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Sampling plans for use case 2

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Summary

The main objective of work package 3 (WP3) is to draft a sampling guide for initial nuclear site characterization in constraint environments, before decommissioning, based on a statistical approach. The first task consisted in providing an overview of available sampling design methods described in standards and guides followed by a brief presentation of the statistical methods that can be used to demonstrate meeting the objectives in the context of initial nuclear site characterization in constraint environments (deliverable 3.1). The second task aimed at developing a strategy for sampling in the field of initial nuclear site characterization in view of decommissioning, with the most important goal to guide the end user to appropriate statistical methods to use for data analysis and sampling design (deliverable 3.2). The data analysis and sampling design strategy was furthermore translated into a web based tool (deliverable 3.3). The third task is targetting at applying the strategy developed on three reference use cases: ? Use case 1: Decommissioning of a back/end fuel cycle and/or research facility; ? Use case 2: Decommissioning of a nuclear reactor; and ? Use case 3: Post accidental land remediation. This document describes the implementation of the strategy and the resulting sampling plan for use case 2, the radiological characterization of the biological shield of the BR3 reactor.

Approval

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Improved Nuclear SIte characterization for waste minimization in DD operations under constrained EnviRonment

Research and Innovation action
NFRP-2016-2017-1

WP3 –Sampling strategy

Sampling plan for use case 2 BR3 Bioshield

Deliverable 3.5

Version n° 1

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Contents

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- Use case 1: Decommissioning of a back/end fuel cycle and/or research facility;
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- Use case 3: Post accidental land remediation.

This document describes the implementation of the strategy and the resulting sampling plan for use case 2, the radiological characterization of the biological shield of the BR3 reactor.

Abbreviations

BR3	Belgian Reactor number 3
BSS	Basic Safety Standards (Directive)
DDW	Dismantling, Decontamination and Nuclear Waste (Expert Group)
DTM	Difficult-To-Measure radionuclide
ETM	Easy-To-Measure radionuclide
FANC/AFCN	Federaal Agentschap voor Nucleaire Controle – Agence Fédéral de Contrôle Nucléaire – Federal Agency for Nuclear Control
INSIDER	Improved Nuclear SIte characterization for waste minimization in Decommissioning under constrained EnviRonment
ISOCS	In Situ Object Counting System
NIRAS/ONDRAF	Nationale Instelling voor Radioactief Afval en verrijkte Splijtstoffen - Organisme National des Déchets Radioactifs et des matières Fissiles enrichies - Belgian National Agency for Radioactive Waste and enriched Fissile Material
nm	Not measured
NST	Neutron Shield Tank
PWR	Pressurised Water Reactor
SCK•CEN	Studiecentrum voor Kernenergie – Centre d'Etude de l'Energie Nucléaire – Belgian Nuclear Research Centre
RB	Reactor Building
SG	Steam Generator
RPV	Reactor Pressure Vessel
UC	Use Case
WP	Work Package

1 Introduction and goal

The EURATOM work programme project INSIDER (Improved Nuclear SIte characterization for waste minimization in Decommissioning under constrained EnviRonment) was launched in June 2017. It aims at improving the management of contaminated materials arising from decommissioning and dismantling (D&D) operations by proposing an integrated methodology of characterization. The methodology is based on advanced statistical processing and modelling, coupled with adapted and innovative analytical and measurement methods, in line with sustainability and economic objectives. The overall objective of INSIDER is to develop and validate a new and improved integrated characterisation methodology and strategy during the D&D process, based on three main use cases – a nuclear R&D facility, a nuclear power plant and a post accidental site remediation. For the second use case, a nuclear power plant, the bioshield of the Belgian Reactor number 3 (BR3) has been selected.

BR3 is a relatively small 10 MWe (about 41 MWth) Pressurised Water Reactor (PWR) of the SCK•CEN (Belgian Nuclear Research Centre). The pilot PWR for the later Belgian nuclear commercial Power Plants BR3, was brought into operation in October 1962 and was definitively shutdown in 1987 after 25 years of operation and eleven campaigns. Figure 1 shows an aerial view with an indication of the main buildings. The heart of the reactor, the reactor pressure vessel (RPV) and primary circuit, was located in the reactor building (RB). Figure 2 shows a cross section of the reactor building during exploitation. The whole primary circuit (RPV, Steam Generator (SG), pumps & primary loop, etc.) has been dismantled, as well as the ventilation and the anti-missile slabs. The bottom part of the reactor building consists of reinforced ordinary concrete. The remainder of the reactor building consists of reinforced heavy concrete. The concrete of the reactor pool close to the reactor pressure vessel is activated. Since the reactor pressure vessel was surrounded by a neutron shield tank, the surrounding concrete is not activated, except locally near the hot and cold legs. Historical measurements confirm this. In a first, very rough estimation, the activated part of the concrete is marked in red colour (Figure 2). Figure 3 shows a picture after dismantling the main components and before dismantling the pool liner (2016). Figure 4 gives a 3D model of the part of the BR3 bioshield taken into consideration for the radiological characterisation programme. The main goal of the radiological characterisation programme is to economically optimise the bioshield dismantling strategy using a waste-led approach.

The main objective of Work Package 3 (WP3) within the INSIDER project, is to draft a sampling guide for initial nuclear site characterization in constraint environments before decommissioning, based on a statistical approach. This is done by selecting state-of-the-art techniques concerning sampling design optimization, using prior information and multiple iterations, testing the approach through different case studies and reviewing the feedback from overall uncertainty calculations. The process followed to meet the main WP3 objective consists of four steps:

1. Status: provide an overview of the available sampling design methods and state-of-the-art statistical techniques.
2. Development: develop a strategy/methodology and implement it in a software package by making use of (and possibly extending) state-of-the-art techniques.
3. Implementation: apply the methodology to the different test cases considered in order to test its adequacy.
4. Guidance: summarize all the findings in a comprehensive sampling strategy guide.

This document is connected to step number three; the implementation. The strategy developed in step 2 is being implemented on the radiological characterisation of the BR3 bioshield, defined as use case 2 in the INSIDER project. This document hence defines the sampling plan for the BR3 bioshield. Sections 2 and 3 describe the objectives and constraints. A preliminary data analysis is performed based on pre-existing data (sections 4 and 5). This leads to a preliminary sampling plan. Results of the in-situ total gamma mapping of the inner surface of the bioshield results in the adjustment of the initial sampling plan (section 6). Section 7 is dedicated to the general arrangements related to integrated management issues.

The results of the characterisation programme will later on be evaluated and compared to the initial objectives. This evaluation is not part of the scope of the current document.

