



Horizon 2020  
Programme

***INSIDER***

*Research and Innovation Action (RIA)*

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 755554.

Start date : 2017-06-01 Duration : 48 Months  
<http://insider-h2020.eu/>



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**Criteria for characterization, RN & materials-cartography**

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INSIDER - Contract Number: 755554

Improved Nuclear Site characterization for waste minimization in DD operations under constrained Environment

Document title	Criteria for characterization, RN & materials-cartography
Author(s)	Mr. Khalil AMGAROU
Number of pages	75
Document type	Deliverable
Work Package	WP02
Document number	D2.2
Issued by	CEA
Date of completion	2017-11-24 16:22:09
Dissemination level	Public

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## Summary

The present deliverable is focused on the initial characterisation of nuclear facilities subject to a decommissioning programme, in order to determine the physical, chemical and radiological properties of their structures and equipment. It also deals with the cartography of plants and wide-areas to map their associated radiation and contamination levels. The document is based on a literature review of several reference publications, namely the ones issued by the International Atomic Energy Agency in this particular field of interest. It likewise discusses the information gathered by means of a questionnaire that has been submitted to several partners, experts and end-users from different EU member states, plus Japan and Ukraine, with a consolidated experience in the domain. Although the focus here is on the radiological aspect as it is the key differentiator between conventional and nuclear dismantling activities, other hazardous substances exhibiting chemical toxicity are briefly addressed. Efforts have been made to keep this report in a clear and concise manner. The interested reader can consult the cited bibliography for further information about the multi-disciplinary aspects related to nuclear decommissioning.

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## **Summary**

The present deliverable is focused on the initial characterisation of nuclear facilities subject to a decommissioning programme, in order to determine the physical, chemical and radiological properties of their structures and equipment. It also deals with the cartography of plants and wide-areas to map their associated radiation and contamination levels. The document is based on a literature review of several reference publications, namely the ones issued by the International Atomic Energy Agency in this particular field of interest. It likewise discusses the information gathered by means of a questionnaire that has been submitted to several partners, experts and end-users from different EU member states, plus Japan and Ukraine, with a consolidated experience in the domain. Although the focus here is on the radiological aspect as it is the key differentiator between conventional and nuclear dismantling activities, other hazardous substances exhibiting chemical toxicity are briefly addressed. Efforts have been made to keep this report in a clear and concise manner. The interested reader can consult the cited bibliography for further information about the multi-disciplinary aspects related to nuclear decommissioning.

## 1 Introduction

Decommissioning of nuclear facilities (power reactors, fuel cycle plants, research or medical accelerators, etc.) refers to the final step, after shutdown, in their life cycle and covers the whole process whereby the considered site is properly dismantled and its near environment is cleaned up to a predetermined endpoint (unrestricted release or further reuse), from any dangerous and radioactive substance (IAEA, 2016; NEA, 2014). It is a long, expensive, complex and non-revenue-generating activity with a multidisciplinary nature (Laraia, 2012), thus representing a global challenge for the XXI<sup>th</sup> century as an increasing worldwide demand in this industrial sector is expected for the next years (NEI, 2016) whereas the lessons learnt from past experiences are still limited<sup>1</sup>. In addition, the followed strategy is specific to each country depending, amongst other considerations, upon the facility's characteristics, own regulatory policies, environmental protection requirements and availability of waste disposal routes (CSW, 2013).

For this purpose, it is undoubtedly essential to act on the upstream stage of a given decommissioning programme for the optimum definition of viable and cost-effective dismantling scenarios as well as for the safe classification and segregation of all radioactive wastes. This constitutes a complex issue considering the wide variety of involved structures and equipment, so that a proper characterisation of these latter become a necessary precondition for a successful quantification of the different contaminated materials (IAEA, 1998).

According to the latest ISO<sup>2</sup> standard that have been recently published in this domain (ISO, 2017), knowledge of the physical and radiological condition of a nuclear facility subject to a decommissioning programme is the major characterisation goal. Nevertheless, although contributing to meet the corresponding remediation or decontamination objectives, characterisation must also consider other specific targets such as complying with Health & Safety requirements during clean-up and dismantling activities, mastering the estimation of radioactive contaminated volumes to be sent to a temporal storage or a final disposal facility, obtaining relevant information to carry out a radiation protection or environmental impact study, and/or evaluating intervention costs, etc. All over, characterisation of nuclear facilities may thus need to address some or all of the following objectives:

- Determination of the radionuclide vectors and chemical compositions for each expected type of radioactive wastes;
- Identification of different areas as regards their radiological/chemical characteristics and impacted environments;
- Estimation of the spatial extent of radioactive contamination in all the facility structures, systems and components, as well as the soils around the facility itself and possibly outside the nuclear site;
- Determination of the radiological and chemical background around the site;
- Validating results of the numerical simulations (e.g. activation, migration or diffusion);

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<sup>1</sup> The first nuclear facilities built in the 1950's were not designed to be readily decommissioned, and early tendency in preferring the deferred dismantling strategy caused loss of tacit knowledge and a missed opportunity to gain a consolidated know-how in this industrial sector.

<sup>2</sup> International Organization for Standardization.

- Identification and quantification of hard-to-measure radionuclides;
- Helping in modelling the radiation levels, in order to determine the remediation criteria for buildings and soils;
- Helping in selecting decontamination or remediation techniques;
- Estimation of occupational doses during clean-up and dismantling;
- Helping in defining the radiation protection actions to be implemented during the dismantling and clean-up activities;
- Facilitating the waste categorization in order to decide on its treatment/conditioning, packaging, shipment options and management route (clearance, recycling, reuse, storage, disposal);
- Evaluation of the dismantling and remediation costs;
- Helping in defining the dismantling and remediation optimal scenarios to be undertaken;
- Helping in designing any possible easements needed depending on the own characteristics of the nuclear facility or the whole site;
- Demonstrating that the remediation objectives of the clean-up of all or part of a site have been reached;
- Giving formal inputs to the documentation to be used for final approval/decisions.

Incomplete characterisation often leads to reliance on overly conservative assumptions and with considerable uncertainties, which at end are costly and time consuming.

In the remainder of the present document, Chapter 2 provides a comprehensive explanation of the characterisation steps that are in common use before starting the dismantling processes of any nuclear facility. Although characterisation is also needed during the dismantling activities to evaluate the efficacy of the applied decontamination procedures and to certify the final quality of the produced waste drums, these two aspects are beyond the scope of the INSIDER project and will not be discussed at all here.

Chapter 3 is fully devoted to the common sources of the radionuclides likely to be present in the facility structures and equipment to be dismantled, with a special emphasis on the methodology used to derive their associated vectors and scaling factors.

Chapter 4 deals with the evaluation of the questionnaires that have filled by identified partners, experts and end-users from different EU member states, plus Japan and Ukraine, with a consolidated experience in the domain.

Finally, Chapter 5 concludes the document with some recommendations regarding pre-decommissioning characterisation and cartography.

Although some basic concepts are assumed, the interested reader is referred to Adloff & Guillaumont (1991), Knoll (2010) Krane (1988), and ICRP (1991) for more details about nuclear physics, measurement of radioactivity, radiochemistry and radiation protection.

## 2 Characterisation steps

Figure 1 summarizes the different steps that must be followed up during the characterisation of a nuclear facility subject to a decommissioning programme. First of all, it is necessary to carry out a huge effort of analysing the whole historical documentation of the facility in order to establish its physical description together with an inventory of the associated radioactive source term. In most cases, the above physical description is complemented with visual inspection (using video imaging, laser scanning and/or photogrammetry), whereas the radioactive source term is usually validated by means of in-situ measurements (gamma spectrometry, alpha/beta contamination, dose cartography, etc.) and laboratory analysis of representative samples. The whole process can be improved, updated and consolidated with the help of 3D modelling and numerical simulations. For instance, by performing more visual inspections to refine the 3D models in one side and on the other by comparing the calculation results with the measured dose cartography. All of these steps are discussed below.

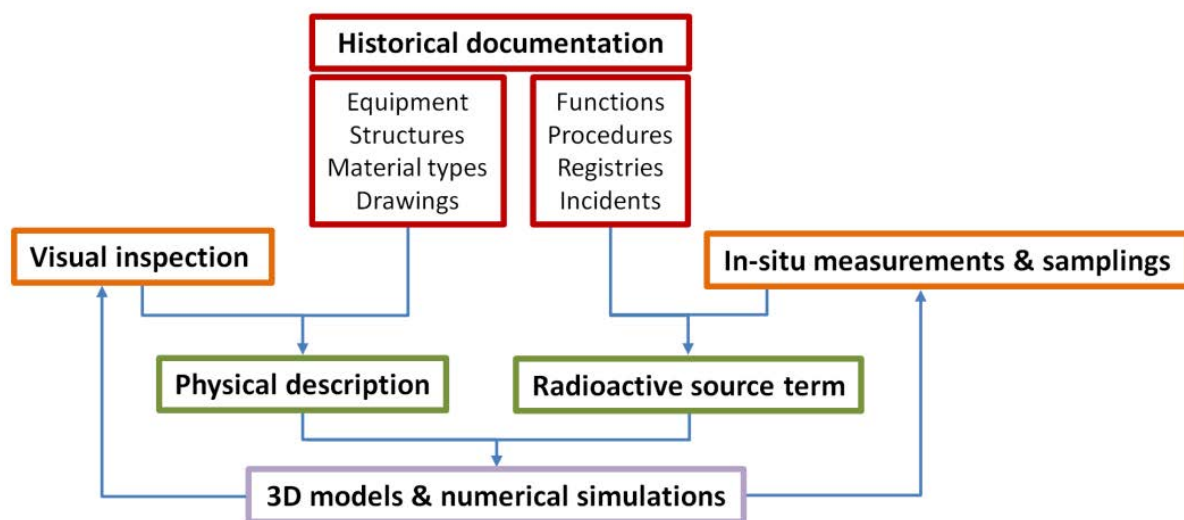


Figure 1: Different steps followed during the nuclear facility characterisation.

### 2.1.1 Study of historical documentation

The study of historical documentation should consider:

- the site context (geology, hydrogeology, occupations in the surroundings, etc.),
- the authorization licenses about the initial function of the facility and its possible reconfigurations or upgrades,
- the whole architectural plans and perspective drawings showing the geometry of its different equipment and their exact location,
- all the technical procedures implemented over the years giving rise to nuclear waste generation,

- the facility registries indicating the operation cycles, outage periods and other relevant aspects, such as nominal power or production rate,
- the safety reports explaining past radioactive discharges, unusual events or serious incidents and their consequences,
- any other information about the original background radioactivity in the zone as well as about eventual former characterisations carried out before the facility shutdown<sup>3</sup>.

This study must also account for key interviews and testimonies of former employees with a good knowledge of the facility history and non-written practices. These interviews may also provide further information identifying additional zones of interest.

Other documents to be consulted are original topographical maps and aerial pictures of the site to identify the location of first constructions, easements, storage areas, digging works or underground structures to help catching up for what has never been written or has been forgotten.

The in-depth processing of all the above information will enable an assessment to be made of past events, including possible contamination, and their consequences for the facility functioning. Such an investigation may also focus on natural catastrophes, which could have impacted the site permanently or temporarily concerning its basic operations and possible transfers of radioactive contaminants to the near environment.

Consultation of maintenance reports would give relevant inputs as well. For example, any change on surface coating or painting can have an impact on the local subsurface contamination.

### **2.1.2 Visual inspection**

Visual inspection, with or without optic aids, is the most simple, inexpensive and reliable technique for quality control process in industry. Its main purposes when applied for the pre-decommissioning characterisation of a nuclear facility are:

- to gather as much information as possible about the crucial characteristics (e.g., geometries, locations, material types, etc.) of its different equipment,
- to cross-check such an information with the existing database,
- to compensate for the lack of architectural plans and perspective drawings, and
- to determine the integrity of those materials (e.g., concrete walls and/or metal components) that may deteriorate with age and usage.

