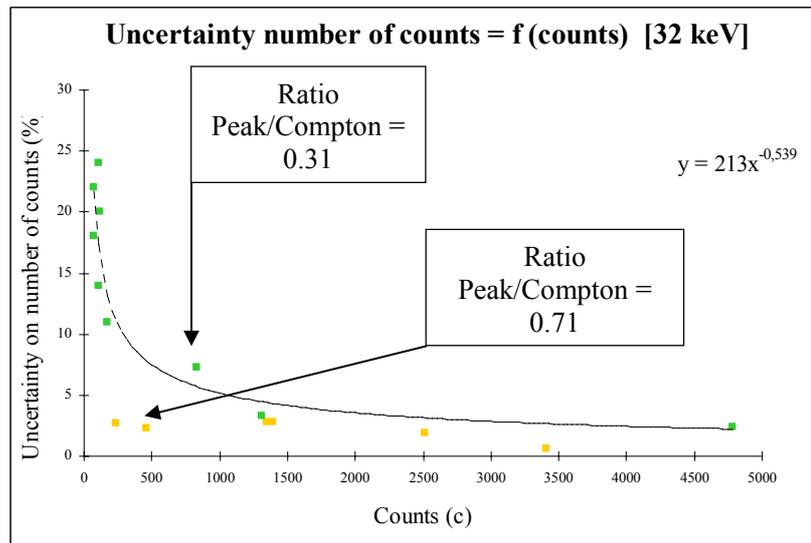Figure 13 shows the standard uncertainty calculated by the software in function of the number of counts.

Figure 13: Standard uncertainty on number of counts for 32 keV

The most important parameters for the standard uncertainty are the Peak/Compton ratio and the counting time. During an acquisition, if the background is high, the part of the Compton will become important in the ratio. The standard uncertainty will be high because of the low value of the ratio.

For calculating a standard uncertainty on a number of counts during an acquisition, a trend line was traced. Moreover, for obtaining a penalising value, I chose to use a Peak/Compton ratio equal to 0.3, corresponding to a high level of background.

I considered for a minimum counting case, a value of 1000 counts in 32 keV and a value of 500 seconds for counting time. In this case, I obtain 4.3% of standard uncertainty.

$$(6) \qquad \sigma_{NumberCou} = 4.3\%$$

We consider, the standard uncertainty on number of counts is 4,3 %.

IV.3 Efficiency curve and relaxation length uncertainty

IV.3.1 Identification

The parameters necessary to generate the efficiency curve and the relaxation length are listed in table N°4.

Sources of uncertainty	Description
Distance detector-ground	Due to the distance between detector and ground
Irregularity of surface	Due to an irregularity of ground (holes)
Axis of symmetry	Eccentricity between detector and matrix
Penumbra	Due to diameter hot spot more large than diameter seen by detector
Atmospherical conditions	Influence of pressure, relative humidity and temperature
Range on relaxation length	Due to an error on RL
Size hot spot	Due to error of hot spot size
Axis of symmetry hot spot	Due to error of hot spot axis
Chemical composition of concrete	Influence of chemical composition variation
Density concrete	Due to uncertainty on density concrete
Composition layer	Influence of epoxy, paint and mortar layer
Efficiency curve generate by ISOCS	Precisely by user
Efficiency curve by constructor	Precisely by constructor data
Extrapolation of 32 keV	Influence of different mathematic model used
Focusing collimator	Due to errors on focusing collimator dimension
Radiation pathways	Precisely by user, accurately of calculus
Density collimator	Due to error on collimator density
Height collimator	Due to errors on collimator dimension

Table 4: Description of uncertainty sources

IV.3.2 Evaluation

First and foremost, we have determined the possible range for each source of uncertainty. Three kinds of ranges may occur. The first is during acquisition (example: the distance ground-detector) the second range is between hypothesis and experimentation (example: homogeneity of source, chemical composition of concrete or extrapolation on 32 keV). And finally, the last range is due to constructor data, as collimator dimension.

The first kind of range is evaluated during acquisition or determined according to experiences. For example, a test has determined the maximum range on axis of symmetry between matrix and detector. For a value above of 2,5° the operator can notice the mistake therefore adjusts the detector symmetry.

The second kind of range is evaluated according literature. For example, the chemical composition of concrete has been evaluated following the specifications during construction of BR3, and general chemical composition of the concrete [17].

For the last kind of range, the constructor provided us the tolerance on collimator dimensions. The ranges of each single parameter are presented in chapter IV, figure 10.

IV.3.3 Methodology of uncertainty calculus

Once a range of a specific parameter is estimated, its effect on in-depth contamination is calculated. The standard uncertainty on a parameter is calculated as the ratio between the maximum range and the standard value of parameter. The following steps have been conducted:

1. To establish a link between the ratio efficiency of 32 keV and the efficiency of 662 keV in function of the depth contamination, I have generated an efficiency curve according to different values of relaxation length. Once this efficiency ratio is established in function of the relaxation length, I have calculated, for each case of efficiency ratio and according expression (2), the depth contamination. Indeed, I imposed an arbitrary value for "Ao" and I used a clearance level (77Bq/dm²) for "Ax", in order to calculate for each efficiency ratio a depth contamination. The arbitrary value of "Ao" has no importance since I used absolute values for the standard uncertainty.

2. Secondly, I varied the parameters in the ISOCS model and I generated the efficiency curve. For each variation, I calculated the efficiency ratio between 32 and 662 keV.