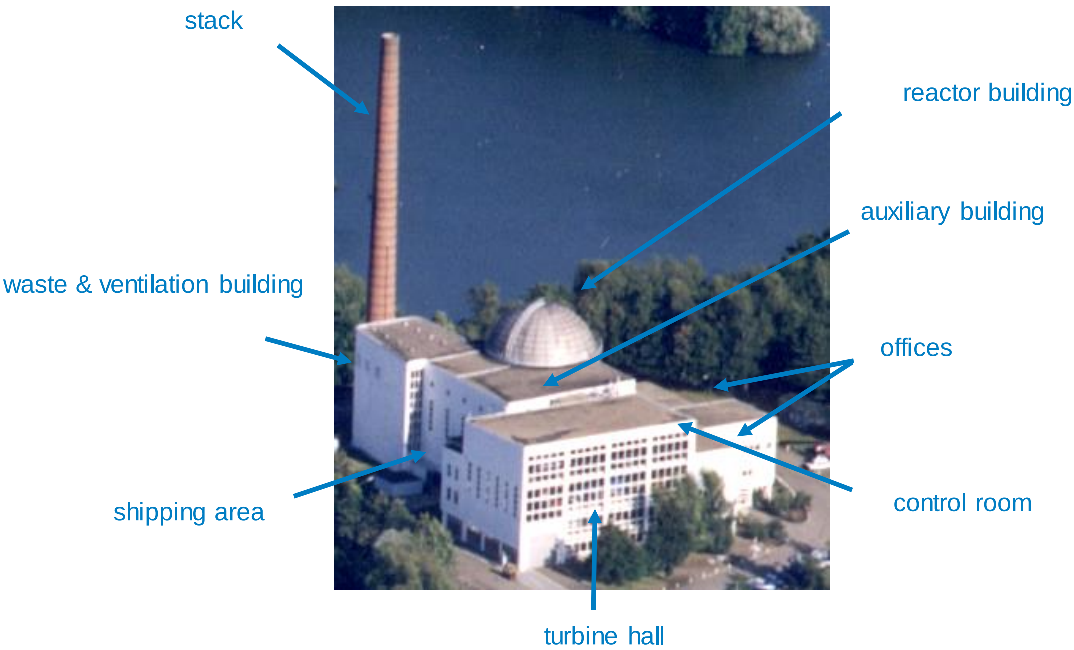

Figure 1: Aerial view of the BR3 with the indication of the main buildings

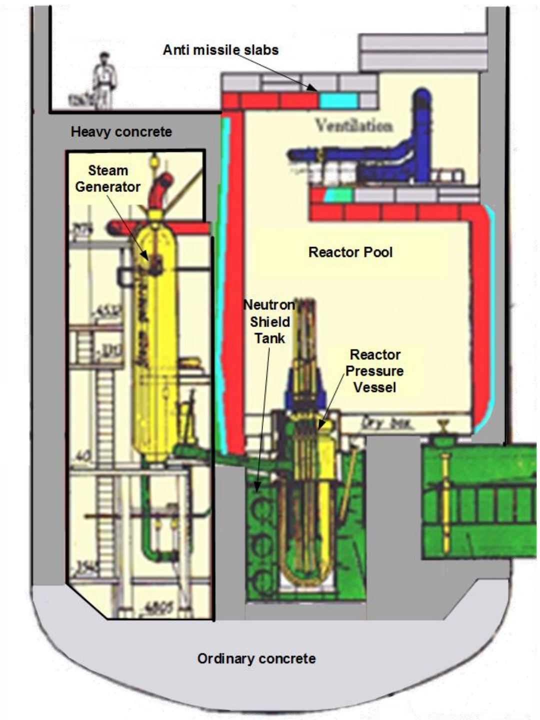

Figure 2: Cross section of the BR3 reactor building during exploitation showing the main components (RPV, NST and SG). The building structure is mainly composed of reinforced ordinary concrete (bottom plate) and reinforced heavy concrete (upper structure). The activated concrete is marked in red colour (first rough estimation).

Figure 3: Pictures of the reactor pool after dismantling the main components and before dismantling the pool liner (2016). On the bottom of the picture, we look into the NST pit. In the left corner at the back of the reactor pool, the plinth of the temporary storage bin for highly activated components is visible (left picture). The picture on the right has been taken after dismantling of this component.

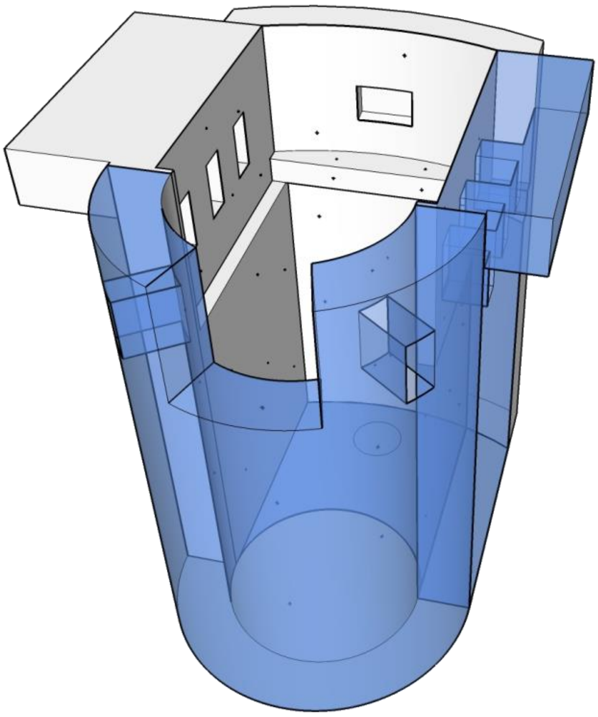

Figure 4: 3D Model of the part of the BR3 bioshield taken into consideration for the radiological characterisation programme.

2 Objectives

Figure 4 provides a model of the part of the BR3 bioshield taken into consideration for the radiological characterisation programme. The total volume is roughly estimated at about 300 m³ or 1050 tons (only walls including the area until 1 m below the bottom of the reactor pool, but excluding the anti-missile slabs and the lower part of the NST pit). According to original plans, the thickness of the walls is 1167 mm. There is no estimation on the mass or volume fraction metal/concrete. To ensure high strength and good gamma radiation shielding properties, the BR3 bioshield consists of reinforced high-density concrete (heavy weight concrete). The heavy weight concrete used at BR3 is barite concrete. This kind of concrete contains considerable amounts of barite (>40 wt.%) due to the use of $BaSO_4$ to increase the density to about 3.5 $g.cm^{-3}$.

The activation products that could be expected in the concrete are the following (IAEA, 1998), (Klein M. , 2001):

- H-3, produced by the $^{6}Li(n,\alpha)^{3}H$ reaction with a 953 barn cross section.
- C-14, produced by the activation of trace nitrogen by the $^{14}N(n,p)^{14}C$ reaction with a cross-section of 1.81 barn. Additional minor routes are via $^{13}C(n,\gamma)^{14}C$ from 1.1% abundant C-13 with a cross section of 0.9 mbarn and C-12 (98.89%, 3.4 mbarn) indirectly via C-13.
- Fe-55, produced by the $^{54}Fe(n,\gamma)^{55}Fe$ reaction in the 5.9% abundant stable iron isotope F-54 with a cross section of 2.25 barn.
- Ca-41, produced by the $^{40}Ca(n,\gamma)^{41}Ca$ reaction in the 96.9% abundant isotope Ca-40.
- Co-60, produced by the $^{59}Co(n,\gamma)^{60}Co$ reaction in the 100% abundant stable cobalt isotope Co-59 with a cross section of 18.7 barn.
- Ba-133, produced by the $^{132}Ba(n,\gamma)^{133}Ba$ reaction in the 0.097% abundant isotope.
- Cs-134, produced by the $^{133}Cs(n,\gamma)^{134}Cs$ reaction from Cs-133.
- Eu-152, Eu-154 and Eu-155 produced by neutron capture in Eu-151 (47.8%) and Eu-153 (52.2%). Other routes to the production of Eu isotopes occur because of chain absorptions in Sm. Eu-152 and Eu-154 are the two dominant europium activation products in bioshield concrete, since both have very large neutron capture cross-sections. Eu-152 is produced primarily by thermal neutrons, whereas Eu-154 also has a substantial resonance integral.

The activation products that could be expected in the reinforcement bars are the following:

- Co-60, produced by the $^{59}Co(n,\gamma)^{60}Co$ reaction in the 100% abundant stable cobalt isotope Co-59 with a cross section of 18.7 barn.
- Ni-63, produced by the $^{62}Ni(n,\gamma)^{63}Ni$ reaction in the 3.6% abundant isotope Ni-62 with a cross section of 14.2 barn.

The following radionuclides are considered to be easy-to-measure (ETM) by total gamma or gamma spectroscopy: Co-60, Ba-133, Cs-134, Eu-152, Eu-154 and Eu-155.

The following radionuclides are considered to be difficult-to-measure (DTM): H-3, C-14, Fe-55 and Ca-41.