This last aspect is of key importance and should consider the following structural degradation mechanisms that might strongly limit the possible decommissioning scenarios:

- degradation of concrete by cracking, rebar corrosion and carbonation;

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<sup>3</sup> In fact, a progressive characterization over the years while the nuclear facility is still on the operational phase will always be much easier, more efficient and less expensive than delaying this activity until the end of the facility life cycle.



- degradation of metals due to corrosion, radiolysis and galvanization effects;
- degradation of other materials (e.g., graphite or beryllium) after neutron irradiation.

Optical aids vary from the remote video-cameras based on Si photodiodes or fiberscopes for the inspection of restricted access zones to more specialized devices such as laser-scanners to generate a point cloud in the three-dimensional (3D) space coordinates, often Cartesian, of the whole scene under study (Far, 2015). Application of high-speed photogrammetry techniques may also enhance automatic volume recognition (Kolyvas, 2015). Much of the success of visual inspection depends a lot on the indoor conditions (background radiation, temperature, humidity, illumination, etc.).

### **2.1.3 In-situ measurements and samplings**

On the basis of the preliminary information gathered regarding the zones of interest, lists of radioactive contaminants and means of measurement, it is possible to determine the adequate number of field measurements and samples, together with their locations. Extrapolating the obtained data to the whole area under study should be done with care, requiring a suitable inspection strategy to be beforehand planned based on advanced statistical approaches (CETAMA, 2017; Desnoyers & Dubot, 2014; EPA, 2000; MARSSIM, 2001). This particular topic will be the core of the workpackage WP3 in the present project.

#### **2.1.3.1 In-situ measurements**

In-situ measurements are mainly based on non-destructive assay methods to detect all types of ionizing radiation emitted by radionuclide such as helium nuclei ( $\alpha$ ), electrons ( $\beta^-$ ), positrons ( $\beta^+$ ), energetic photons (X- or  $\gamma$ -rays) and neutrons in the item under investigation, without affecting the physical or chemical form of this later.

The simplest, fastest and inexpensive method that can be used in most cases is the one based on measuring the radiation dose levels at predefined locations to map the associated spatial distribution or cartography of wide-areas (Mikami et al., 2015). The results of such measurements serve to localize the potential presence of radioactive singularities or hotspots and can be roughly correlated with the activities of the major gamma emitting radionuclides.

Although this method is widely applied, it is seriously affected by uncertainties on the measurement geometry, on the own characteristics of the measurement probe used and on the environment parameters to be considered. Cartography of alpha/beta contamination on surfaces would also be of great utility (Leskinen et al., 2013).

Other methods that can be applied for in-situ measurements are gamma spectrometry, passive neutron counting, digital autoradiography, and gamma, alpha or neutron imaging (Amgarou et al., 2016; Baschenko, 2004; Cieřlak, 2016; Knoll, 2010; O. Gal et al., 2001; Haudebourg & Fichet, 2016; Lamadie et al., 2005; Reilly et al., 1991; Woolf et al., 2015; Takeda et al., 2012). They will be broadly discussed in the deliverable D5.1 of the present project.

### 2.1.3.2 Laboratory analysis of representative samples

The main purpose in collecting samples for laboratory analysis is to obtain a small and informative portion of the population under investigation. Usually, representative samples that are expected to adequately reflect the properties of interest in the population being sampled are sought. However, targeted samples are sometimes needed in expected accumulation spaces.

Taking samples is easy in liquids but drilling techniques are needed in the case of hard materials (e.g., concrete). In turn, smears are the best choice to control any possible radioactive contamination on surfaces. Sample constitution and representativeness are important issues for the later data processing and for the confidence given to the results, namely the activity concentration of the sought radionuclides. Composite samples may be appropriate in tackling these issues.

The detection limits of measurement methods are central criteria in deciding on the number of samples and the quantity of materials to be collected for laboratory analyses. The choice of methods has to be optimised to fit the characterisation objectives. This decision should consider the relevance and importance of the contaminant in the safety or environmental assessment, technical performances, the cost of the measurement, etc.

In general, the taken samples are analysed by means of destructive assay (DA) methods in dedicated radiochemical laboratories. These methods can be applied directly to the dissolved sample or after the separation process, depending on the complexity of the mixture and the resolution of the measurement instruments used (Adloff & Guillaumont, 1991). In the case of solids, digestion process (with microwave or oxygen bomb) are often needed, whereas pyrolysis are systematically used for volatile radionuclides.

The selection of the radionuclides to be analysed and of associated chemical compounds depends on the preliminary historical and functional analyses. Chemical separations must be designed to isolate the element to be determined from the chemical and radioactive interferences. Basic methodologies are liquid-liquid extraction, precipitation, distillation, catalytic combustion, extraction chromatography, ion exchange chromatography, and mass spectrometry.

Counting systems for isolated alpha emitting radionuclides are solid scintillation counters, such as ZnS(Ag), or proportional counters. In the case of a mixture of alpha emitting radionuclides, liquid scintillation counters (LSC) can be used, which give alpha spectra with low resolution. More accurate measurement can be achieved with high-resolution alpha spectrometers. Activity concentration of pure beta emitting radionuclides can be determined by proportional counters but the most effective method is LSC with alpha-beta discrimination. That of X-ray and low-energy gamma emitting radionuclides can be directly measured using thin planar Ge detectors. Low amount of these emitters in the samples means that chemical separation has to be carried out prior to measurements to avoid unwanted interferences.

In the case of high-energy gamma emitting radionuclides, such as  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ , direct measurements in an appropriate geometry by solid scintillation gamma spectrometry (low resolution) can be performed. Semiconductor detectors with high resolution may be applied when a complex mixture of gamma emitters is treated before separation or if the sample has a simple composition of

gamma emitters. Most of the above techniques will be widely discussed in the workpackage WP4 of the present project.

Further chemical measuring kits are available (laser induced breakdown spectroscopy (LIBS), photoionization detection (PID), heavy metals testing, X-fluorescence, etc.) for situations where chemical pollutants also need to be characterised. The previous technique can be used on line. For laboratory measurement after digestion process technique such as ICP are widely used.

For responsiveness and effectiveness purposes, while reducing logistic and cost aspects, CEA has recently developed a mobile radiochemical laboratory for the characterisation of contaminated soils, called SMaRT<sup>4</sup> (Goudeau et al., 2017).

#### **2.1.4 3D models and numerical simulations**

The term 3D modelling refers the process of developing a mathematical representation in the form triangle or other polygon mesh (i.e., a collection of vertices, edges and faces) of any polyhedral object in the space coordinates, via computer-aided design (CAD) systems like CATIA<sup>5</sup>, Autodesk<sup>6</sup>, Solidworks<sup>7</sup>, etc. The formed 3D models are not technically graphics until they are displayed through rendering, which consists in converting them into images with a very high degree of visual realism by using ray tracing or high dynamic range (HDR) techniques to simulate several optical effects, such as lighting, shallow depth-of-field and diffuse reflection (Chopine, 2011).

In nuclear decommissioning, a precise digital mock-up of the whole scene under study is then designed based on the information gathered from the historical documentation and the results of visual inspection. This mock-up can be of valuable help for the scenario optimization or for training purposes in an immersive room (Szöke, 2015). In practice, simplifications are made to reduce the size of the 3D model by removing, for instance, fastenings (screws, nuts, washers, etc.), deleting small holes, smoothing extrusion profiles, and eluding non-visible or hidden objects. As an illustrative example, Figure 2 shows a comparison between a real picture and its associated 3D model of the APM<sup>8</sup> cell 414 (Chabal & Soulabaille, 2016) at the CEA Marcoule site, which currently represents one of the largest nuclear dismantling worksites in the world.

After adding the needed information concerning the material compositions and densities, the updated 3D models can also be used for numerical simulations, using Monte-Carlo and/or deterministic methods, of the relevant physical phenomena associated with the production and transport of nuclear particles as well as their interactions with matter. The leading calculation codes for this purpose are: MCNPX (Pelowitz, 2011), FLUKA (Ferrari et al., 2005), GEANT (Agostinelli et al., 2003), DARWIN (Tsilanizara et al., 2000), NARMER (Visionneau et al., 2017), ORIGIN (Bell, 1973), TRIPOLI (Brun et al., 2015), SCALE (Bowman, 2011), APOLLO (Sanchez et al., 1988), FISPIN (Burstall, 1979), etc.

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<sup>4</sup> Shelter for Monitoring and nucleAR chemistry.

<sup>5</sup> Further information available here: <https://academy.3ds.com/en/software/catia-v5-student-edition>

<sup>6</sup> Further information available here: <https://www.autodesk.fr/products/autocad/overview>

<sup>7</sup> Further information available here: <http://www.solidworks.com/>

<sup>8</sup> APM is a French acronym for *Atelier Pilote de Marcoule* and this facility was built in 1960's as an industrial demonstrator of the different spent fuel reprocessing techniques developed in CEA research laboratories.

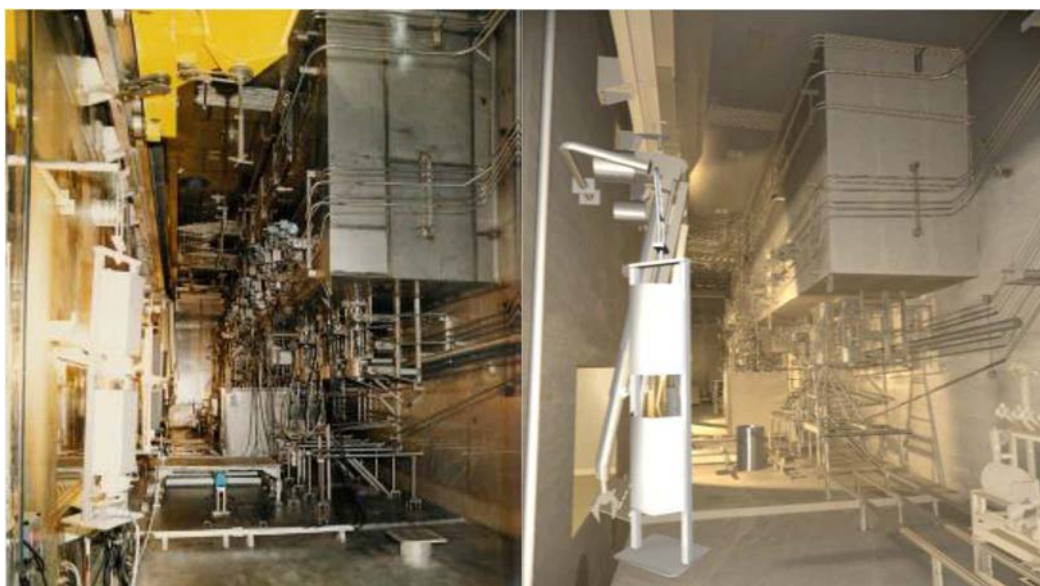


Figure 2: Comparison between real picture (left) and 3D model (right) of the APM cell 414.

### 3 Radionuclide inventory

The good knowledge of the processes giving rise to radioactive waste generation is one of the most effective ways to limit the range of the radionuclides likely to be present in the facility structures and equipment to be dismantled as well as the in-situ measurements and the radiochemical analytical methods to be used.

In a fuel manufacturing plant, for example, uranium and its attendant products may be the only radionuclide species possible, so that their specific quantification is all what must be required avoiding thus unnecessary broad characterisations to be carried out.