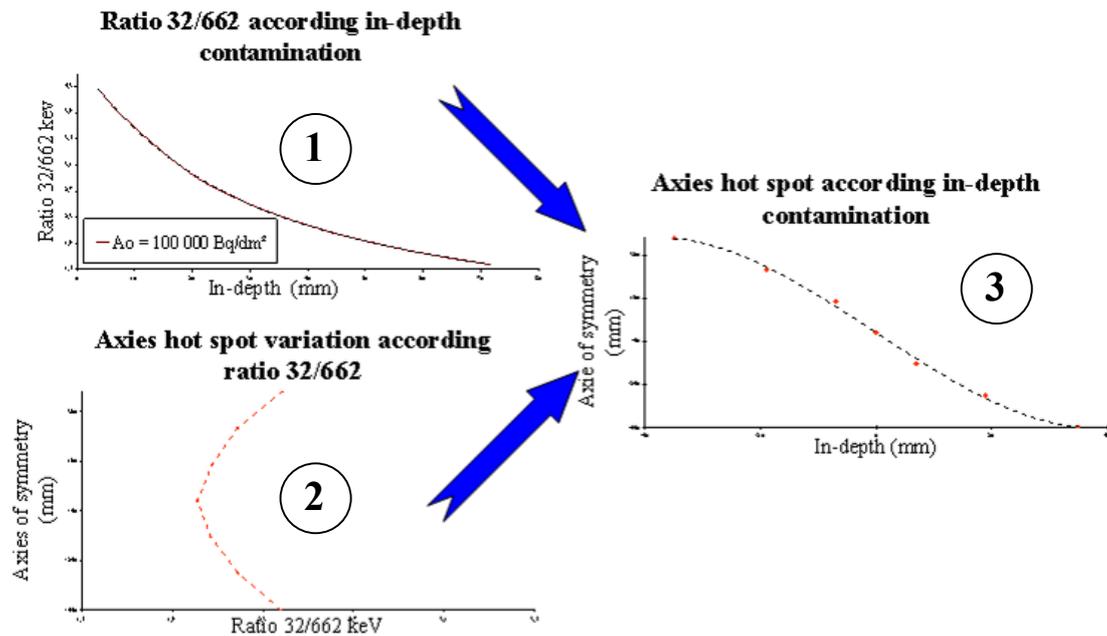Illustration of this method is shown in figure 14.

Figure 14: Principle of uncertainty determination

3. Finally, I could establish a link between the variation of the parameter and the in-depth contamination. In the way to have an absolute value, I calculated the in-depth ratio between the new value and the standard value of the parameter.

IV.3.4 Results of standard uncertainties

The standard uncertainties have been calculated for a routine scanning set-up and are presented in figure 15.
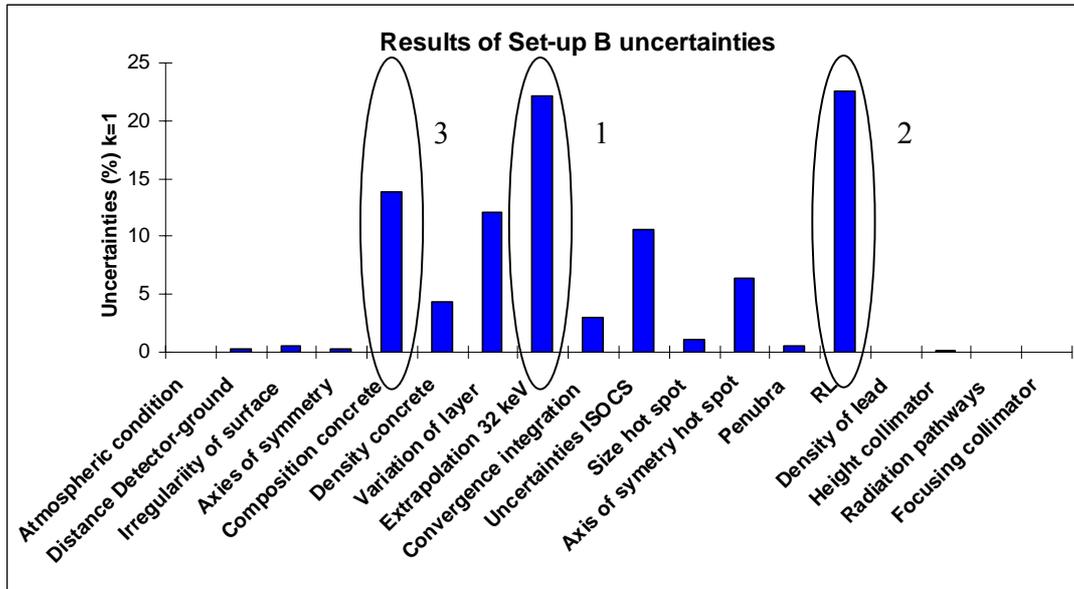

Figure 15: Standard uncertainties values for ISOCS parameters

The first main contribution to the standard uncertainty is estimated to about 22.6%. It is the relaxation length. We will see in chapter V.3.1, that the experimental contamination profile does not follows always an exponential decreasing. Consequently, the standard uncertainty on the relaxation length is very high.

The second main contribution to the standard uncertainty, is estimated at 22.2%. It is the extrapolation to 32keV. Indeed, ISOCS can generate an efficiency curve from 45 keV to 7 MeV. That is why we need to extrapolate this curve down to 32 keV. This mathematical extrapolation leads to the major standard uncertainty.