The main goal of the radiological characterisation programme for the BR3 bioshield is to economically optimise the bioshield dismantling strategy using a waste-led approach. In order to reach this main goal, the main objective is divided into three sub objectives:

1. Create a 3D activity concentration distribution map
2. Quantify and localise the different end-stage volumes
3. Economically optimise volumes in view of a waste-led approach

2.1 Create a 3D activity concentration distribution map

The first sub goal is to create a 3D activity concentration distribution map. Figure 5 represents a typical activity concentration as a function of depth in the concrete for neutron activation. In the first centimetres in the concrete, the activity concentration increases to reach a maximum concentration at depth D_{max}. This is explained by the neutrons being thermalized, reaching a maximum interaction at distance D_{max}. Penetrating more into the concrete, the neutrons lose their energy and their ability to interact. The drop in activity concentration due to activation within depth follows an exponential curve. The relaxation length RL is the distance where the activity concentration decreases with a factor 1/e or about 0.37. In the case of the BR3 bioshield, it is not known if the D_{max} is located at the surface of the concrete or at a few centimetres depth into the concrete, and it may vary depending on the specific location. The activity concentration depth is also expected to vary with height and angle (compared to the former reactor core). Typical relaxation lengths are 8 to 15 cm.

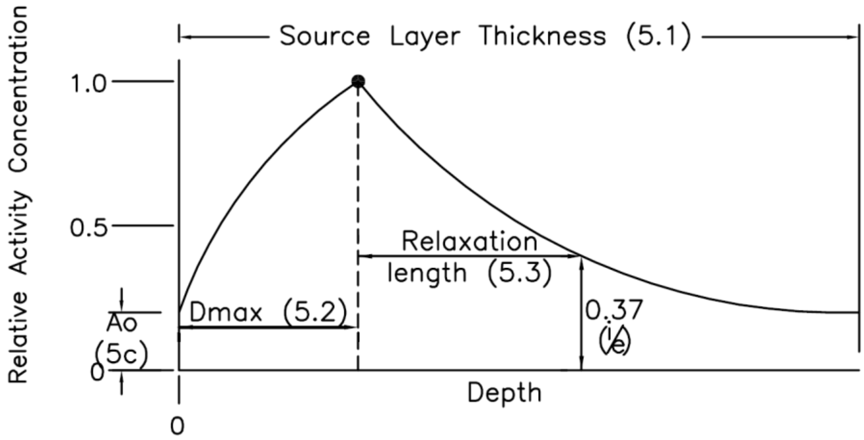

Figure 5: Activity concentration as a function of depth in the case of neutron activation

2.2 Quantify and localise the different end-stage volumes

The second sub goal is to quantify and localise the different end-state volumes.

The possible end-stages of the various parts of the bioshield are the following:

- Unconditional release according to article 35 of the Belgian legislation (FANC, 2001);
- Conditional release according to article 18 of the Belgian legislation (FANC, 2001); or
- Radioactive waste according to the waste acceptance criteria of the Belgian National Agency for Radioactive Waste and enriched Fissile Material (NIRAS/ONDRAF).

We currently do not consider the option of conditional re-use, since the total volume is too limited and this option has never been used in Belgium for this kind of materials.

Table 1 lists the radionuclides of potential importance including their half-life, activity concentration for clearance (unconditional release) and for exemption (conditional release) according to the European Basic Safety Standard Directive (EURATOM, 2013). The values will be implemented in the Belgian legislation (FANC, 2001) by 2018. The Basic Safety Standards Directive does not mention any values for Ca-41 and Ba-133. For those radionuclides, we took the rounded value 10^{x+1} for the calculated value between $3{\cdot}10^{x}$ and $3{\cdot}10^{x+1}$, mentioned in (Thierfeldt S. et al, 2012).

Table 1: Radionuclides of potential importance with their half-life (LNHB, 2018) and the activity concentration levels for unconditional release and conditional release (EURATOM, 2013). For Ca-41 and Ba-133 the rounded calculated values of (Thierfeldt S. et al, 2012) are reported.

Radionuclide	Half-life years	Activity concentration for clearance $kBq.kg^{-1}$	Activity concentration for exemption $kBq.kg^{-1}$
H-3	12.312	100	1E+06
C-14	5730	1	1E+04
Ca-41	1.002E+05	100 (*)	? (*)
Fe-55	2.747	1000	1E+04
Co-60	5.2711	0.1	10
Ni-63	98.7	100	1E+05
Ba-133	10.539	0.1 (*)	100 (*)
Cs-134	2.0644	0.1	10
Eu-152	13.522	0.1	10
Eu-154	8.601	0.1	10
Eu-155	4.753	1	100

Since we are dealing with a mixture of radionuclides, the following summation formula should be used for unconditional and conditional release:

$$\sum_{i=1}^{n} \frac{c_i}{c_{li}} < 1.0$$

Where

c_i is the activity concentration of radionuclide i (Bq/g),

c_{li} is the activity concentration for clearance of radionuclide i in case of unconditional release (Bq/g) or the activity concentration for exemption of radionuclide i in case of conditional release (see Table 1),

n is the number of radionuclides in the mixture.

In the above expression, the ratios of the concentration of each radionuclide to the clearance (or exemption) level is summed over all radionuclides in the mixture. If this sum is less than one, the material complies with the requirements for unconditional (in case of exemption levels of conditional) release. In case of a sum above 1 for conditional release (exemption levels), the concrete has to be considered as radioactive waste. Furthermore, the radionuclides considered can be limited to the most important ones, that together make up 99% or more of the total sum.

The current strategy for conditioning as radioactive waste would be to directly cast the concrete in monoliths. This option is however not available yet. The only current available solution would be to dispose the concrete in 200-L drums for super compaction at Belgoprocess. For the BR3 bioshield concrete being considered as radioactive waste disposed of in monoliths or 200-L drums, there would be no problems with physical or radiological aspects.

2.3 Economically optimise volumes in view of a waste-led approach

The third sub goal is to optimise volumes for the three possible end-stages in view of a waste-led approach. In terms of waste costs, we take the unit prices for the following end-stage options into account: unconditional release, conditional release and radioactive waste (super compaction 200-L drums, monoliths). For the determination and localisation of the three end-stage volumes, we furthermore consider the following directions:

- The probability to exceed the threshold should be less than 5%, for demonstrating compliance with the threshold;

- The threshold value can be considered as an average value for a mass of 1 ton. For the BR3 bioshield (density concrete of 3.5 g/cm^3) this means that we can average over cubes of 0.66 x 0.66 x 0.66 m.

The effect of in-situ and ex-situ decay storage of respectively 10 and 30 years could be envisaged as a potential optimization in terms of waste costs and dismantling strategy/costs.

We need to take potential practical constraints in view of, and costs related to, dismantling and separation of end-stages materials into account.

3 Constraints

Decommissioning is usually about finding a balance between safety and budgetary issues. For this specific characterisation programme, there are however some specific additional constraints.

3.1 Safety

The expected activation levels are relatively low (maximal activity concentrations up to 50 Bq/g). We expect that the radiation levels after removing the liner are very limited (100 nSv/h up to a few µSv/h). As the activity concentrations are low, the contamination hazard is relatively low as well. Due to the presence of reinforcement bars with diameters of 32 mm, the sampling will be performed by wet core drilling reducing the contamination risk. The technique is very well known. Standard precautions for radiation and contamination hazards will be sufficient.

Important hazards are related to conventional safety aspects (working at height). Access to the major part of the walls of the reactor pool (> 10 m height, excluding the depth of the RPV/NST pool) is secured through a movable platform, attached to a hoisting structure. Therefore, the platform is not stable and can only be operated by qualified staff. The total maximum working load on the platform is 520 kg (max. 3 people & 250 kg). For accessing the bottom part of the reactor pool, a specific procedure needs to be drawn up and approved by the internal safety department. All working procedures are subject to an approval by the internal safety department.

Figure 6: Platform installed into the reactor pool

3.2 Planning & Budget

Planning and budgetary restrictions limit the number of core drilling samples to 30. The number of gamma spectrometry measurements per core will be limited to 10.

3.3 Other

In situ (non-destructive) measurements are only possible on the inner or outer surface of the reactor pool walls. Moreover, acquiring results in terms of activity concentrations is extremely challenging due to the activity distribution profile depending on the depth and angle.
For sampling and ex-situ measurements the amount (mass, volume, number) is limited compared to the total volume of the bioshield. Sampling will be hampered by the presence of thick reinforcement bars. Due to the wet drilling, precautions are needed to prevent cross contamination. The order of sampling would be preferable from low activity concentration to higher activity concentration.