#### 3.1 Origin of radioactivity in nuclear facilities

##### 3.1.1 Background radioactivity

Background radioactivity may be different not only from one site to another but also within the same site and the associated radiation dose rate varies with time. Radionuclides present in the environment (Shahbazi-Gahrouei et al., 2013) can be classified into:

- The primordial or natural radionuclides that may be typically found in the earth's crust. Such radionuclides consist of the principal ones in the decay series of  $^{232}\text{Th}$  and  $^{238}\text{U}$  (see Figure 4 and Figure 3)<sup>9</sup>, which half-lives are comparable to the estimated age of the universe, together with  $^{40}\text{K}$ .
- The cosmogenic radionuclides such as  $^{14}\text{C}$  and  $^3\text{H}$  that are produced in the upper atmospheres as a result of the nuclear interactions,  $^{14}\text{N}(\text{n,p})^{14}\text{C}$  and  $^{14}\text{N}(\text{n},^3\text{H})^{12}\text{C}$ , of cosmic

<sup>9</sup>  $^{234}\text{U}$  and  $^{235}\text{U}$  may also be present in natural uranium but with very small isotopic abundances (0.0057% and 0.72%, respectively).

neutrons with nitrogen nuclei. Their corresponding equilibrium activity is controlled by their production rate and their residence times in the surrounding air, oceans and soils.

- The anthropogenic or manmade radionuclides that have been released in the nature because of the different worldwide nuclear activities carried out in the past, namely the ones associated with:
  - the nuclear weapons' tests;
  - the nuclear power plants;
  - uranium mining and milling;
  - nuclear spent fuel reprocessing;
  - nuclear disasters like at Chernobyl (1986) and Fukushima (2011).

Several anthropogenic radionuclides, including  $^{90}\text{Sr}/^{90}\text{Y}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{137}\text{Cs}/^{137}\text{Ba}^m$ ,  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ , as well as the uranium and plutonium isotopes, have attracted the attention of health specialists. Together with the natural radionuclides, they can be concentrated in different parts of the soil column. This makes very difficult to determine the variation range of the background radioactivity in a built-up area and the sole alternative would be to measure that of the nearby soils, known as uncontaminated.

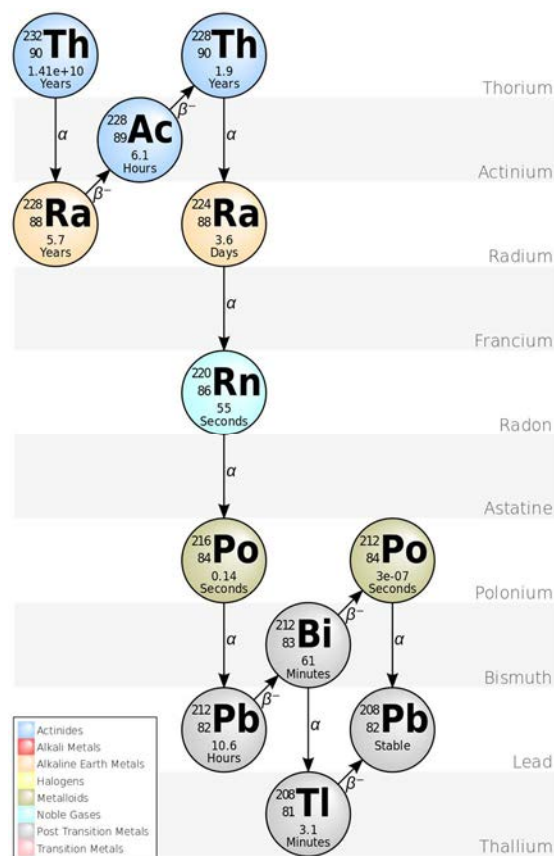


Figure 3: Decay chain of  $^{232}\text{Th}$  (source: Wikipedia).



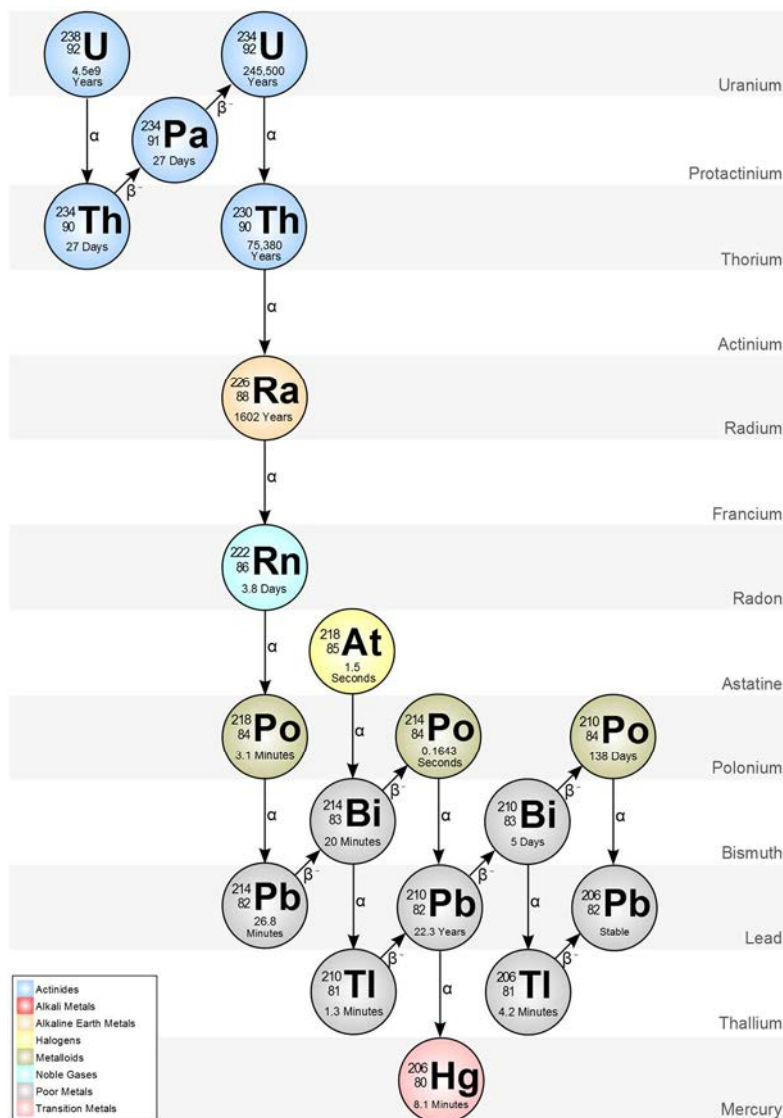


Figure 4: Decay chain of  $^{238}\text{U}$  (source: Wikipedia).

### 3.1.2 Nuclear activation

Nuclear activation is the process whereby an ionizing radiation causes an unintended induction of radioactivity in stable substances. The physical processes behind the interaction of nuclear particles with matter are very complex and have been largely described in the literature (see Leroy & Rancoita, 2016).

During the normal operation of nuclear reactors, thermal<sup>10</sup> or fast neutrons are used to trigger the necessary fission reactions for the energy production. These neutrons are hence able to activate several materials within the same reactor core and in the peripheral structures such as graphite moderator and bulk biological shield made of concrete (whether or not containing barite aggregates) and steel reinforcements.

<sup>10</sup> Neutrons that are in thermal equilibrium with a surrounding medium and their most probable kinetic energy at a temperature of 290 K is 0.025 eV, which is equivalent to a speed of 2200 m/s.

High-energy accelerators generate showers of secondary particles through cascades of successive electromagnetic and hadronic interactions, which ultimate result is a predominant emission of fast neutrons outside the beamline. These fast neutrons can travel large distances until being thermalized and possibly undergo radiative capture or  $(n,\gamma)$  reactions. As these accelerators are commonly housed in buildings with thick concretes walls, these latter become radioactive over time, just like all the machine components, heavy shielding materials (lead or tungsten) and any other infrastructure in the irradiation rooms.

Nuclear activation may also take place in the ITER reactor (Rosanvallo et al., 2007) because of the fusion reaction between deuterium and tritium emitting 14 MeV neutrons. For that reason, efforts have been during the conception of this facility to reduce the potential waste amounts and category, in particular thanks to a proper choice of the materials to be used for its construction. It is expected that around 100 years of radioactive decay after the reactor shutdown, no long-lived radionuclides should remain in the generated wastes and, even without considering recycling, no deep repository would be necessary.

In view of the above, we can conclude that, regardless of the type of nuclear facilities, neutrons always represent, by far, the major cause of nuclear activation. An initial estimation of the extent and levels of neutron activation can be carried out on the basis of numerical simulations using one or more of the calculation codes cited in Section 2.1.4 (ISO, 2013). The calculation process is threefold. Firstly, the spatial and energy distributions of the neutron flux throughout the system are derived. Then, individual reaction rates are estimated for the parent elements that give rise to radioactive daughters after neutron activation. Lastly, the activity concentration (in units of  $\text{Bq g}^{-1}$ ) is computed for each generated radionuclide.

It is thus crucial to know the exact geometry of all the facility structures and equipment as well as their material compositions, including impurities and trace elements (like cobalt in stainless steels) that are susceptible to produce this kind of radioactivity. In addition, it is mandatory to have an access on the facility registries indicating the operation cycles and other relevant aspects, such as nominal power or production rate, to derive the total integrated neutron flux during the whole period that it has been in use.

The possible radionuclides of concern in this domain are:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{22}\text{Na}$ ,  $^{36}\text{Cl}$ ,  $^{39}\text{Ar}$ ,  $^{41}\text{Ca}$ ,  $^{46}\text{Sc}$ ,  $^{51}\text{Cr}$ ,  $^{54}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{57}\text{Co}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{93}\text{Mo}$ ,  $^{93}\text{Zr}$ ,  $^{94}\text{Nb}$ ,  $^{108}\text{Ag}^{\text{m}}$ ,  $^{110}\text{Ag}^{\text{m}}$ ,  $^{125}\text{Sb}$ ,  $^{133}\text{Ba}$ ,  $^{134}\text{Cs}$ ,  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$ ,  $^{155}\text{Eu}$ ,  $^{166}\text{Ho}^{\text{m}}$ . As  $^{60}\text{Co}$  use to be the predominant radionuclide, almost all of its initial radioactivity (i.e.,  $\sim 99.9\%$ ) decays after an elapsed time 50 years and may leave alone the long-lived ones, even they were initially present at small amounts.

### 3.1.3 Radioactive contamination

Radioactive contamination refers to the unintended and unwanted presence of a radioactive substance on structural surfaces and/or within equipment, or the process giving rise to its presence in such locations. It is more likely for an activated component to be also contaminated rather than the opposite case.

In the case of nuclear reactors, it results from the unpredictable release from the fuel, generally made up of enriched uranium and MOX<sup>11</sup>, of fissile elements, fission products, together with their progeny, during normal operation or unwanted incidents (e.g. rupture of fuel rod cladding tubes). It also includes corrosion and erosion within the refrigeration circuits of activated materials that tend to accumulate near the region where elbows and adjacent wedges are welded into the piping loops.

Such a phenomenon is generally known as “crud” in water-reactors and “deposit” in gas-cooled ones. It may be strongly adherent (i.e., “fixed”) or easily removed by mechanical means (i.e., “loose”). It cannot be excluded the radioactive contamination caused when handling, treating and storing (e.g., spent fuel pool) any radioactive substance during the facility normal operation. Moreover, the formation of airborne or volatile radionuclides may give rise to a spread of radioactive contamination in unsuspected zones (walls, ceilings and ventilation systems), until reaching and filling any available structural cracks or crevices.

A particular attention must be paid to nuclear fuel-cycle plants that can use a variety of fabrication processes and very complex equipment. For example, enrichment of <sup>235</sup>U can be carried out by centrifugation or by gaseous diffusion, and fuel fabrication may involve different methods or different fissile materials. The gaseous diffusion process uses uranium hexafluoride UF<sub>6</sub>, which hydrolyses in contact with water vapour, even in trace amounts, to form aerosols or insoluble fine particles that may deposit on any surface encountered by the fluid, resulting in radioactive surface contamination. All the tubes and vessels in contact with the process fluid are contaminated to some extent.