The third main contribution to the standard uncertainty is estimated to 13.9%. It is the chemical composition of the concrete.

Then, the variation of layer in matrix, is estimated at 12.1%, and the ISOCS uncertainty given by the constructor, is estimated at 10.6%. The density of the concrete and the source characteristics represent a relatively low contribution to the standard uncertainties. They are estimated at 5%. Even lower standard uncertainties are found to be the measurement geometry (example: distance detector-ground), the atmospherically condition and the collimator data. They are estimated at 0.5%. Details of standard uncertainties using different geometries measurement, are shown in *annex 2*.

The combined standard uncertainty on the efficiency curve, excluding the result on the relaxation length, is calculated following expression (7).

$$(7) \quad \sigma_{EfficiencyCur} = \sqrt{\sum_{Parameters}\left(\left(\frac{\sigma_{Parameter1}}{100}\right)^2 + \left(\frac{\sigma_{Parameter2}}{100}\right)^2 + ...\right)} = 31.8\%$$

We consider, the combined standard uncertainty on the efficiency curve is 31.8 %.

We consider, the standard uncertainty on the relaxation length is 22.6 %.

IV.4 Expanded uncertainty

The combined standard uncertainty is calculated as follows:

$$\sigma_{CombinedUnc} = \sqrt{\left(\frac{\sigma_{NuclearDat}}{100}\right)^2 + \left(\frac{\sigma_{NumberCou}}{100}\right)^2 + \left(\frac{\sigma_{EfficiencyCur}}{100}\right)^2 + \left(\frac{\sigma_{RelaxationLen}}{100}\right)^2}$$

$$\sigma_{CombinedUnc} = \sqrt{\left(\frac{8.1}{100}\right)^2 + \left(\frac{4.3}{100}\right)^2 + \left(\frac{31.8}{100}\right)^2 + \left(\frac{22.6}{100}\right)^2} = 40.1\% \quad \textbf{For k=1}$$

$$\sigma_{ExpandedUnc} = 80.2\% \quad \textbf{For k= 2}$$

The main contribution to the expanded uncertainty is coming from the combined standard uncertainty on the efficiency curve calculation and the standard uncertainty on relaxation length calculation. The expanded uncertainty is calculated with a confidence level of 95%.

IV.5 Reduction of main sources of uncertainty

In order to reduce the expanded uncertainty, we have to reduce the main contributions.

1. First contribution: The relaxation length
   We cannot directly reduce the uncertainty on relaxation length. However if the expanded uncertainty is reduced, the standard uncertainty on the relaxation length is reduced.

2. Second contribution: Extrapolation of 32 keV.
   After several requests by the users, Canberra has taken the decision in 2008 to develop an update of ISOCS including an efficiency curve generation from 30 to 7000 keV.

3. Third contribution: Chemical component of concrete
   The composition of the matrix is an important factor for the expanded uncertainty. Consequently, we have sent samples of concrete and mortar to a chemical laboratory. These samples originate from different premises of BR3, in order to have a representative result. Unfortunately, due to lack of time the results will not be ready for the end of my study.

Figure 16 shows an estimation of standard uncertainties before this study, the standard uncertainties calculated during this study, and an estimation of the standard uncertainties in the next future.
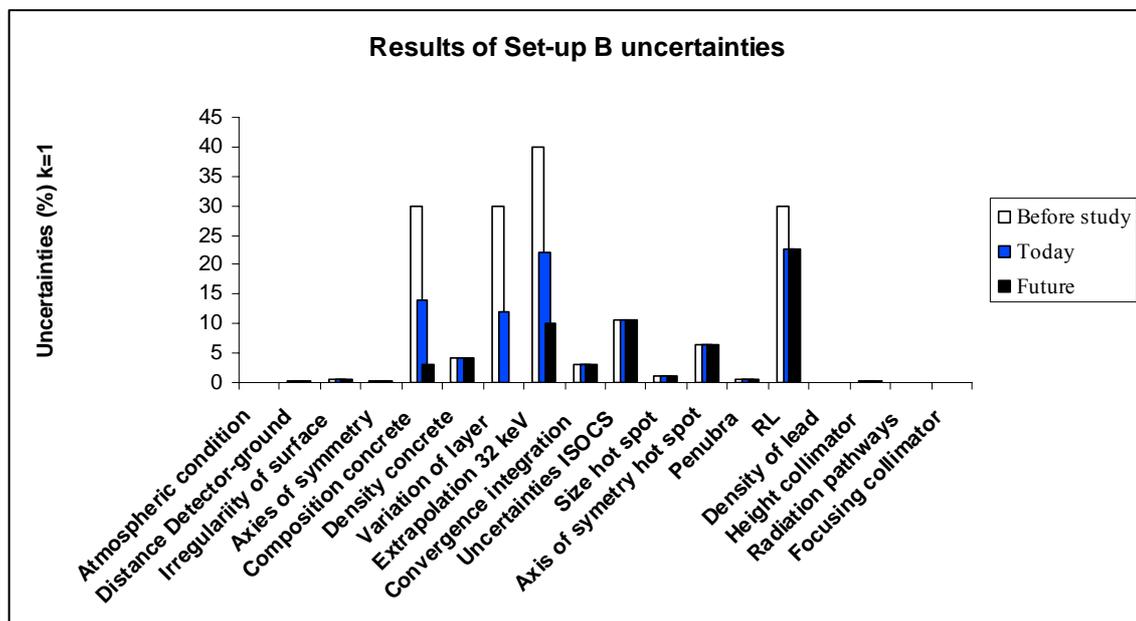

Figure 16: Estimation of old, actual and future standard uncertainties

The expanded uncertainty for this methodology was estimated to amount more than 100% before this study. Today, we have seen in chapter IV.4, the expanded uncertainty is about 80 %. And in future, thanks to the reduction of the main sources of uncertainty, we hope to reach a combined uncertainty of about 60%.