4 Pre-existing data

We dispose of the following pre-existing data:

- Original 2D plans and 3D models based on the original 2D plans (not "as-built");
- Operational history;
- Results from neutron activation calculations; and
- Results from historical sampling and characterisation programmes

4.1 Plans and models

Table 2 lists the original plans of the BR3 reactor building. Based on those plans various 3D models have been built (e.g. screenshot of Figure 4 using Sketchup). We will use the language and environment for statistical computing and graphics "R" (https://www.r-project.org/) for the activity distribution calculations and mapping.

Table 2: Original plans Reactor Building

Letter	Number	Comment
TE	100 382	Bottom reactor building
TE	100 392	Bottom reactor building reinforcement
TE	100 399	-4.8 up to +1.6 m
TE	100 481	+0.2 up to +16.5 m
TE	100 482	+0.2 up to +16.5 m
TE	100 483	Reinforcement of 100 482
TE	100 556	Openings reactor pool
TE	100 333	Work phases
TE	100 332	Work phases (openings in operating deck)
TE	100 475	Edge
TE	100 478	Salbs 2-14
TE	100 479	Slabs 15-27
TE	100 331	Floor (galery incl.)
TE	100 474	Legs passages NST
TE	100 418	Framework Operating Deck
TE	100 651	Plugs manholes Operating Deck
TE	100 359	Drainage NST
Cockerill Ougrée	50 406	Refueling Channel
Cockerill Ougrée	50 407	NST piping
Cockerill Ougrée	50 408	Refueling Channel piping
Cockerill	904 438	Embedded piping
TE	100 393	Intermediate floors 0 – 0.53 m
TE	100 409	Intermediate floors -3.505 m
TE	100 410	Intermediate floors 0.0 m
TE	100 451	Intermediate floors +3.353 m
TE	100 566	Intermediate floors 7.214 m
	15143/05/105	Rail 40 T portal crane
	15107/852	Trolley 40T/ Frame 40T
	15142/07/144	40 T skid
	416 618	Extraction tubing ventilation

4.2 Operational history

The BR3 bioshield is equipped with a stainless steel liner of 3 mm thickness to secure water tightness (see Figure 3). Historical records do not report any incident regarding potential leakages; nor during reactor operation, nor during dismantling and cutting activities. Also during the removal of the liner, no signals of potential leakages have been found. Before removing the liner, we performed a cleaning operation followed by a radiological characterisation. Apart from the Co-60 activation, we only detected limited traces of Am-241 and Cs-137 (see §4.4), probably as a result of the cutting of the main reactor components. We assume that there is no contamination behind the liner.

4.3 Results from neutron activation calculations

In order to have an idea of the potential radionuclides being present and the activity concentration levels concerned, several calculation campaigns were performed:

- (Vincent, 1995) and (Aït Abderrahim, 1996) compared calculated Ba-133 activity concentrations in the lateral anti-missile slabs using the Monte Carlo neutron transport code Tripoli 3.2 with actual sample measurements.
- (Smaizys, 2006) focussed on the neutron shield tank and bioshield and provided results for various radionuclides (H-3, C-14, Cl-36, Ca-41, Fe-55, Co-60, Ba-133, Cs-134, Eu-152 and Eu-154) for two different neutron fluxes: 7.18E+06 n/(cm².s) at approximately 300 cm height and 1.98E+10 n/(cm².s) at 400 cm height.
- (Klein M. , 2001) defined the scaling factors; and
- (Aoust, 2008) attempted to produce a 3D activation distribution map based on neutron activation calculations.

From the different neutron activation calculation exercises, we notice the following:

- The activity concentration obtained differs considerably from sample measurements for the main radionuclide present (Ba-133), while Ba is one of the main elements present in the concrete.
- Ratios of activity concentration of other radionuclides to Ba-133 considerably differ between different calculations, considering different chemical compositions of concrete (trace elements).

The following chapter reports on the comparison of the results of neutron activation calculations with the results of sample measurements in view of end-stage objectives.

4.4 Results from historical sampling and characterisation programmes

In view of the general decommissioning planning, various measurement programmes have been performed, such as the removal of the anti-missile slabs, creation of an access for the decontamination of the steam generator, removal of the hot leg and removal of the pool liner. In order to get a first idea on the activation of the concrete, we organised two specific sampling and characterisation programmes. Various sampling and characterisation programmes have been performed:

- ***Removal of anti-missile slabs (1994)***: the dismantling team removed the anti-missile slabs in the '90s (Klein M. , 1991) (Mandoki, 1994). During operation, they served as a protection in case of a missile incident (e.g. RPV lid).

- ***Bioshield sampling programme 1 (1996)***: a first specific bioshield sampling and characterisation campaign took place in 1996. A total of eight core drillings were taken on the cylindrical part of the bioshield. After cutting the core into pieces of 70-90 mm length, the SCK•CEN Low-Level Radioactivity Measurements Lab measured each piece individually (Mandoki, 1996).
- ***Creation of an opening for the decontamination of the steam generator (2001)***: in order to decontaminate the Steam Generator (SG) it had to be lifted and placed horizontally in the reactor building (see Figure 7). For this reason the dismantling team created an opening in the bioshield, centrally on the top of the circular wall (see also Figure 4). (Plateau, 2001) describes the instruction to create the opening and (Plateau, 2002) reports on the results.

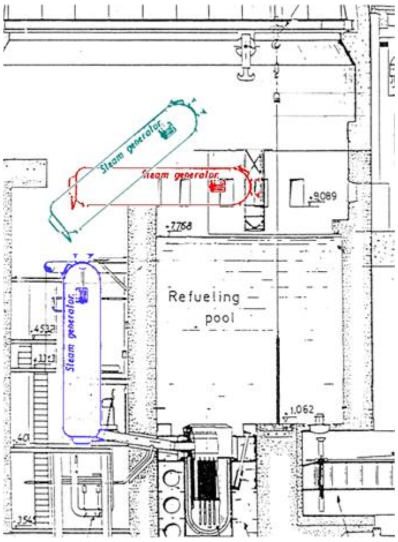

Figure 7: Plant container: in order to decontaminate the SG it had to be lifted and placed horizontally.

Figure 8: Block 10 resulting from opening in the bioshield that was created to decontaminate the SG. The block was ex-situ measured using the ISOCS. A sample has been taken by core drilling and was analysed.

- ***Bioshield sampling programme 2 (2008)***: a second specific sampling and characterisation campaign took place in 2008 as result of a thesis (Piccini, 2006). In this campaign seven cores drillings were performed on the two lateral walls and one core was taken on the circular wall close to the hot leg. Due to time restrictions only a part of the samples have been analysed using a NaI detector.

- ***Removal of the hot leg (primary loop, 2012)***: in 2012, the dismantling team removed the hot leg and shutdown tube from the bioshield by 36 core drillings (Figure 9). The cores have not been drilled following the radius of the RPV nor perpendicular to the rounded wall of the bioshield. They were drilled horizontally and parallel to the surface of the hot leg and the shutdown tube. Some of the cores were sliced using a stonecutter and analysed by gamma spectroscopy (determination Co-60, Ba-133 and Eu-152; traces of Eu-154 were below the MDA) using the ISOCS (In Situ Object Counting System). The results show a very specific activation profile in the volume surrounding the hot leg (Verstrepen, 2013).