Radionuclides of concern in this domain are: <sup>79</sup>Se, <sup>90</sup>Sr/<sup>90</sup>Y, <sup>99</sup>Tc, <sup>106</sup>Ru/<sup>106</sup>Rh, <sup>129</sup>I, <sup>137</sup>Cs/<sup>137</sup>Ba<sup>m</sup>, <sup>144</sup>Ce/<sup>144</sup>Pr, <sup>231</sup>Pa, <sup>232</sup>Pa, <sup>228</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th, <sup>234</sup>Th, <sup>232</sup>U, <sup>233</sup>U, <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>238</sup>U, <sup>237</sup>Np, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>242</sup>Pu, <sup>241</sup>Am, <sup>242</sup>Am, <sup>243</sup>Am, <sup>242</sup>Cm, <sup>243</sup>Cm, <sup>244</sup>Cm.

In contrast to nuclear activation, it is very difficult to theoretical predict the extent of radioactive contamination. The most appreciate attempt is that carried out by CEA (Pérot et al., 2004) to simulate transport and deposition of corrosion and/or erosion activated products throughout the PWR<sup>12</sup> primary circuit. Such kind of calculations can only provide orders of magnitude estimation on an averaged basis, often neglecting the formation of radioactive hotspots.

### 3.2 Radionuclide vectors and scaling factors

As revealed above, the inventory of the radionuclides to be declared is habitually numerous and varied. Radionuclide vectors with known activity ratios are then used as a radioactive “fingerprints” of the different contaminated materials (ISO, 2007).

Some radionuclides are easy-to-measure (ETM), namely the ones emitting high-energy  $\gamma$ -rays such as <sup>137</sup>Cs or <sup>60</sup>Co, but most of them are hard-to-measure (HTM)<sup>13</sup> and need complex radiochemical analytical methods (e.g. pure alpha or beta emitters). The scaling factor methodology allow estimating the radioactivity of HTM radionuclides using correlations between them and the chosen key radionuclide, also called radio-tracer, among the ETM radionuclides (IAEA, 2009). Specifically,

<sup>11</sup> MOX is an acronym for Mixed OXides and consists of plutonium blended with natural or depleted uranium.

<sup>12</sup> PWR is an acronym for Pressurized Water Reactor.

<sup>13</sup> Alpha, pure beta and X ray and low energy gamma emitters.



each HTM radionuclide can be predicted via the radio-tracer by simply multiplying the concentration of this latter with their associated scaling factors, previously obtained by means of appropriate radiochemical analytical methods and which represent the average correlation between their activities in the item to be assayed.

When NDA measurements provide the activity of radio-tracer as well as that of other ETM radionuclides, these data are kept for periodic control of the validity of the scaling factors used for the inventory.

To establish representative correlations, radio-tracers are selected according to the following criteria:

1. They must be present in the assay mixture in significant quantities;
2. Their radioactive half-lives must be long compared with the time necessary for sampling, measurement, and physicochemical processing;
3. They can be non-destructively measured beyond the detection limit.

Conversely, a technical basis is required for the existence of these representative correlations, as for instance HTM and ETM radionuclides with identical chemical behaviour (like solubility) or the ones formed by a similar mechanism. Any change regarding the facility process may produce different mixture of radionuclides, so the corresponding scaling factors need to be reassessed again.

If there is not a technical basis for the correlation, it may not be valid or defensible in practice. Especially, in cases where correlations are only observed on log-log plots as it is difficult to justify the corresponding spread data points or variance that may be up to several orders of magnitude.

Last but not least, tritium ( $^3\text{H}$ ) has a particular problem due its extremely high mobility and must be considered separately. It is often encountered on its own (i.e., not accompanied by the presence of a scalable radio-tracer) and, as it is a low energy  $\beta$ -emitter ( $< 18.6$  keV), it is very difficult to detect.

### **3.3 Presence of other hazardous substances**

The eventual existence of other hazardous substances also needs to be controlled during pre-decommissioning characterisation of nuclear facilities (IAEA, 2006). Above all, there is the particular case of asbestos that was the preferred electrical and thermal insulator in the past. Further possible chemical toxicants that must be controlled are: antimony, arsenic, beryllium, boron, cadmium, chromium, cyanides, mercury, nickel carbonyls, lead, polychlorinated biphenyls, selenium and sodium or sodium-potassium alloy. Materials exhibiting both radiological and toxic properties, commonly known as “mixed wastes”, may suppose a non-trivial problematic regarding dismantling, matrix encapsulation and disposal routes.

## **4 Evaluation of the questionnaires**

In parallel with Task 2.1, a complete questionnaire (see the annex) has been prepared in order to draw a clear picture of the current practices in an international context regarding the characterisation of nuclear facilities that are currently undergoing a decommissioning programme. Then, several partners, experts and end-users from different EU member states, plus Japan and Ukraine, with a

consolidated experience in the domain, have been asked to fill this questionnaire according to the best of their knowledge.

Albeit the short deadline (less than two months including the summer holidays period), we have finally registered nine participations representing up to seven countries: Belgium, France, Germany, Italy, Japan, Spain and Ukraine.

However, as almost all of the participants asked to treat confidentially their contribution, a decision was made to henceforth publish here only a general overview specified for each country, while trying to be extremely careful to not modify the meaning of the whole answers received.

It should nevertheless be noticed that question (2.1) of the questionnaire did not allowed to gather additional value-added data compared to what already exists in the IAEA's Power Reactor Information System (PRIS)<sup>14</sup>. Consequently, this question was thereafter withdrawn and instead a summary of NPPs<sup>15</sup> currently undergoing a decommissioning programme in each country is given.

#### **4.1 Belgium**

The only reactor shutdown in Belgium is the small size BR3 reactor with a nominal power unit of 10 MW<sub>e</sub><sup>16</sup>. BR3, which is located at the Nuclear Research Centre in Mol, was the first PWR reactor to be built in Europe. It was used for 25 years, first as a teaching tool for the future commercial reactors at Tihange and Doe, and then for the Vulcain Project (spectral shift using heavy water moderation). It was afterwards extensively used for testing high burn-up and gadolinium type fuels and, finally, for extensive testing of MOX fuels. It operated between 1962 and 1987. It was selected by the European Commission (EC) in 1989 as one of the four pilot dismantling projects.

Other nuclear facilities currently undergoing a decommissioning programme are: the Eurochemic for fuel reprocessing and Belgonucleaire for MOX production. Moreover, licences for decommissioning of two high-energy accelerators (INW cyclotron at Ghent and Best Medical at Fleurus) are in progress.

The physical nature, composition and estimated volumes of radioactive wastes generated in the above decommissioning programmes are detailed by ONDRAF/NIRAS, which is the Belgian National Agency for Radioactive Waste and enriched Fissile Material, in its third report (ONDRAF, 2013).

The main objectives to be achieved during the radiological and/or chemical characterisation of the considered nuclear facility and its nearby soils are:

- to confirm the historical information,
- to identify radionuclide vectors and associated scaling factors,
- to localised radioactive contamination zones,

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<sup>14</sup> Available at : <https://www.iaea.org/pris/>

<sup>15</sup> Nuclear Power Plants.

<sup>16</sup> Megawatt electric.

- to well address safety issues, including ALARA<sup>17</sup> planning,
- to select decontamination techniques,
- to define the best waste management strategy,
- to perform an environmental impact assessment,
- to optimise cost estimations, and
- to check that the site at the end of decommissioning complies with the national general regulation, known as ARBIS, of the protection of the population, the workers and the environment against the dangers of ionising radiation.

The sought radionuclides are the ones listed in Sections 3.1.2 and 3.1.3 but a preliminary inventory is often established based on the historical information and numerical simulations.

Several of the radiochemical analytical methods expounded in Section 2.1.3.2 are applied to analyse irradiated and non-irradiated samples, including the chemical composition of potential impurities, depending on the practical restrictions. Whereas the most in-situ measurement techniques commonly used are: dose rate cartography, surface contamination (direct and indirect), total gamma counting, gamma spectrometry and gamma imaging.

There is not a specific protection level concerning the measured radionuclide concentrations and their associated uncertainties during the pre-decommissioning characterisation of a nuclear facility. The only acceptance criteria, imposed by ARBIS, are the ones linked to the produced waste drums during dismantling and decontamination. That is the sum of the activity concentration and associated uncertainty (with at least a coverage factor  $k = 2$  or a 95% confidence interval) for each of the declared radionuclides per drum mass or volume must be below an established upper limit).

The experimental procedures that are normally undertaken to qualify each one of the different radiochemical analytical methods used are the ones described in Prichard & Barwick (2007), whereas in-situ measurement techniques are validated in accordance with ISO/IEC 17025 (ISO, 2005) and/or ISO 9001 (ISO, 2015).

In the opinion of the questionnaire participant, the specific objectives of a benchmarking exercise for both the laboratory analysis of samples and *in-situ* measurements should be linked to the overall objective and global uncertainty of the full characterisation process.

The way to derive the radionuclide fingerprints and their associated scaling factors is that described in Section 3.2.

The participant response with respect to no presence of hard-to-measure radionuclides (i.e., after a cooling period  $\gg 10$  half-lives) is that the characterisation is relatively easy and straightforward.

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<sup>17</sup> As Low As Reasonably Achievable. Justification of radiation exposures, optimisation of radiation protection and application of individual dose limits are the three ALARA principles. The International Commission on Radiological Protection, in its Publication 103 (ICRP, 2007), states that: "*the likelihood of incurring exposures, the number of people exposed, as well as the magnitude of their individual doses should be kept as low as reasonably achievable taking into account economic and societal factors*".

However, if there are only HTM radionuclides present he assumed that characterisation could only rely on samplings and destructive analyses.

The main difficulty in defining a radionuclide vector for a given facility is that it is a long, costly and multi-parameter process, which can suppose large uncertainties.

Considering experimental uncertainties in combination with waste acceptance criteria, a global radionuclide vector for the entire facility is preferred, whenever possible, instead of specific ones for individual subsystems.

In what respects chemo-toxicity, its assessment is usually linked to certain types of materials or substances so that it can be correlate with radio-toxicity but, according to the participant, it is sometimes difficult to find laboratories that can handle both problems.

Extensive theoretical calculations of neutron activation in the case of BR3 reactor have been carried out using the TRIPOLI code (Brun et al., 2015).

However, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for:

- subsequent clearance process,
- waste classification and waste treatment process,
- planning decontamination actions,
- planning site remediation actions,
- performing the final site release process,
- estimating radiation doses to workers during the decommissioning activities,
- estimating the environmental impact during the decommissioning activities,
- estimating the amount of wastes produced during the decommissioning activities, and
- estimating of decommissioning costs.

Finally, the participant confirmed that operational characterisation is carried out during the dismantling process, but without providing any further precision about how it is used for estimating radiation doses to workers during the decommissioning activities.

## 4.2 France

France is the nuclear country by excellence in Europe<sup>18</sup> with 58 operational reactors (900 - 1600 MW<sub>e</sub>) representing more than 70% of the national electricity production. All of the French nuclear power plants are operated by the EDF (*Electricité de France*) company.

During the 1960s, in line with the overall target of industrial independence and domestic technological development, basic prototype designs were promoted, such as the Chooz-A 305 MW<sub>e</sub>.

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<sup>18</sup> France is not so far from the United States of America, which is first position with up to 99 operational reactors (500 - 1500 MW<sub>e</sub>) representing almost 80% of the national electricity production.