V. Measurement procedure and parameters

First and foremost, the choice of spectral analysis parameters using the standard software "GENIE 2K" will be explained. Subsequently the test on geometry measurement and on ISOCS model will be presented, in order to conclude on the set-up for the measurement of specific spots.

V.1 Different parameters

V.1.1 Spectral analysis parameters

In view of using ISOCS standard models, the first task has been to optimise the spectral analysis parameters. Indeed, using a different mathematical function or different parameter values in GENIE 2K could lead to different results, during the analysis of the spectrum. Important parameters are the peak localisation, the peak area calculation and the nuclide identification. Therefore, I have studied the impact of these parameters on the count rate, Minimal Detectable Activity (MDA) and the peak's deconvolution.

Figure 17, shows the importance of having a good photopeak. I have used three spectra for analysis. These spectra correspond to an acquisition in three different conditions. The first two spectra correspond to good count rate statistics with both low and high background (Spectrum left shown in figure 17). The third spectrum corresponds to degrade condition (Spectrum right shown in figure 17), namely, a low statistic count rate and high background. This study has been conducted for the 32.keV, 36keV and 662 keV photopeaks. It is obvious that the impact of the uncertainty on MDA and Nnet is getting more important in case of low emission rates.
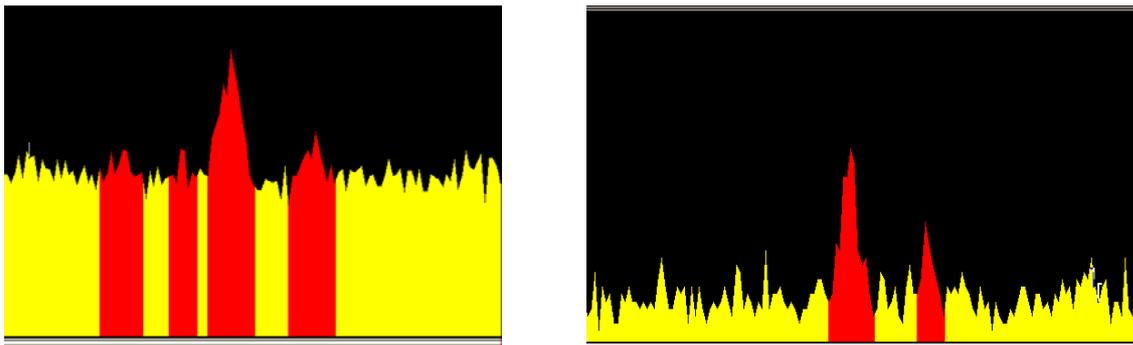

Figure 17: Spectrum of 32 and 36 keV in good and degrade condition

V.1.2. Results

This section only presents the results corresponding to the main spectral analysis parameters. The remainder of the results are presented in *annex 3*. The photopeak used is 32 keV and the spectrum corresponds to a bad condition (Figure 17).

Type of parameter: Peak location
Parameter study: Significance threshold
This parameter corresponds to the sensibility for seeking peaks. A number greater than 0 allows to "eliminate" insignificant peaks. The variation of the "significance threshold" according Nnet and MDA 32keV is shown in figure 18.
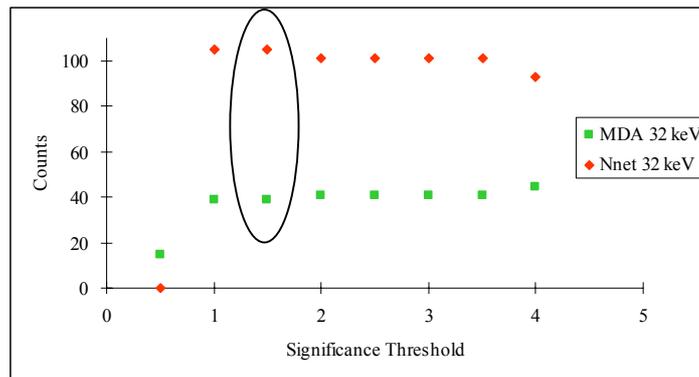

Figure 18: Variation of "significance threshold" on Nnet and MDA at 32 keV

I chose for a value of 1.5, which seems to be a good compromise between the elimination of an insignificant peak and an acceptable value for the MDA.