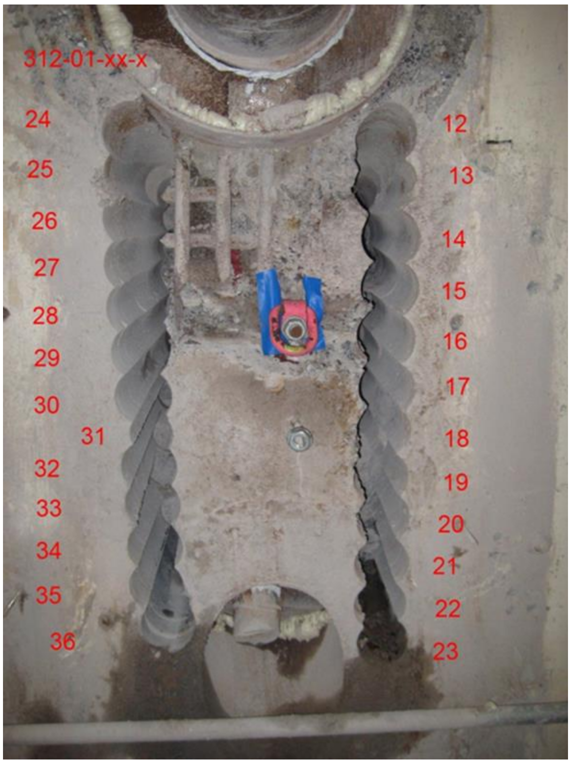

Figure 9: Core drillings for the removal of the hot leg and shutdown tube. A total of 36 core drillings have been performed.

- ***Bottom of the reactor/NST pit (2016)***: As indicated in Figure 10, we took 3 samples from the reactor/NST pit in 2016 and analysed them using the ISOCS (Verstrepen, 2016). No traces of Ba-133 and Eu-152 were found. The main radionuclide measured was Cs-137 including traces of Co-60 and Am-241, probably all as a result from contamination.

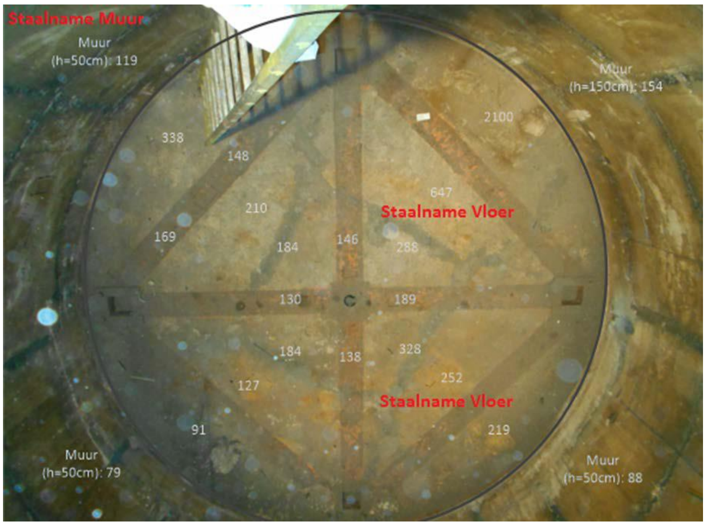

Figure 10: Sampling from the reactor/NST pit: two samples have been taken from the bottom and one sample has been taken from the wall (marked in red colour)

- ***Removal of the pool liner (2018)***: in view of a waste led approach for the reactor pool liner, we organized a sampling and characterisation campaign before removing it. Circular disks were taken using a crown drill bit. The samples were analysed by gamma spectroscopy using the ISOCS. The Co-60 activity concentration distribution of the stainless steel pool liner gives a good idea on the (relative, not absolute) activation of the concrete just behind it. The distribution appeared to be relatively symmetric, apart from the liner behind the former temporary storage bin for highly activated components, where the activation appeared to be lower. (Verstrepen, Staalnamecampagne april 2017: BR3_Plan Container_Lokaal 087, Inox wand piscine, Analyserapport, 2017).

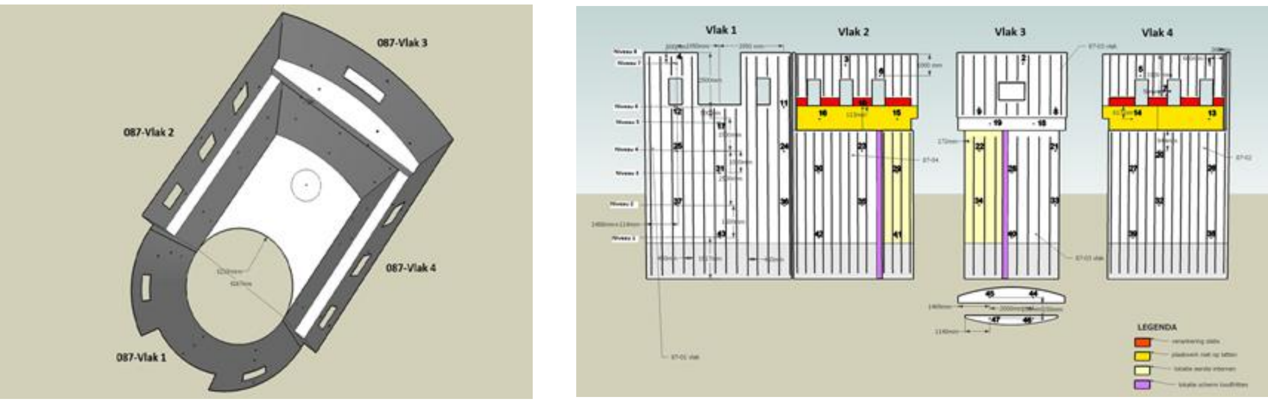

Figure 11: Sampling from the pool liner walls. Four walls are identified (left) and a total of 47 samples have been taken using grid sampling (right)

Results of neutron activation calculations and sample measurements in view of end-stage objectives: theoretical assumptions and various neutron activation calculations resulted in the identification of the following radionuclides of importance:

- ETM: Co-60, Ba-133, Cs-134, Eu-152, Eu-154 and Eu-155.
- DTM: H-3, C-14, Ca-41, Fe-55 and Ni-63.

For the ETM, Cs-134 and Eu-155 should not be taken into account due to their initial low abundance and short half-lives. Especially older measurements showed traces of Eu-154. Applying the summation formula (see chapter 2.2) on 1 January 2020, Eu-154 will clearly contribute less than 1% compared to Ba-133, Eu-152 and Co-60 and therefore it should not be taken into account according to (FANC, 2001). The DTM radionuclides H-3, C-14, Ca-41 and Fe-55 would contribute much less than 1% applying the sum formula. The Ni-63 is expected to be present in the reinforcement bars. Based on neutron activation calculations we expect a Ni-63/Co-60 ratio of about 2 (1 January 2020). Various sample measurements showed a ratio of about 4. It should be only necessary to start taking the contribution of Ni-63 into account for release purposes from the year 2027 on.

We conclude that for the end-stages of unconditional release and conditional release (see chapter 2.2) the only radionuclides to be taken into account are:

- Co-60, Ba-133, Eu-152 for the activated concrete
- Co-60 for the reinforcement bars (only from the year 2027 on, we should start to take Ni-63 into account as well).

Moreover, the Co-60 concentration in the reinforcement bars is about 5 times lower than the Ba-133 concentration in the concrete at the same location on 1 January 2020. This means that, when the Ba-133 concentration would be below the clearance level in the concrete at a specific location, this will be as well the case for the Co-60 in the reinforcement bars at the same location.

5 Preliminary data analysis based on pre-existing data

Although the pre-existing data is rather heterogeneous as it results from different campaigns with different goals, the amount of information and consistency seems sufficiently adequate to justify performing a preliminary analysis for informing the sampling design. The analysis has been performed following the strategy described in (Rogiers, B., Boden S., Pérot N., Desnoyers Y., Sevbo O., Nitzsche O., 2018), and we follow and discuss the different steps from the flow charts here, and the choices made based on the data analysis and sampling design methods diagrams.

The main objective we try to address with this preliminary analysis is the first one, *i.e.* the estimation of the 3D activity concentration distribution. The other objectives were considered not to be of major importance at this stage.