PWR, which was built jointly with Belgium, and the Brennilis experimental 70 MW<sub>e</sub> heavy water reactor. However, international developments in the nuclear industry in that period led to the recognition that the French reactor designs could not compete with light water reactors. A decision was made in 1969 to build LWRs under license, whilst restructuring the domestic industry to improve competitiveness. Subsequently, the French government envisaged an ambitious construction programme of one or two PWRs a year.

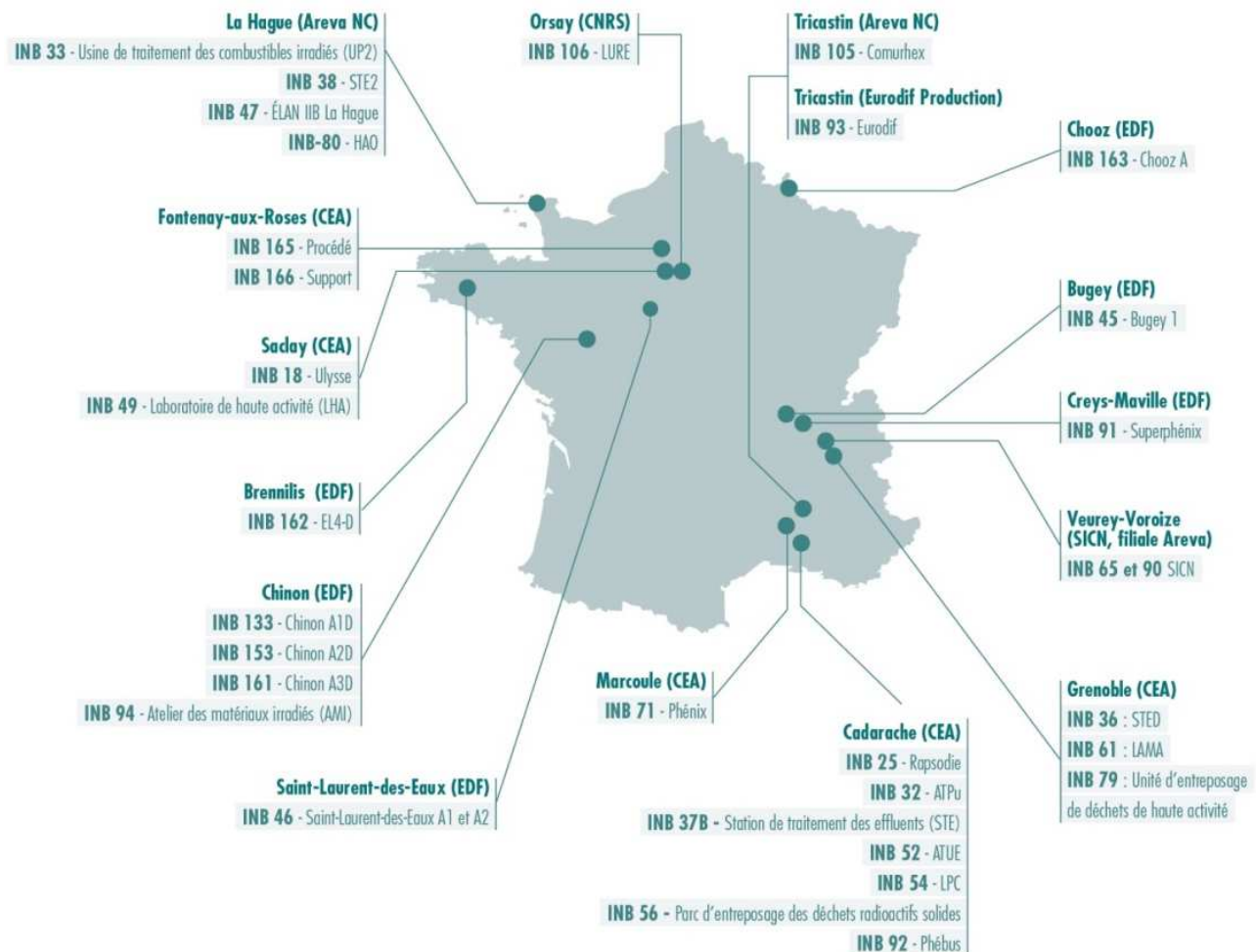
From 1974 to 1981, emphasis was put on adaptation of the Westinghouse design for the development of a French standard. The nuclear programme accelerated the pace with the 1970s oil crisis. The nominal power capacity of French reactors increased from 900 MW<sub>e</sub> to 1 300 MW<sub>e</sub> and later to 1 500 MW<sub>e</sub>. France developed and implemented in parallel a strong domestic fuel cycle industry, built upon the infrastructure originally established by CEA. In 1981, Framatome terminated its license with Westinghouse and negotiated a new agreement, gaining more autonomy. This allowed to develop a wide range of servicing expertise as also capabilities in reactor operation and maintenance services. In the same year, France had to adapt its energy policy to a lower than expected economic growth, alongside with the occurrence of over-capacity in the national electricity supply system. The achievement of the 1 450 MW<sub>e</sub> N4 model was the landmark for a totally autonomous French reactor design.

Then, a new period started with the formation of the AREVA Group, the construction of a 1 600 MW<sub>e</sub> European Pressurized Reactor started in Olkiluoto (Finland) and another one in Flamanville (France). At the same time, the implementation in Marcoule of the first GEN IV 600 MW<sub>e</sub> research reactor, called ASTRID, was envisaged and it is currently at the design phase.

Thirteen experimental and nuclear power reactors are being decommissioned in France (see Figure 5), nine of them are the first-generation gas-cooled or graphite-moderated types, while the other four include the 1 240 MW<sub>e</sub> Superphénix (Creys-Malville) reactor, the veteran 233 MW<sub>e</sub> Phénix reactor, as also with the above Chooz-A and Brennilis prototypes. EDF points to Chooz A as the most representative NPP of those currently operating, and dismantling work on it is on schedule for completion in 2022 and on budget. Other star nuclear facilities subject to decommissioning programme are UP1 and UP2 400 spent fuel reprocessing plants at the Marcoule and La Hague sites, respectively, together with the Eurodif gaseous diffusion enrichment plant at the Tricastin site.

CEA has a unique position in the nuclear decommissioning domain because of the number of facilities involved (nearly two thirds of the ones currently being dismantled in France), the wide variety of these latter (experimental reactors, fuel cycle processing plants, effluent treatment stations and legacy waste storage sites), as well as their huge and complex equipment (reactor vessels, hot cells, heat exchanger, dissolvers, centrifuges, evaporators, pulsed columns or filters, mixer-settler banks, metering wheels, tanks, pipeline circuits, etc.). The CEA's decommissioning programmes do not therefore benefit from any kind of a "serial effect". This difficulty is further increased by the age of the facilities to be dismantled, some of which were commissioned in the sixties and where the traceability of their activities has not always been preserved. Beyond these complications, all the nuclear facilities at the Grenoble site have been fully dismantled. The three reactors (Siloé, Siloette and Mélusine) have already been decommissioned. That of the active materials analysis laboratory

(LAMA) and two radioactive effluent and waste treatment stations is currently being examined by the French Nuclear Safety Authority (ASN).



**Figure 5: Overview of the nuclear facilities under decommissioning in France (source: ASN<sup>19</sup>).**

According to the questionnaire participants, the current dismantling of the first generation reactors will produce 800 000 metric tons of conventional waste (free of radioactivity) that can be reused and 170 000 tons of radioactive waste for final storage. The classification of these later will be 11% of Very Low Level Waste, 4% of Low Level Short Lived Waste, 2% of contaminated graphite, 0.5% of radioactive sodium and 0.06% of Intermediate Level Long Lived Waste. The physical nature, composition and estimated volumes of all the radioactive wastes to be generated in France are detailed in the report published by the National Radioactive Waste Management Agency (ANDRA) on 2015 (ANDRA, 2015).

<sup>19</sup> The French Nuclear Safety Authority.



In their opinion, the specific difficulties and obstacles during the above decommissioning programmes are basically:

- Limit financial resources;
- Extension of the scope of nuclear facilities undergoing dismantling;
- Lack of input data on the initial state of the earliest nuclear facilities;
- Variation in waste disposal costs, storage specifications and commissioning deadlines;
- Disposal availability for intermediate-level wastes and alpha-contaminated graphite.
- The increasing number of safety rules, and regulation requests.

The main objectives to be achieved during the radiological and/or chemical characterisation of the considered nuclear facility and its nearby soils are:

- To provide qualitative information: extend and kind of contamination? Which radionuclides are present? Presence of toxic chemicals?
- To provide quantitative information: chemical composition, content of toxic chemicals, radioactive contamination levels, radionuclide activities, dose cartography, and 3D radiological models (gamma/alpha imaging for example);
- To anticipate the radioactive waste management (e.g., waste classification and purchase of storage casks).

ANDRA has defined a list of 143 radionuclides that may have an impact on both the population and environment, including the short-lived ones that are in secular equilibrium with their parents (ANDRA, 2013). For practical purposes, the following radionuclides are commonly measured during the characterisation of nuclear facilities:

- Gamma emitters:  $^{137}\text{Cs}$ ,  $^{46}\text{Sc}$ ,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{110}\text{Ag}^m$ ,  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$
- Beta emitters:  $^{55}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}/^{90}\text{Y}$ ,  $^{36}\text{Cl}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$
- Alpha emitters:  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Cm}$ ,  $^{244}\text{Cm}$
- Other potential demand (less frequently):  $^{93}\text{Zr}$ ,  $^{107}\text{Pd}$ ,  $^{121}\text{Sn}^m$ ,  $^{126}\text{Sn}$ ,  $^{10}\text{Be}$ ,  $^{41}\text{Ca}$ ,  $^{241}\text{Pu}$ ,  $^{93}\text{Mo}$ ,  $^{129}\text{I}$ ,  $^{99}\text{Tc}$ ,  $^{94}\text{Nb}$

In line with what has already been pointed out in Section 3.1.2, the participants specified that impurities are systematically measured in order to assess the extent and levels of the neutron activation effects by means of numerical simulations. They assured that such measurements are always preferred as they are easier and less costly on non-irradiated samples. It should be noted that the chemical analysis of irradiated items might present other interests, as for instance, to correlate the statistical variability of impurities with that of the generated radionuclides.

Regarding the protection levels to be targeted in terms of activity concentration and associated uncertainty for each of the declared radionuclide, most of the acceptance criteria are given in the

different ANDRA technical specifications<sup>20</sup> as well as the several guides established by ASN<sup>21</sup>. As a best practice, the radiological inventory must be reasonably penalizing.

Most of the radiochemical analytical methods expounded in Section 2.1.3.2 are applied to analyse representative samples. Uncertainties on the corresponding results depend on the laboratory, the protocols followed and the instruments used. The choice of the laboratory is related to the radioactivity levels, analytical capacities and cost analysis. In what respect numerical simulations (including the step for validating the model), the combined uncertainty is determined as the square root of the quadratic sum of the individual uncertainties arising from different sources. For the most part, there is a need of a mineralization process and, from time to time, complicated chemical separation processes to isolate the chemical element of interest.

The participants remarked that each of the different laboratory analytical techniques used is generally set-up according to the state of the art and the chemical separation procedures are properly standardized. Validation procedures are particularly important for the detection limit determination.

In-situ measurements are usually carried out through dose rate cartography referenced to a predominant gamma emitting radionuclide, typically <sup>60</sup>Co. When there are several gamma emitters (<sup>60</sup>Co, <sup>108</sup>Ag<sup>m</sup>, <sup>110</sup>Ag<sup>m</sup>, <sup>137</sup>Cs), complementary measurements with a gamma spectrometer are performed. In both cases, the chosen detector must be conveniently collimated. Other used techniques are alpha/beta surface contamination counting, beta spectrometry, digital autoradiography and radiation imaging by means of gamma and/or alpha cameras.