Type of parameter: Peak area
Parameter study: Region Of Interest max
This parameter allows the user to select the maximum limits for the determination of Region Of Interest (ROI). The unit is FWHM, in order to vary the size of ROI across the spectrum. The variation of "ROI max" according Nnet and MDA 32keV, is shown in figure 19.
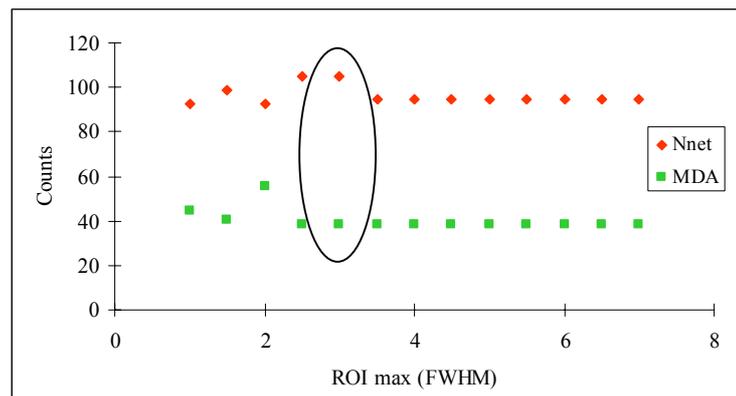

Figure 19: Variation of "ROI max" on Nnet and MDA at 32 keV

A value of 3 and more appears to be a good value for limiting the determination of a ROI.

V.2 Measurement geometries

In order to implement a set-up measurement for specific spots, we have performed many tests with different geometries. The geometry includes distance detector-ground and collimator.

One the one hand, we have tested the influence of the distance detector-ground on statistical count rates. To do this, we have used three different distances, 584mm, 300mm and 220mm imposed by the constructor's cart.

Figure 20 gives an overview of the different collimators that have been tested.
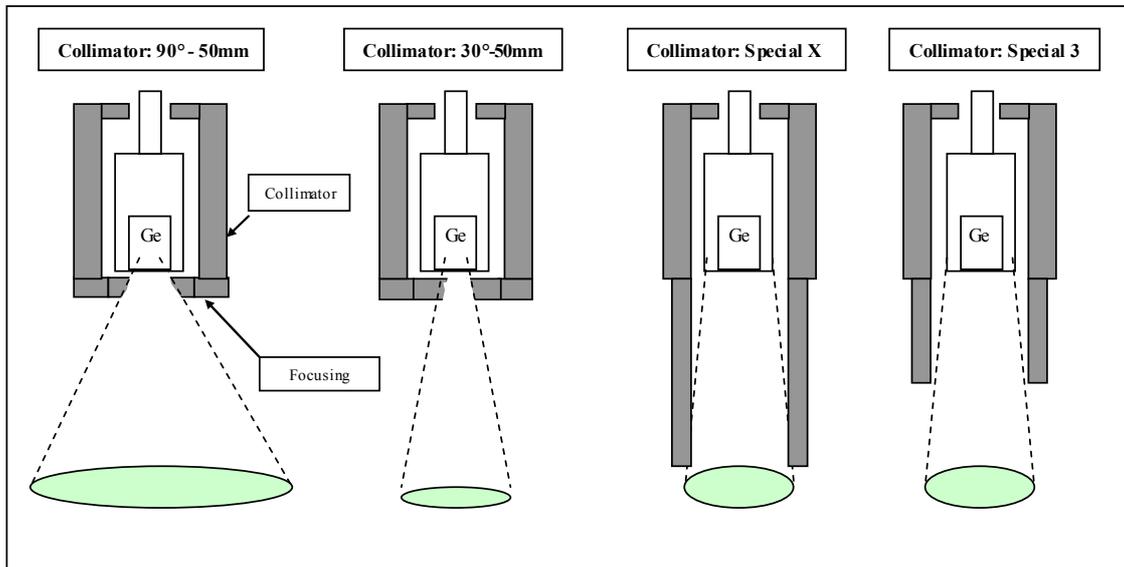

Figure 20: Four kinds of collimators tested

The tests took take place in room 143 "Ventilation" at BR3. Four hot spots were observed using a hand-held plastic scintillator "Selectra". For each hot spot, we have tested different geometries.

On the other hand, we have tested the influence of collimator focusing on statistical count rates. Three types of focusing were used, 0°, 30° and 90°. Additionally we have tested two special collimators called "Special 3" and "Special X". For "Special X", the focusing collimator is taken out, but another ring collimator is added in the way to have ring collimator down to the ground. The "special 3" is comparable with "special X", but some ring collimators have been removed to increase the active surface of detector.

V.3 Choice of the ISOCS model

V.3.1 Contamination profile

According to literature, the mathematical profile of the contamination follows an exponential decrease. Starting with a maximum activity concentration at the surface of the ground, and decreasing down to zero (if the matrix thickness allows it). However, a study shows that the real mathematical function is similar to an exponential but has specific characteristics [16]. In practice contamination can follow a preferential path like a crack present in a ground. Nevertheless, in this study we use an exponential profile.

V.3.2 ISOCS model

As described in chapter III.2.2, in order to generate efficiency curve, we have to choose an ISOCS model. This model depends on the source geometry and on the profile of the contamination. In our case, I chose to study two models.

In the first case, the model used is called "**Exponential circular plane**" (ECP). This is a model for a plane surface such as a floor or a wall. First and foremost, in order to use the exponential profile of activity, I have to precise the value of relaxation length and the value of matrix thickness. Once the characteristics of the exponential curve are specified, I have to enter the physical characteristics of the matrix. Parameters such as diameter of the matrix, density and chemical composition of the matrix are asked. Then I have to precise parameters related to count condition such as, axis of symmetric between detector and matrix, distance ground detector or thickness of absorber present between detector and matrix. At last, I have to choose the appropriate collimator.