5.1 Pre-processing

The pre-processing of the data basically consisted of three different aspects:

1. The 3D location of the different measurements was not directly recorded, and had to be inferred based on the 3D model of the bioshield, and the recorded positions in terms of 2D coordinates for the specific concrete elements, the angles of the boreholes and the depth of the samples with respect to the inner or outer surface of the bioshield. As this is not straightforward given the complexity of the 3D geometry of the bioshield, it required different iterations between the 3D model and the data analysis experts. Both the liner and the borehole sample locations are shown in Figure 12.
2. The 3D geometry of the bioshield itself had to be converted into an effective format that would allow efficient estimation of the activity concentrations across the entire volume. We opted for using regular grids, with point locations separated by a constant distance d in x, y as well as z directions. In this way, estimates could be made on a point-by-point basis, and every point is approximately representative for a volume of d^3. We provide an overview of different grids in Figure 13. Of course the coarser grids were used for model development, prototyping and testing purposes, while the final calculations were always performed for the 5 cm spacing.
3. To handle the different times since measurement of activity concentrations, all values were rescaled, so they represent the activity concentrations at January 1, 2020.

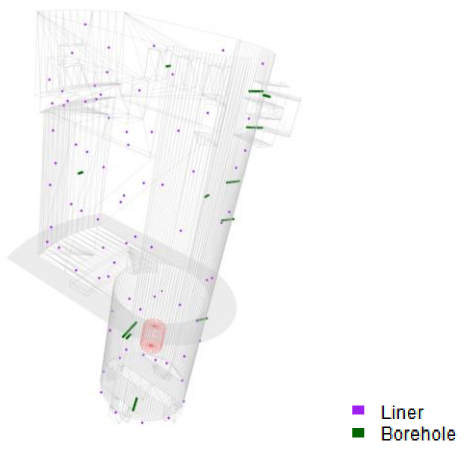

Figure 12: Overview of the liner and borehole sample locations, corresponding to the measurement results used in the preliminary data analysis.

Figure 13: Different regular grids used for estimating the activity concentrations. From left to right: 50, 25, 10 and 5 cm spacing, resulting in 3174, 28239, 420449 and 3354028 points.

Other outliers and/or errors were not present at first sight, and the representativeness of the data was judged to be sufficiently adequate for a first analysis., Although the boreholes were focused on one side of the bioshield, the liner data covered all walls, so at least some information (even if just secondary) on all parts of the bioshield was available.

5.2 Exploratory data analysis

During the exploratory data analysis, we focused on the multivariate aspect, and the potential relation between the liner activity concentrations and those in the concrete. It was already very clear from the start that there would be a complex trend related to the distance from the neutron source, and the depth in the concrete (see *e.g.* Figure 5), and that characterization of this trend would be the main aim in this phase. If any spatial structure would be present, on top of the trend, it would not be obvious from the limited data in this case, and hence this was not further investigated. Furthermore, robust approaches were considered out of scope as well.

For the borehole analysis, an idea on the missing data and the pairwise correlations is provided in Figure 14. This clearly illustrates that we have Ba-133 results at all locations, but the other remaining radionuclides are not always available. The scatterplots and correlations on the other hand suggest that, certainly in this stage, we can focus on estimating Ba-133 activity concentrations, and derive the other values from this (*i.e.* using simple linear regression for the log-transformed values). Hence, we decided to fall back to a univariate problem, at least as far as the borehole data is concerned.

Since the liner data is more systematically distributed over the inner surface of the bioshield, we of course tried to account for it in this stage. In a later stage, when a similar distribution of borehole measurements would be available, accounting for this secondary data will of course not be so useful anymore. Indeed, from theory, we expect that the distribution of *e.g.* Co-60 activity in the liner provides some information on at least the relative distribution of Ba-133 activity concentrations at the concrete surface. Given the different spatial distribution of the liner and borehole data, it is however difficult to confirm this by an exploratory data analysis. We therefore do not discuss this relation here, but explain it as part of the workflow for the actual data analysis.

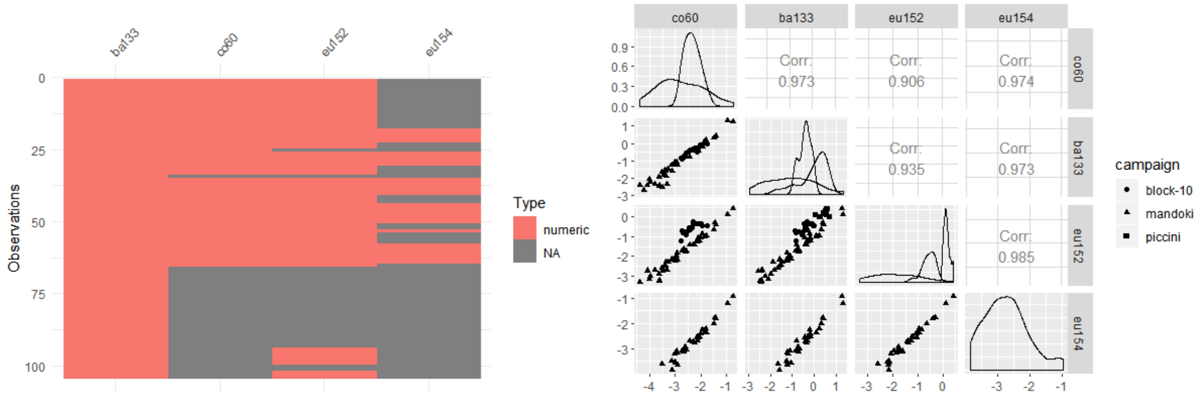

Figure 14: Overview of the borehole data. Left: indication on missing values. Right: scatterplot matrix of the logarithmic (base 10) activity concentrations.

5.3 Data analysis

We are thus currently focussing on modelling the complex trend of Ba-133 activity concentrations, and therefore we fall back, in accordance to the strategy from D3.2, to the class of generalized linear models and potentially generalized additive models if the former would not provide sufficient flexibility. Different features are available for use as regressors in this case. We considered of course the spatial coordinates, including the absolute x coordinate because of the expected symmetry. Also the distance to the reactor fuel, the depth within the concrete, the spatial coordinates of the point projected to the inner surface of the bioshield, and the corresponding liner Co-60 activity concentration were considered as potentially informative.

The liner Co-60 activity concentration is of course only available at the measurement locations. Therefore, we used a generalized additive model to basically interpolate these measurements on the inner surface of the bioshield, as a smooth function of the projected x and z coordinates, and the corresponding distance to the fuel. A linear model did not suffice, because of the non-linear relation of the Co-60 activity concentration with some of the considered regressors. The result is illustrated in Figure 15.

For similar reasons, the trend modelling for the Ba-133 activity concentrations was done with a generalized additive model as well, using a smooth function of the liner Co-60 activity concentration and the depth within the concrete. The results of this model are illustrated in Figure 16. A comparison with the measurement data is provided in Figure 17. Although there are clearly still issues to be addressed, this already seems to provide an idea on the order of magnitude to expect across the entire volume of the bioshield, which is very useful for informing the sampling design.

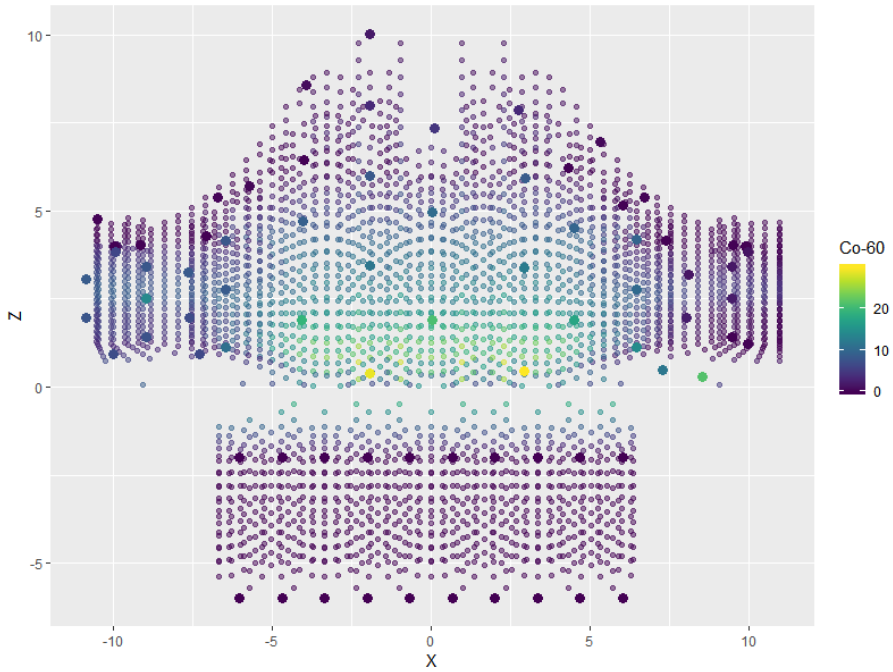

Figure 15: Overview of the liner Co-60 activity concentrations (large, non-transparent points) and the predictions of the generalized additive model (small, transparent points) for different points within the bioshield, based on an unwrapped projection to the inner surface of the bioshield.