The qualification procedures for such measurement systems are based on:

- Calibration with point/extended standard radioactive sources;
- Theoretical simulations of the instrument response (MCNP);
- Experience tests on reference drums with well-known chemical compositions.

Another aspect highlighted by the participants is that the implementation principle of any quality assurance plan is to assess the accuracy and the detection limit of a technique. They indicated that QA/QC/QM<sup>22</sup> guidelines mostly come from environmental measurement and there are a lot of national regulations in this matter.

The specific objectives of a benchmarking exercise for the laboratory analysis could be:

- Improvement of sampling processes;
- Comparison of results (accuracy evaluation);
- Uncertainty evaluation (digestion process + measurement).

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<sup>20</sup> Available at: [http://www.andra.fr/index.php?id=itemmenu\\_article\\_484\\_1681\\_8\\_1&itemracine=462](http://www.andra.fr/index.php?id=itemmenu_article_484_1681_8_1&itemracine=462)

<sup>21</sup> Available at: <http://www.french-nuclear-safety.fr/References/ASN-Guides-non-binding>

<sup>22</sup> Quality Assurance/Quality Control/Quality Management.



The participants expressed that it is preferable to perform inter-comparison exercises with real samples representative of radioactive contaminated concrete and/or soil. Although it is difficult to know *a priori* the "true values" of such samples, this kind of exercises will consolidate, harmonize and identify the actual limits the different methodologies applied.

Concerning in-situ measurements, the specific objectives of a benchmarking exercise are:

- Mastering of the operational procedures;
- Mastering of data analysis methods (namely in regards to gamma spectrometry);
- Mastering of the numerical simulation codes used to well interpret the reading of the instruments used to derive the sought radioactive source term;
- Mastering of statistical and geo-statistical approaches;
- Mastering of the methodology followed to evaluate the associated uncertainties.

Moreover, the participants recommended performing a common benchmarking exercise for both in-situ measurements and destructive analysis. By considering for example a piece of concrete that can be measured in-situ before taking representative samples for laboratory analysis.

The way to derive the radionuclide fingerprints and their associated scaling factors is that described in Section 3.2 but correlations are established either by the available experimental data or by means of numerical simulations.

When HTM radionuclides are predominant, various options can be followed:

- Assigning all of the dose rate to one of the easy to measure radionuclides;
- Assigning all of the dose rate successively to each easy to measure radionuclide;
- Carrying out a gamma spectrometry to differentiate between the different easy to measure radionuclides.

The choice of one or other of these options depends a lot on the radioactivity level.

Otherwise, minimum detectable activity concentrations are declared in the case of the no presence of hard-to-measure radionuclides (i.e., after a cooling period  $\gg 10$  half-lives). Sometimes these radionuclides are simply ignored. Nonetheless, the characterisation can be referenced to another parameter than radioactivity, for example through a calorimetric approach.

One of the main difficulties that may be encountered in the definition of a radionuclide vector is the multiplicity of subsystems with different operating conditions. A functional analysis of each of these subsystems may allow grouping them by classes linked with elementary functions to limit the number of applicable radionuclide vectors. Sometimes, this number is simplified by itself after the ageing process. Other negative issues are:

- Samplings in constrained environments;
- Few statistics;

- Samples are not representative enough;
- No evident correlations between ETM and HTM radionuclides;
- Diffusion or migration of radionuclides ( $^3\text{H}$ ).

What is the most desirable and feasible is to set a global radionuclide vector for every nuclear facility but it is not the case in reality.

The participants considered that chemo-toxicity is independent of radio-toxicity. To accomplish the ANDRA technical specifications, toxic materials or substances can be assessed separately after the inventory of the radioactive waste.

They also clarified that the available experimental data are often complemented by numerical simulations of the neutron activation in reactor components, especially within reactor vessel and its peripheral structures. The case of graphite moderator blocs is a good example of application, where the measurements are used to modify the calculation hypotheses (i.e., adjustment of the impurity chemical concentrations).

As in the Belgium case, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for but the participants shared the following case-by-case feedbacks:

- Subsequent clearance process: Initial characterisation is used to build the Decommissioning Safety Report. This report is used to define the strategy of dismantling, to optimize the schedule and also to study the environmental impacts.
- Waste classification and waste treatment process: The pre-decommissioning characterisation allows anticipating casks supply. This latter can be modified during the effective waste production through direct measurements of ETM radionuclides. Only these last measurements are used to finally classify the waste and to declare the corresponding radionuclide inventory.
- Planning decontamination actions: The pre-decommissioning characterisation is used to define the optimum dismantling scenario and working conditions (individual protection, occupational dose rates, etc.).
- Planning site remediation actions: Cartography based on in-situ measurements and geo-statistical approaches can be efficient to localize radioactive hotspots and some particular zones where to sample cores in order to evaluate contamination in depth.
- Performing the final site release process: Cartography is taken as a reference to demonstrate that the removal of residual radioactivity levels during dismantling activities. However, a last cartography may be also performed before the final site release process (see the ASN guide n°14<sup>23</sup>).

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<sup>23</sup> Available at: <http://www.french-nuclear-safety.fr/References/ASN-Guides-non-binding>

- Estimating radiation doses to workers during the decommissioning activities: With the help of the updated 3D models and numerical simulations that have been previously validated with in-situ measurements, occupational dose rates can be computed for each intervention scenario.
- Estimating the environmental impact during the decommissioning activities: The pre-decommissioning characterisation allows defining the necessary controls to limit any environmental impact during the decommissioning activities.
- Estimating the amount of wastes produced during the decommissioning activities: The pre-decommissioning characterisation can be used to estimate:
  - activated material and structures,
  - the extent and levels of the radioactive contamination
  - radioactive contaminated structures ;
  - volumes of wastes and their classification, and
  - potential radioactive waste to be produced during the decontamination process.
- Estimating of decommissioning costs: Costs are estimated by means of a dedicated IT tool.

Finally, the participant confirmed that operational characterisation is carried out during the dismantling process, but without providing any further precision about how it is used for estimating radiation doses to workers during the decommissioning activities.

### 4.3 Germany

Following the Fukushima disaster in March 2011, the German Government decided to abandon the use of nuclear power plants to meet its energy needs by gradually phasing them out. Figure 6 gives an overview of the nuclear power plants under decommissioning in Germany, as well as those already either dismantled, permanently shutdown but awaiting granting of the decommissioning licence, or in operation with the foreseen end dates.

In addition to the power and prototype reactors, more than 30 research reactors of various size as also tens of nuclear fuel cycle facilities were shutdown and have been or will be decommissioned.

The legal framework for the decommissioning of German nuclear facilities results from the national Atomic Energy Act (AtG). It stipulates that nuclear decommissioning is subject to licensing by the competent authority, allowing two different strategies to be followed by the facility operator: immediate dismantling or dismantling after safe enclosure. Most operators have opted for dismantling immediately.

Last year, Germany announced a milestone in the complete dismantling of the reactor pressure vessel of the Obrigheim nuclear power plant. After 36 years of operations the plant was shutdown in 2005 and has been in the process of dismantling since 2008.

The physical nature, composition and estimated volumes of radioactive wastes generated in the above decommissioning programmes are detailed in the report published by the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety on August 2015 (BMUB, 2015).

The specific difficulties and obstacles during the above decommissioning programmes are:

- Disposal uncertainties, costs and technical requirements<sup>24</sup>: a spent fuel repository has not yet been determined and there is a huge delay regarding the Konrad mine disposal expected for low and intermediate level wastes. Liquids and bitumen are not yet allowed.
- Licensing process: the scale of the national decommissioning programmes overwhelmed the regulatory departments and most of them are considerably delayed.

The sought radionuclides are the ones listed in Sections 3.1.2 and 3.1.3 and most of the radiochemical analytical methods expounded in Section 2.1.3.2 are applied to analyse representative samples.

In-situ measurement techniques commonly used are: dose rate cartography, alpha/beta surface contamination and gamma spectrometry.

The questionnaire participants agreed with Section 3.1.2 in the sense that measurement of impurities is very important to well assess the extent and levels of the neutron activation effects by means of numerical simulations.

Regarding the protection levels to be targeted in terms of activity concentration and associated uncertainty for each of the declared radionuclide, the participants provided the following responses:

- Depending on the measurement goal but most of the acceptance criteria are given in the German standards DIN 25457 about the activity measurement methods for the clearance of radioactive substances and nuclear facility components (DIN, 2013a; DIN, 2013b; DIN, 2014; DIN, 2017a; DIN, 2017b).
- 95% of radioactive wastes have to be declared, only 5% of them are allowed to be a maximum 10% above the declared radioactivity. Uncertainties associated to the HTM radionuclides are addressed through a conservative approach so that they are mostly over- than underrated.

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<sup>24</sup> GNS (*Gesellschaft für Nuklear-Service*) has been recently transferred its interim storage activities to BGZ (*Bundes Gesellschaft für Zwischenlagern*), which is a new joint venture fully managed by the federal government.

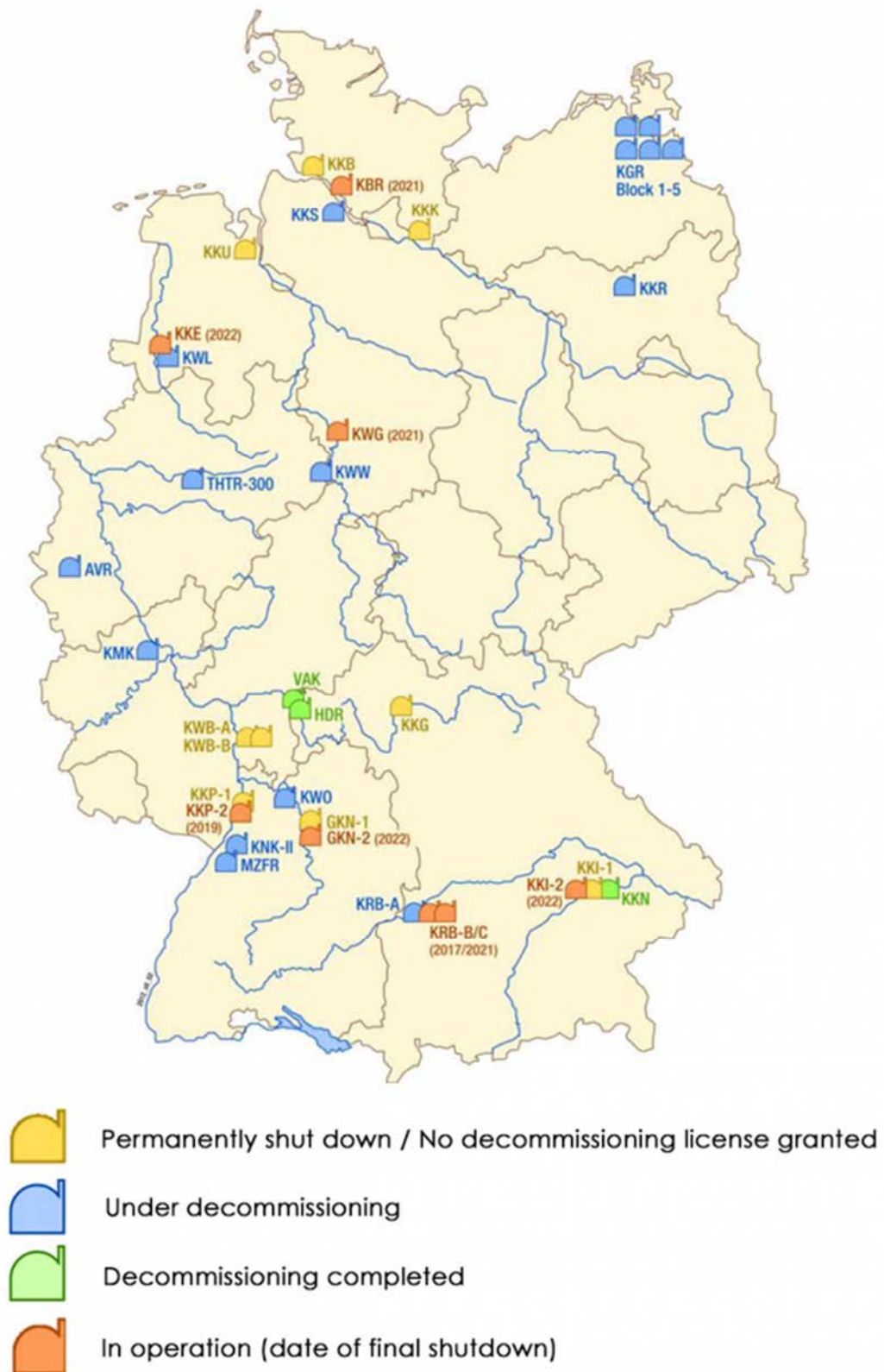


Figure 6: Overview of the nuclear power plants in Germany (source: IAEA).