In the second case, the model studied is called "**Circular plane**" (CP). I have to precise the physical characteristics of the matrix, the parameters of count condition and the collimator. But instead of defining the exponential profile parameters, I have the possibility to cut the matrix into several layers. Therefore, I can associate each layer with different chemical compositions, and different percentages of activity, in such way to create the appropriate profile. Figure 21 shows, the model "Circular Plane" .
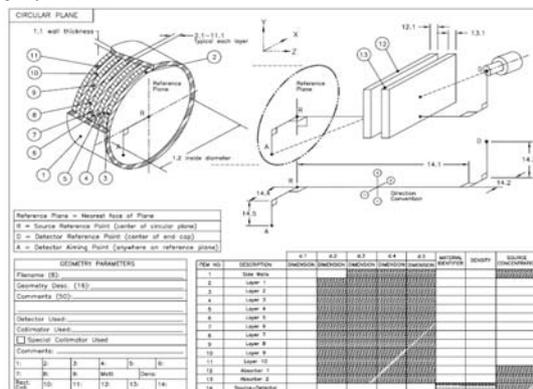

Figure 21: Model Circular Plane

Generally, the model "Circular plane" seems to be more flexible than the first model. I can adjust the profile of activity and the chemical composition of layer component this source. However, we will see that it is rare to have only one component, concrete for example. In most cases, we have layers of paint, epoxy or mortar.

V.4 Set-up for specific spots

We consider two types of measurements. A first fast measurement (scanning) should allow us to determine the coordinates of hot spots in a room, in short time. To do this, we use a geometry with a diameter seen by detector enough large. A second longer measurement should allow us to more accurately determine the in-depth contamination. Moreover, according to literature [15], the median values of lateral distribution activity are between 17 and 41 cm. Consequently, the diameter seen by detector should be about 30 cm.

The purpose is to estimate the geometry susceptible to be more adapted for the second measurement. Then the measurement of in-depth contamination should be determined.

Table 5 shows the results of the geometries studied. The distance ground-detector and the focusing both influence the counting statistics.

Geometry	Distance ground-detector	Focusing (°)	Count rate 32 keV (c/s)	Count rate 662 keV (c/s)	Time required (s)	Diameter seen (mm)
A	585mm	30	0,5	3,9	21600	313
B	585mm	90	6,8	33,8	1470	1170
B	585mm	90	6,8	33,8	1470	1170
C	230mm	30	2,5	21,9	3980	123
D	230mm	90	38,3	173,9	261	460
SPX	300mm	Special	26,8	86,7	373	108
SP3	300mm	Special	34,6	109,3	289	145

Table 5: Results of the geometries studied

The time required for having at least 10 000 counts in each peak is considered as good statistics. Moreover, following diameter seen by detector and distance ground-detector tested, I have calculated the surface seen by detector. Therefore we can assess the time required for scanning the ground of a room, according to geometry used.

The geometry B corresponds to the first fixed scanning set-up.

During the uncertainty calculation in chapter IV, we have observed relatively low uncertainties, for the parameters regarding the set-up (example: distance detector-ground, penumbra or axis of symmetry hot spot). Consequently, in view of the minimal time required and the diameter seen by the detector, the geometry B, seems to be also appropriate for the second type of measurement (increased measurement time).

VI. Comparison of measurement methods

In order to verify the validity of the non-destructive method, a comparison with the traditional method, using core drilling has been performed. During the measurement geometries study, four hot spots have been selected among the most contaminated. However, only the results of hot spot N°1 are presented. The in-depth contamination of the other hot spots was less than 1 cm.

VI.1 Drilling core

The drilling was performed by a BR3 operator, assisted by a technician. The operation was supervised by a radiation control agent. During this work, the drilling had to be stopped because a cable that was not mentioned in the room's plan. Fortunately it was a disconnected cable but it is shown that we must take maximum care during working in a controlled zone. The waste water of the drilling system was recovered and processed by evaporation at BR3. Figure 22 shows an example of concrete core.

Figure 22: Concrete core of hot spot N°2 in room 143

After drilling the core was removed and cut in several thin discs, destroyed and put in a container for counting. Each sample was crushed and milled in order to homogenise. The power was brought into a standard geometry (20cc) and analysed using a Sodium Iodide (NaI) detector. Sodium Iodide is a plastic scintillator detector. As an additional control, the samples were analyses as well using ISOCS. It is possible to use ISOCS for sample measurements since certain ISOCS models were made for this application. The interest is to use a germanium detector, in order to have access to low activity.

VI.2 Results

Due to the low specific activity of most of the hot spots, only the results of hot spot N°1 are presented. As shown in table 6, the values by NaI detector and Germanium detector are similar.

Drilling	Matrix	Slop (mm)	A (Bq/g) in Cs-137	
			Ge	NaI
N°1 x=4840 y=6200	Epoxy	2.9	5164	/
	Mortar	13.1	47	45
	Mortar	24.5	0,7	0,6
	Mortar	36.5	0,2	0,3
	Cement	52.4	0,1	<MDA