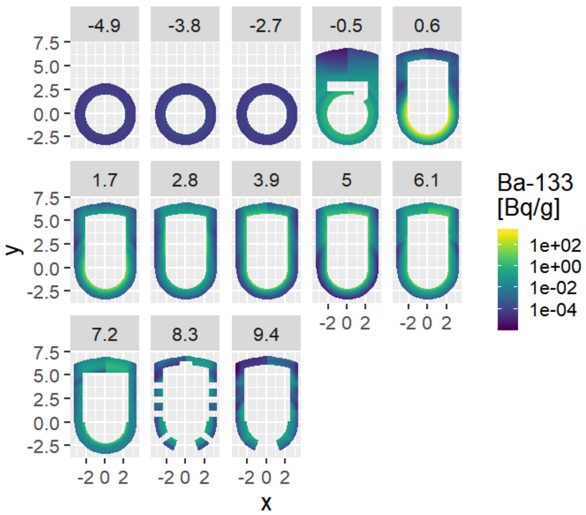

Figure 16: Series of horizontal slices through the preliminary Ba-133 3D activity concentration model, for different z coordinates.

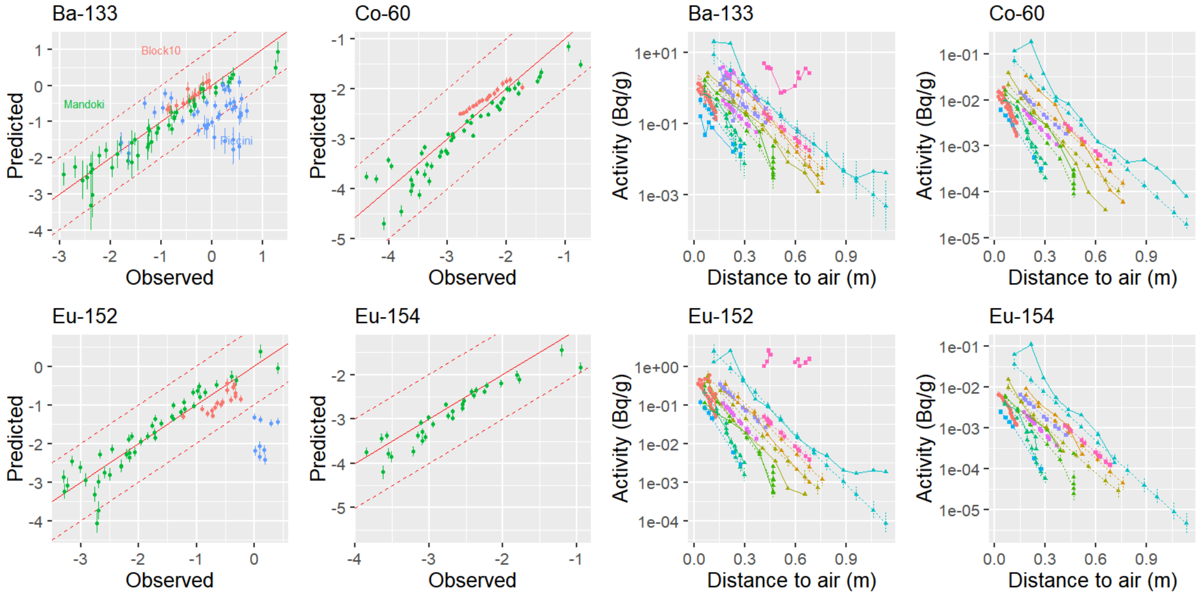

Figure 17: Overview of the predicted/fitted versus the observed data (for the logarithmic (base 10) transforms) (left four plots) and the results for the different boreholes, with observed values in solid lines and predictions in dashed lines (right four plots).

5.4 Post-processing

As the main output of the data analysis is the individual radionuclide activity concentrations across the entire bioshield, no post-processing is required for the first objective. As mentioned above, an initial very rough estimate of end-stage volumes was made, but discussing the outcome is out of the scope of this document.

5.5 Achievement of the objectives

As the data on which this analysis is based is very limited, and *e.g.* a proper quantification of the uncertainties on the results was not even considered relevant at this stage, it is very clear the objectives are not achieved at this point. The preliminary analysis just serves the purpose of informing the sampling design.

6 Sampling design

After removing the liner, the SCK•CEN performed an in-situ total gamma surface mapping (June 2018), consisting of about 300 individual measurements using a Como 300G plastic scintillator of 300 cm² surface. This roughly amounts to about one measurement per square meter. We used regular grid sampling (Figure 18) to achieve full coverage, as the idea is to use these data as secondary information for the activity concentrations within the concrete, in a similar way as how the liner data was used for the preliminary data analysis.

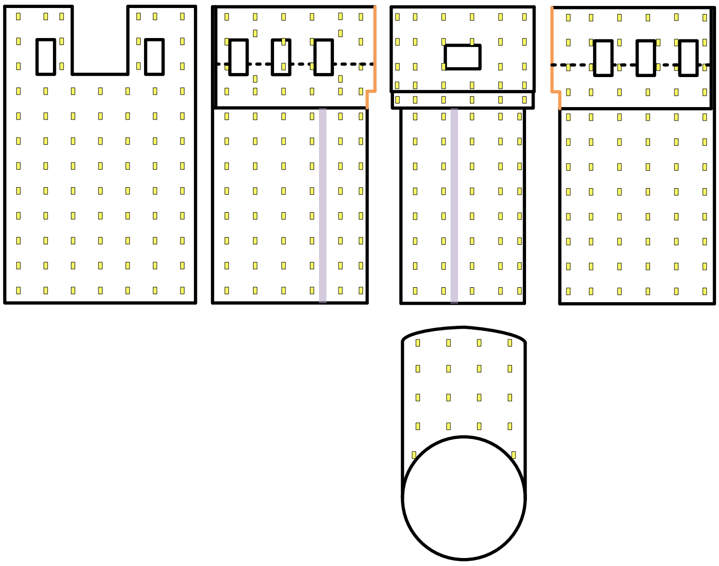

Figure 18: In-situ total gamma surface mapping of the inner pool (top) walls and floor (bottom). The yellow rectangles indicated the measurement locations

Following the basic principles described in (Rogiers, B., Boden S., Pérot N., Desnoyers Y., Sevbo O., Nitzsche O., 2018), the sampling design mainly consisted of systematic sampling (equal probability of selection/probabilistic) supplemented with judgemental selected sampling locations (specific structures such as the “poubelle” and the refuelling channel and close to the location with the maximal activation level). In addition, the expected trend extreme locations were selected as well, and we rely on the symmetry of the activation to maximize the results with a minimum number of samples. Figure 18 shows the sampling plan.

This combination of sampling approaches basically ensures that:

1. We cover all the concrete elements, to reduce the risk of missing anything,
2. We include (approximately) the minimum and maximum values across the entire bioshield, but also within every element, to reduce the required amount of extrapolation during the data analysis,
3. We investigate specific features for which it is known that they deviate from the general trend.