Concerning the quality assurance plan, the participants assured that schedules have to be respected. Detailed and transport documentation are crucial because they are constantly checked by the German authorities.

In their opinion, the specific objectives of a benchmarking exercise for both the laboratory analysis of samples and *in-situ* measurements needs a strict frame that must be followed by all the involved laboratories to get sound and reliable results.

The way to derive the radionuclide fingerprints and their associated scaling factors is that described in Section 3.2 but correlations have to be conservative with respect of different clearance options as well as to sensitivity of different measurement procedures. When HTM radionuclides are predominant, corrosion/activation products are correlated to  $^{60}\text{Co}$ , fission products (such as  $^{90}\text{Sr}/^{90}\text{Y}$ ) are correlated to  $^{137}\text{Cs}$ , Pu-isotopes are correlated to  $^{241}\text{Am}$  that in turn can be sometimes correlated to  $^{137}\text{Cs}$ , and for special clearance options, correlations are done on the sum  $^{60}\text{Co}+^{137}\text{Cs}$ .)

The main difficulty in defining a radionuclide vector for a given facility are:

- heterogeneous contamination/activation history of systems,
- different radioactive materials,
- different clearance options,
- different measurement strategies (e.g., in-situ gamma spectrometry versus surface beta contamination), and
- organisation problems with use of different radionuclide vectors.

What is the most desirable and feasible is to set a global radionuclide vector for every nuclear facility but, in reality, there are as many radionuclide vectors as the existing subsystems and periods.

Chemo-toxicity evaluation is also carried out since there are regular limits for this risk as well. Chemo-toxic vectors must be established to deliver confident results.

The participants remarked that the available experimental data are often complemented by numerical simulations of the neutron activation in reactor components, especially within reactor vessel and its peripheral structures.

Over again, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for:

- subsequent clearance process,
- waste classification and waste treatment process,
- planning decontamination actions,
- planning site remediation actions,
- performing the final site release process,
- estimating radiation doses to workers during the decommissioning activities,



- estimating the environmental impact during the decommissioning activities,
- estimating the amount of wastes produced during the decommissioning activities, and
- estimating of decommissioning costs.

#### 4.4 Italy

Italy, which was a pioneer of civil nuclear energy, definitively discarded this sector by closing its last operational NPP in 1990 following a nationwide referendum held on November 1987, after the Chernobyl accident.

The history started at the end of 1946, with a small centre for nuclear energy research, called *CISE*<sup>25</sup>. Afterwards, the National Committee for Nuclear Research (CNRN<sup>26</sup>) was established in 1952 to become the National Committee for Nuclear Energy (CNEN<sup>27</sup>) in 1960 and the National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA<sup>28</sup>) in 1982.

In the early 1960s, three nuclear power plants from different companies (Westinghouse, General Electric and Npcc) were commissioned at Trino Vercellese, Sessa Aurunca and Latina. Meanwhile, in 1972 a 40 MWe prototype reactor of Italian design, called CIRENE, was built, tested, but never operated. The construction of the 4th NPP near Caorso was ordered one decade later and completed in 1978.

The phase out involved all the operating NPPs (see Figure 7), two new ones almost finished (Montalto di Castro 1 & 2) and the PUN (Progetto Unificato Nucleare) reference design for the construction of six PWRs plants. Other nuclear facilities that were closed are: 2 pilot plants for spent fuel reprocessing, located in Saluggia and Trisaia, one fuel fabrication facility at Bosco Marengo and several small experimental reactors distributed in different research centers or universities.

In 2008, government policy towards nuclear changed and an ambitious program was planned to increase the associated power share up to 25% of the country's electricity generation capacity by 2030. However, this policy was once more rejected after another nationwide referendum held on June 2011. Since then, Italy has been engaged in decommissioning, under the responsibility of Sogin (*Società Gestione Impianti Nucleari*, Nuclear Plant Management Company), its four nuclear power reactors and associated nuclear fuel-cycle facilities. Plans for waste management rely on the development of a National Radioactive Waste Repository for the disposal of low- and medium-level waste and interim storage of high-level waste.

The current decommissioning activities also include a high-energy cyclotron at the JRC-Ispra site (Characteristics: protons and alphas up to 38 MeV, deuterons up to 19 MeV and <sup>3</sup>He up to 53 MeV; current 50  $\mu$ A).

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<sup>25</sup> Centro Informazioni Studi ed Esperienze.

<sup>26</sup> Comitato Nazionale per le Ricerche Nucleari.

<sup>27</sup> Comitato Nazionale per l'Energia Nucleare.

<sup>28</sup> Energia Nucleare ed Energie Alternative.



**Figure 7: Overview of the nuclear facilities under decommissioning in Italy (source: IAEA).**

According to the questionnaire participants, approximate estimation of volumes of radioactive wastes and other contaminated materials at national level are: 90 000 m<sup>3</sup>, 60% of which are from power reactors and 40% from other installations. Moreover, they consist of 75 000 m<sup>3</sup> of VLLW and LLW along with 15 000 m<sup>3</sup> of ILW and HLW.

In their opinion, the major difficulty is the long delay in the authorisation process that makes harder any attempt to make a time and budget planning. Another problem derives from the fact that practically all the national nuclear industries ceased their activity after the referendum of 1987, so there is a very limited market.

They expressed that the main objectives to be achieved during the radiological and/or chemical characterisation of the considered nuclear facility and its nearby soils are:

- To collect most of information available in order to plan the decommissioning activities and the related waste management; this includes (but not limited to):
  - Estimation of produced waste in term of type of material, amount (mass and volume), physical form and radiological content
  - Planning of waste handling/treatment/conditioning facilities
  - Dimensioning of storage needs
  - Planning of activities (including time schedule and human resources)
  - Estimation of cost



- To detect evidence of leakage or spillage, identify hotspots and plan remediation actions in view of unconditioned site release (green-field<sup>29</sup>).

Because of the varied nuclear facilities subject to a decommissioning programme in Italy (research reactors, pilot reprocessing plants, hot cells for post-irradiation analysis, high-energy cyclotron for radioisotope productions, waste handling installations), the range of potential radionuclides is extremely wide. It includes:

- Spent fuel with all trans-uranium elements (U, Np, Am, Pu, Cm) and main long-lived fission products (<sup>137</sup>Cs, <sup>90</sup>Sr/<sup>90</sup>Y, <sup>152</sup>Eu, ...);
- Activation products from stainless steel (<sup>60</sup>Co, <sup>59</sup>Ni, <sup>63</sup>Ni,...), graphite (<sup>14</sup>C, <sup>36</sup>Cl) and heavy water (Tritium);
- Specific radionuclides connected with radiopharmaceutical production.

In line with what has already been pointed out in Section 3.1.2, the participants specified that impurities are measured for instance in the composition of the concrete of the biological shielding in order to assess the extent and levels of the neutron activation effects by means of numerical simulations.

Regarding the protection levels to be targeted in terms of activity concentration and associated uncertainty for each of the declared radionuclide, there are two major metrological challenges:

- The strong requirement in extremely low MDA (minimum detectable activity) for clearance purposes;
- The uncertainty in NDA measurement for low energy gamma emitter, in particular for the key nuclide <sup>241</sup>Am.

For site mapping, the participants explained that they use to use a large plastic scintillator mounted on a caddy to perform a first screening of the entire area and to detect eventual radioactive hotspots. Then, samples are taken for each of the detected hotspot and in-situ measurements by means of a portable gamma spectrometer with geometry-dependent calibration like ISOCS<sup>30</sup>, ISO-CART<sup>31</sup> or equivalent, are performed as well.

Several of the radiochemical analytical methods expounded in Section 2.1.3.2 are applied in ISO/IEC 17025 certified laboratories (ISO, 2005) to analyse the above collected samples. The corresponding procedures are in place compliant with the standard for qualification of the technique and of the personnel, including calibration procedure and data management. In turn, the validation of in-situ measurement techniques are carried out through inter-comparison exercises.

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<sup>29</sup> The green-field term is understood to be an end state for a decommissioned nuclear facility which allows the land to be released from regulatory control, whereas the brown-field term is used when the corresponding site remains under regulatory control, and the land's reuse and redevelopment is subject to certain restrictions.

<sup>30</sup> Further information available here: [http://www.canberra.com/products/insitu\\_systems/isocs.asp](http://www.canberra.com/products/insitu_systems/isocs.asp)

<sup>31</sup> Further information available here: <http://www.ortec-online.com/products/radiochemistry-health-physics-research-industrial/waste-assay-systems/iso-cart-85>

Concerning the quality assurance plan, the participants said that they use to operate in conformity with ISO 9001 standards (ISO, 2015).

The specific objectives of a benchmarking exercise for both the laboratory analysis of samples and *in-situ* measurements, should be:

- Adequateness verification of the available technical means, methodologies and analytical procedures to reach the objectives of plant characterisation;
- Inter-comparison of different techniques;
- Inter-comparison of different laboratories;
- Better assessment of measurement uncertainties;
- Needs identification of innovative/improved techniques;
- Measurements validation of challenging samples and/or matrices;
- Assessment of potential broader use, namely towards radiation imaging.

The way to derive the radionuclide fingerprints and their associated scaling factors is that described in Section 3.2 and must follow the national standard UNI 11194 (UNI, 2006). Care is taken to collect a sufficient number of samples where the HTM radionuclide is contemporary present accompanied by an ETM one. Then, the collected samples are properly analysed in specialized laboratories and average ratios (correlation factors) are computed together with their associated dispersion. Such correlations are considered valid if the observed dispersions are below a defined value depending on the number of analysed samples (e.g., <8 in 20 samples). When HTM radionuclides are predominant, a criterion of physical plausibility is generally applied by preferring, for instance, the correlation of fission products with  $^{137}\text{Cs}$  and activation products with  $^{60}\text{Co}$ . Otherwise, minimum detectable activity concentrations are declared in the case of the no presence of hard-to-measure radionuclides (i.e., after a cooling period  $\gg 10$  half-lives).

The main difficulties to be usually faced when defining a radionuclide vector for a given facility are:

- HTM radionuclides not correlating with any ETM, so that either the maximum concentration in all samples or, when possible, derive a normal distribution are assumed; and
- The number of positive matches, where both the HTM and an ETM are contemporary detected in a sample, is not sufficient to satisfy the statistical criterion. This can be solved with further sampling, but in many cases the required number of samples becomes unrealistic.

In small nuclear facilities a global radionuclide vector can be targeted but this is not the case in complex ones where there are in reality as many radionuclide vectors as the existing subsystems (e.g., research reactors with associated hot-cells and own radiochemical laboratories).