Table 6: Results for the hot spot N°1 in room 143 by core drilling

Due to the poor in-depth contamination and due to the minimal thickness of sample cutting (7mm), only 5 points could be measured. Due to the uncertainty on gamma spectrometric measurement for the results, I have used only the first three points. The in-depth contamination is calculated using the exponential trend line (figure 23).
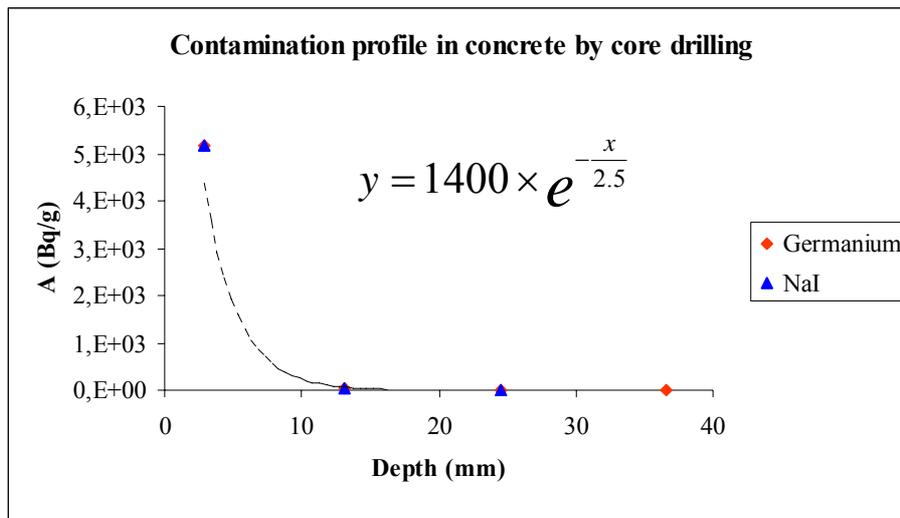

Figure 23: Activity in Cs-137 in function of depth in room 143

According to expression 3, I have calculated the in-depth contamination. I have used the value 1 Bq/g as clearance level for Cs-137 (Ax). Ao and RL are obtained by the trend line equation, they respectively amount to 14000 and 2.5. In this case, I have found 23 mm as in-depth contamination.

$$(3) \qquad x = -Ln\left(\frac{A_x}{A_0}\right) \times RL \quad \rightarrow \quad x = -Ln\left(\frac{1}{14000}\right) \times (2.5) = 23mm$$

Table 7 shows the results for the in-depth contamination for the different geometries as described in chapter V.4.

Geometry	Distance ground-detector	Focusing (°)	ISOCS Model	in-depth contamination (mm)
A	585mm	30	CP	20
B	585mm	90	ECP	12
B	585mm	90	CP	16
C	230mm	30	CP	21
D	230mm	90	CP	17
SPX	300mm	Special	CP	19
SP3	300mm	Special	CP	21

Table 7: Results of in-depth contamination

VI.3 Conclusion

- ISOCS model

During the choice of the ISOCS model in chapter V.3, the model CP seems to be more flexible than the ECP model. In view of the results, the model ECP has obtained a low-level of in-depth contamination. Due to the impossibility of the model to precise the presence of epoxy and mortar in matrix, the value of in-depth appears to be far from the real in-depth contamination. Consequently, the CP model is preferred over the ECP model.

- Geometries measurement

During the calculation uncertainty in chapter IV.3.4, the results show a rather low influence of the different geometries on standard uncertainty. However, the results on in-depth contamination show that the geometries measurement A, C, SPX and SP3 appear to be closer to the destructive in-depth value. Consequently, it might be interesting to perform additional measurements using spots with high specific activity and more in-depth contamination.

- Validity of the calculation using ISOCS

Despite the lack of knowledge on the in-depth contamination uncertainty using the non-destructive method, the results of comparison from both methods are in agreement. The ISOCS measurement results seem to be validat for this one spot. However, it might be useful to perform other tests, in order to compare the both methods in a larger range of in-depth contamination.

VII Conclusion

In view of the validation of a non-destructive methodology to determine the in-depth contamination in concrete structures, the first objective was to calculate an expanded uncertainty. After identifying and quantifying the most important sources of uncertainty, we have found an expanded uncertainty amounting to about 80% (K=2). Optimisation of some of the most important sources of uncertainty will probably decrease of the expanded uncertainty down to about 60% (K=2). A few percentages could still be gained, changing the measurement geometry towards a specific spot measurement. However, this might increase the measurement costs.

A comparison of the non-destructive method with the traditional method (sampling by core drilling, sample preparation and gamma spectroscopy) showed a more than acceptable result. In order to fully complete the validation, an intercomparison between the traditional method and the new non-destructive method on different contamination depths should be performed.

It is clear that the non-destructive method has a great number of advantages compared to the traditional one. One weak point however is the size and weight (nitrogen cooling) of a germanium detector. The possibility to replace it by a less fragile detector and more compact measurement system could be envisaged.

REFERENCES

[1]   G. Collard, "Presentation SCK·CEN", during Master ITDD, November 2007

[2]   www.sckcen.be, September 2008

[3]   Directive DGD N°1, March 26 1993

[4]   "Recommended radiological protection criteria for the clearance of buildings and building rubble from the dismantling of nuclear installation", Recommendation s of the group of experts set-up under the terms of Article 31 of the Euratom Treaty, European Commission, Radiation protection 113, 2000

[5]   M.Klein, O.Edmond, M.Ponnet, "Overview of recycling technologies for decommissioned materials: Lessons learned during the dismantling of a reactor", IAEA Conference, Malta, 5-9 November 2001

[6]   S.Boden, E.Cantrel, "Determination of in-depth contamination in concrete using a non-destructive technique", scientific report, 2006

[7]   E.Cantrel, S.Boden, "Pre decommissioning radiological survey of BR3 infrastructures", Scientific report, 2005

[8]   Canberra, "In-Situ Gamma Spectrometry With ISOCS, an In-Situ Object Counting System", Application note, 2000