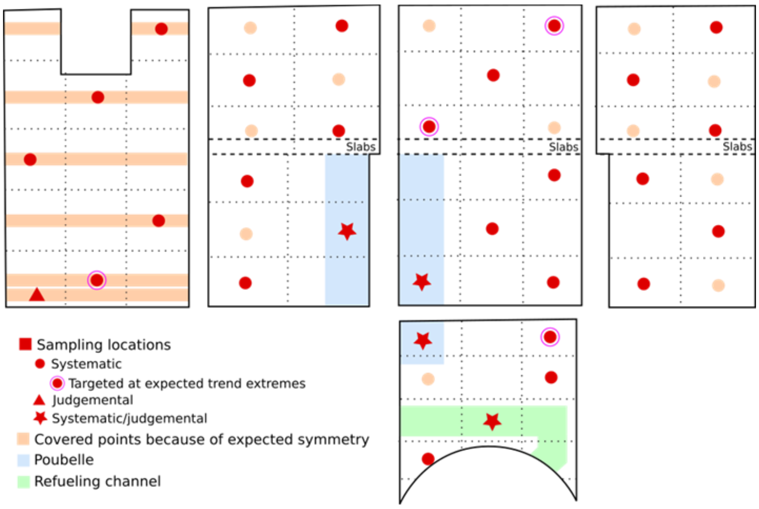

Figure 19: Sampling plan for the BR3 biological shield (30 samples).

As described in chapter 3.2 we foresee to take 30 samples by wet core drilling. The cores (diameter 72 mm, length of about 90 cm down the first outer reinforcement bars) will be segmented and the segments will be analysed by gamma spectroscopy. The results will be used for further data analysis and checking the objectives. The first selection of segments to be analyzed is based on the results of the preliminary data analysis. That is, the focus is on quantifying the absolute levels of Ba-133 activity concentration, and the trend with depth in the concrete. Furthermore, outer parts of the bioshield that seem to be orders of magnitude below the threshold for unconditional release can be omitted for now.

In addition to the 30 samples, 6 cores (diameter 72 mm, length 30 cm) will be taken and transported to the National Physical Laboratory for homogenisation and distribution to other EU laboratories. They will serve as base material for the benchmarking exercises within INSIDER WP6. Benchmarking results will serve as an input for the calculation of the global measurement uncertainty within WP6. Apart from a location at high and medium activity, the exact location of the 6 additional samples is not important (Peerani, Boden, Crozet, & Zanovello, 2018). The locations are not indicated in Figure 18.

7 Integrated management arrangements

The sampling plan is performed in accordance with the SCK•CEN Integrated Management System (Safety, Security, Health, Environment and Quality, https://imsportal.app.sckcen.be/#/) and more specifically the SHEQ-QA manual of the "Dismantling, Decontamination and Nuclear Waste" (DDW) expert group (Demeulemeester, 2018). The sampling is performed in accordance with the scope of the ISO 9001 certification of the DDW expert group. The following main standard operating procedures are involved:

- Dismantling, decontamination and nuclear waste (DDW-SOP-01);
- Characterization measurements DDW (200-SOP-01);
- DDW Dismantling (119-SOP-01); and
- DDW Treatment of material (121-SOP-01).

Specific issues are described in underlying documents such as:

- 119-SUP-834/14-19 (Organisatorische aspecten mbt het uitvoeren van een werf)
- 119-SUP-831/11-33 (Kleine ontmantelings- en niet reguliere onderhoudswerken op BR3)
- 119-SUP-831/12-04 (Strategie voor de sanering van de gebouwinfrastructuur BR3)
- 119-SUP-831/12-05 (Decontaminatie en ontmanteling van beton)
- 200-INS-831/11-16 (Staalname in betonnen constructie d.m.v. watergekoelde kernboring)

The samples will be analysed by the SCK•CEN Low-Level Radioactivity Measurements Laboratory within the scope of their ISO 17025 accreditation (133-SOP-1000: Determination of radioactivity by gamma spectrometry).

8 Bibliography

Aït Abderrahim, H. (1996). *Assessment of the BR3 concrete building activation using the Tripoli Monte Carlo transport code, Topical meeting on Radiation Protection and Shielding, No Falmouth Massachusetts, USA, Proc. 576-583.* American Nuclear Society.

Aoust, T. (2008). *Evaluation neutronique de l'activation du béton autour du réacteur BR3, Situation 30/04/2008, Internal note.* SCK-CEN.

Demeulemeester, Y. (2018). *SHEQ - QA manual DDW, SCK-CEN/1259373 Rev. 1.1, DDW-SHEQ-02.* SCK-CEN.

EURATOM. (2013). *Council Directive 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom a.* Official Journal of the European Union.

FANC. (2001). *Koninklijk besluit van juli houdend algemeen reglement op de bescherming van de bevolking, van de werknemers en het leefmilieu tegen het gevaar van de ioniserende stralingen (ARBIS).* Belgian Official Gazette.

IAEA. (1998). *Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes, Technical Reports Series 389.* Vienna: IAEA.

Klein, M. (1991). *Contamination of the anti-missile slabs, Technical note.* SCK-CEN.

Klein, M. (2001). *Flux des déchets du BR3, Méthodologie de charactérisation (version 3 de l'ancien 164/99-02), 164/01-01.* Mol: SCK-CEN.

LNHB. (2018). *Nucléide - Lara, Library for gamma and alpha emissions, http://www.nucleide.org/Laraweb/index.php.* Laboratoire National Henri Bequerel.

Mandoki, R. (1994). *Caractérisation des dalles anti-missiles du BR3, Note technique RM/35/94-74.* SCK-CEN.

Mandoki, R. (1996). *Rapport de la caractérisation du bouclier biologique du BR3 (partie cylindrique), Internal report R-3107.* SCK-CEN.

Peerani, P., Boden, S., Crozet, M., & Zanovello, F. (2018). *Design of the benchmarking exercise, Deliverable 2.5, INSIDER H2020 project.* EC.

Piccini, G. (2006). *Gestion des bétons activés du BR3, Master 2 ITDD GéDéRA.* SCK-CEN.

Plateau, C. (2001). *Scabbling et découpe du béton de l'Operating Deck (11,475 m) au-dessus du Générateur de Vapeur et du Pressuriseur, Instruction de travail - démantèlement, 117/BR3/VS-419.* SCK-CEN.

Plateau, C. (2002). *Rapport de chantier 118/419-01.* SCK-CEN.

Rogiers, B., Boden S., Pérot N., Desnoyers Y., Sevbo O., Nitzsche O. (2018). *Improved nulcear site characterization for waste minimization in decommissioning and dismantling operations under constraint environment. INSIDER WP3 - Sampling strategy, Report on the sampling strategy development. Deliverable D3.2 pp39.* H2020 INSIDER D3.2.

Smaizys, A. (2006). *Calculation of neutron shield tank activation level and waste management + ALARA planning (optional), internal report.* SCK-CEN.

Thierfeldt S. et al. (2012). *Berechnung von Freigrenzen und Freigabewerten für Nuklide, für die keine Werte in den IAEA-BSS vorliegen, 434.0000-101/11.007303/7918731.* Brenk Systemplanung.

Verstrepen, G. (2013). *Kernboorstalen 312-01 t.o.v. warm been - shutdown leiding: gebariteerde beton, Interne mem, 160/13-01.* SCK-CEN.

Verstrepen, G. (2016). *Cement-/betonlaag, herkomst: BR3_put NST (2 x vloer, 1 x wall), Analyserapport.* SCK-CEN.

Verstrepen, G. (2016). *Cement-/betonlaag, herkomst: BR3_put NST (2 x vloer, 1 x wall), Analyserapport, 165-16/05.* SCK-CEN.

Verstrepen, G. (2017). *Staalnamecampagne april 2017: BR3_Plan Container_Lokaal 087, Inox wand piscine, Analyserapport.* SCK-CEN.

Vincent, T. (1995). *Evaluation de l'activation des bétons du bâtiment réacteur BR3 à l'aide d'un code de transport neutronique.* DEA Radioéléments Rayonnements Radiochimie.