[9]   F.Saintamon, D.Noel, "Measurement of the contamination depth of nuclides in concrete using surface spectrometric methods", EDF R&D, 200

[10]   C.Martin, S.Leclercq, H.Catalette, D.Noel, "Assessment of the penetration depth of radionuclides in concrete", EDF R&D, 2004

[11]   http://laraweb.free.fr, March 2008

[12]   http://nucleardata.nuclear.lu.se/nucleardata/toi/nucSearch.asp, March 2008

[13]   http://nucleardata.nuclear.lu.se/Database/es, March 2008

[14]   http://www.gammaspectrometry.co.uk/appendices/appendixb.htm, March 2008

[15]   L.Hummel, B.Sitte, K-H.Lehmann, "Concepts and methods for the clearance measurements of building structures of nuclear facilities under decommissioning", TUV Süddeutchland, 1999

[16]   C.Martin, S.Leclercq, D.Noel, H.Catalette, "Assessment of the penetration depth of radionuclides in concrete", EDF R&D, 2004

[17]   A.A.Naqvi,M.M.Nagadi,O.S.B.Al-Amoudi, "Non-destructive evaluation of concrete samples using PGNAA technique "King Fahd University of Petroleum and Minerals, Dhahram,2006

LIST OF ACRONYMS

ANDRA: Agence Nationale pour la gestion des Déchets RAdioactifs
AREVA NP: AREVA Nuclear Production
BR3:   Belgian Reactor 3
CP :   Circular plane
ECP :   Exponential Circular Plane
EDM   Electrical Discharge Machining
FBFC: Franco-Belge de Fabrication du Combustible
FWHM   Full Width at Half Maximum
HPGe   Hyper Pure Germanium
IAEA :   International Atomic Energy Agency
ISOCS: In-Situ Object Counting System
LAVA: LaagActieve Vast Afval
LNHB: Laboratoire National Henry Becquerel
MDA: Minimal Detectable Activity
MOX: Mixed OXide
NaI: Sodium Iodide
NIRAS-ONDRAF: National Instelling voor Radioactief Afval en verrijkte Spelijtstoffen-
                        Organisme National des Déchets RAdioactifs et des matiéres Fissiles
                        enrichies
RL: Relaxation Length
ROI: Region Of Interest
R&D: Research and development
TFA: Trés Faible Activité
VLLW: Very Low Level Waste

ANNEX

Annex 1: Nuclear data values used

LNHB (2006)			Lund/LBNL Nuclear data (1999)			Erdtmann and Soyka database		
Energy (keV)	Emission (%)	Error (%)	Energy (keV)	Emission (%)	Error (%)	Energy (keV)	Emission (%)	Error (%)
31,82	1,95	2,00	31,82	2,04	2,45	31,82	1,92	/
32,19	3,59	1,95	32,19	3,76	2,12	32,19	3,70	/
36,45	1,06	2,09	36,35	1,03	4,48	36,4	1,04	/
661,66	84,99	0,24	661,66	85,10	0,24	661,62	84,62	/

Annex 2: Details of standard uncertainties using different geometries measurement

Geometry	A (CP 230 90°)	B (CP 585-90°)	B (ECP 585-90°)
Efficiency curve and relaxation length standard uncertainties (% K=1)			
Distance Detector-ground	1,0	0,3	1,9
Irregularity of surface	0,3	0,5	1,5
Axis of symmetry	0,1	0,3	1,8
Penumbra	1,8	0,6	3,0
Atmospheric condition	0,1	0,1	0,1
Composition concrete	13,9	13,9	16,2
Density concrete	4,4	4,3	5,4
Extrapolation 32 keV	22,4	22,2	33,9
Convergence integration	3,0	3,0	3,0
Size hot spot	2,9	1,1	7,0
Axis of symmetry hot spot	3,5	6,4	14,6
Density of lead	0,1	0,1	0,2
Height collimator	0,3	0,2	/
Radiation pathways	0,0	0,0	0,2
Variation of layer	17,4	12,1	13,9
RL	22,6	22,6	20,3
Uncertainties ISOCS	10,6	10,6	10,6
Focusing collimator	0,6	0,1	/
Other standard uncertainties (% K=1)			
Nuclear data	7,5	7,5	7,5
Nnet	4,5	4,5	4,5

Annex 3: GENIE 2K treatment parameters

Standard sequence GENIE 2K

Step traitment	Parametrers	Values
Peak Locate-Unidentified 2nd Diff	Search region	50-8000
	Significiance threshold	1,5
	Tolerance	1 FWHM
Peak Area-Sum/Non-linear LSQ fit	Peak Area Region	50-8000
	Continuum	3 FWHM
	Continuum fonction	Step
	ROI Limits Determination	3 FWHM
		1,5 FWHM
		1,5 FWHM
	Cheked	Display ROIs
Area Correction-Std. Bkg. Subtract	Background subtract	Bkg standard validation
	Tolerance	1 FWHM
Efficiency Correction- Standard	Efficiency Calib Type	Dual
Nucleide Identification-NID w/Interf. Corr.	NID range	50-8000
	NID library	Cs-137 Co-60 Validation
	Tolerance	1 FWHM
	NID Confidence threshold	0,3
	MDA confidence factor	5%
Detection limits- Currie MDA	Confidence Factor	5%

The bleu parameters represent those who were studied

LIST OF FIGURES

LIST OF TABLES