The participants clarified that correlations between chemo- and radio-toxicity are not often considered. In fact, chemo-toxic substances are dealt separately with identical isotopic vectors as non-toxic materials from the same nuclear facility.

They likewise confirmed that numerical simulations using Monte Carlo codes are habitually endeavoured of the neutron activation in reactor components.

Once more, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for but the participants shared the following case-by-case feedbacks:

- Subsequent clearance process: The definition of the radionuclide vectors is common to waste characterisation and to the clearance process.
- Waste classification and waste treatment process: The pre-decommissioning characterisation is used to estimate the amount of wastes that is expected to fall in the different categories. In any case the final attribution of each item will be done with individual NDA measurements. It is also used to estimate the applicability and expected performance of waste treatment processes, typically for volume reduction, and evaluate them on a cost/benefit analysis. Typical treatments envisaged in this aspect are: super-compaction, decontamination by abrasive blasting, metal melting, incineration, etc.
- Planning decontamination actions: The pre-decommissioning characterisation is used to evaluate the opportunity to perform or not specific decontamination actions. The estimated cost for disposal of non-treated item is compared with that of the treatment plus disposal (or clearance cost) of the decontaminated item and residue.
- Planning site remediation actions: Cartography is used mostly to identify contaminated areas and to give a preliminary assessment of the extension of the remediation action, which is then planned on a case-by-case basis.
- Performing the final site release process: Cartography gives initial information. In any case a complete site survey will be redone after the decommissioning before the final site release.
- Estimating radiation doses to workers during the decommissioning activities: This is a marginal use. In any case estimations of radiation doses to the workers during the decommissioning activities are assessed through specific measurements done by the radioprotection services. In Italy there is a clear distinction between measurement done for radioprotection purposes and those for operational activities.
- Estimating the environmental impact during the decommissioning activities: The pre-decommissioning characterisation feeds the estimation of potential releases during the dismantling activities both in normal and in hypothetic accidental conditions.
- Estimating the amount of wastes produced during the decommissioning activities: The pre-decommissioning characterisation is one of the primary tools to estimate the amount of waste produced. Indeed not the global amount but the expected attribution of waste to the different streams: potentially clearable, expected waste VLLW/LLW/ILW/HLW.
- Estimating of decommissioning costs: The pre-decommissioning characterisation provides a source of useful information for the estimation of a variety of different components of the

costs associated to decommissioning and related waste management, for instance (not exhaustive):

- Estimation of the waste volumes expected in the different categories are used to evaluate the waste disposal costs;
- Estimation of performances of waste treatment and volume reduction processes are used to evaluate the need of treatment facilities and the cost of related infrastructures, the cost of outsourced services and eventually the savings on disposal costs;
- Cartography of buildings can be used to estimate how much of the material generated by demolition will be clearable or generate waste;
- Cartography of land will give indication of the need of remediation actions and the costs of the site release.

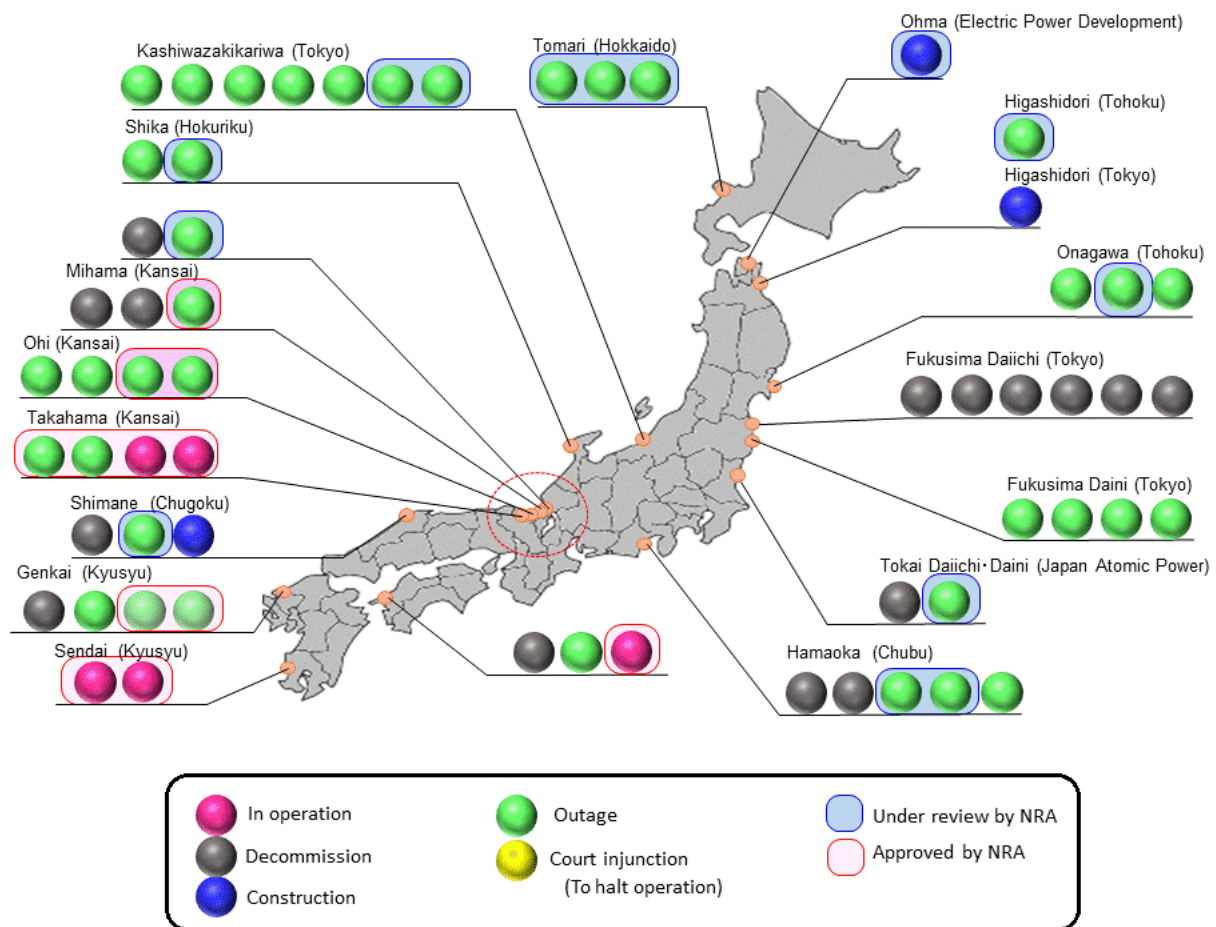
Finally, the participant indicated that there is NOT operational characterisation of the nuclear facility during the dismantling process.

#### **4.5 Japan**

The Fukushima disaster in March 2011 has been a decisive turning point in Japan, stopping at once all of the operational NPPs at that time as well as a vast expansion program for the construction of new ones to increase the associated power share up to 60% of the country's electricity generation capacity. Subsequently, the reformed regulatory agency, the Nuclear Regulation Authority (NRA), introduced, together with very strict requirements for plant safety and disaster resilience, a 40-year maximum operational limit for NPPs, with the option of a single extension for another 20 years. Today, there are 15 Japanese nuclear reactors, together with the TOKAI reprocessing plant, planned to be decommissioned for the next years (see Figure 8).

While the operator of each nuclear facility is the responsible for the management of decommissioning activities, NRA is the only competent authority to define the national strategy in this domain. Such a strategy consists of: a) site preparation (including site characterisation, defueling and decontamination), b) safe storage to reduce the radioactive inventory in the reactor through natural decay processes, and c) final deconstruction/dismantling processes.

The questionnaire participant estimated that between 300 000 and 1 050 000 metric tons of solid wastes, mostly concrete and metals, will be generated during the dismantling of the above nuclear facilities, near 98% of which can be considered as conventional or below the clearance levels to be disposed of, indeed recycled. The remaining 2% are expected to be radioactive containing a wide variety of radionuclides. Some details about the clearance levels of such radioactive wastes and their classification are given in the recent report of the EU-Japan Centre for Industrial Cooperation (Schmitter, 2016).



**Figure 8: Status of the Japanese nuclear power plants.**

The disposal of decommissioning waste poses a serious problem, with existing disposal concepts currently relying on on-site storage for most of the radioactive wastes. Furthermore, many technical – and some regulatory – issues for the decommissioning of the Fukushima Daiichi site remain unsolved, particularly due to the still limited understanding of the situation inside the damaged reactors. Other difficulties are the cost of clearance procedures and the acceptance of cleared materials in the general market.

The participant is fully focused on a comprehensive characterisation, including cartography, of the Fukushima damaged reactors. For this purpose, he would like to fix almost all the unknown parameters, namely towards the optimum in-situ measurement and sampling techniques under excessively extreme radioactive environments.

Many radionuclides are targeted and the whole study is very complex since it may consider the radioactive contamination during the operational period of the accident as well as the severe consequences of this latter and further impacts of the nuclear fuel debris.

In addition, the participant insisted that numerical simulations must be conducted to evaluate the neutron activation such as in core internals. In the first instance, manufacturers were asked to provide as much information as possible about the presence of impurities in most of the construction

materials and some cases identical metals were newly reanalysed. All the gathered data were used as calculation inputs for ORIGEN code (Bell, 1973). Because of the raised radiation dose levels in core internals, very few samplings were carried out to be compared with the calculation results. For the same reasons, there was not yet a specific protection level regarding the measured radionuclide concentrations and their associated uncertainties. This was left for future characterisation activities.

Both the in-situ measurement techniques and radiochemical analytical methods are described in the report No. 7305 of the Nuclear Energy Agency (NEA, 2016). Their experimental qualification is normally based blank samples with known characteristics.

In regulatory framework, the management system should be included in the preservation rule of nuclear facility. Analysis data of waste are used to check the compliance with the waste acceptance criteria of the disposal facility. These data, conjointly with the QA/QC/QM guideline and its observance situation, are constantly supervised by the regulator in the safety inspection.

Concerning the specific objectives of a benchmarking exercise for both the laboratory analysis of samples and *in-situ* measurements, the participant stated the importance of the uncertainty assessment and their applicability.

The correlation between HTM to ETM radionuclides is performed in Japan by collecting samples for radiochemical analyses and applying the same approach as that described in Section 3.2. When HTM radionuclides are predominant, efforts are made through numerical simulations or precise laboratory analysis, particularly by considering another element with almost the same chemical property. Conversely, the full absence of HTM radionuclides facilitates a lot the entire characterisation process.

The main difficulty in defining a radionuclide vector for a given facility is that the high radiation level due to the  $^{137}\text{Cs}$  presence and the unknown characteristics of samples, which complicate the corresponding chemical separation process.

What is the most desirable and feasible is to set a global radionuclide vector for every nuclear facility but, in reality, there are as many radionuclide vectors as the existing subsystems and periods.

The participant remarked that assessment of chemo-toxicity just started in the Japanese nuclear facilities subjected to dismantling, so that it is very premature yet to establish any kind of correlation between chemo-toxic and radiological materials or substances.

He explained that besides numerical simulations of the neutron activation in materials, the theoretical estimation of the radionuclide migration behaviour is also endeavoured.

Another time, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for:

- subsequent clearance process,
- waste classification and waste treatment process,
- planning decontamination actions,
- planning site remediation actions,



- performing the final site release process,
- estimating radiation doses to workers during the decommissioning activities,
- estimating the environmental impact during the decommissioning activities,
- estimating the amount of wastes produced during the decommissioning activities, and
- estimating of decommissioning costs.

Finally, the participant indicated that there is NOT operational characterisation of the nuclear facility during the dismantling process.

#### 4.6 Spain

Spain related experience in nuclear decommissioning includes both the Vandellós I and José Cabrera nuclear power plants. Vandellós I is a 480 MW<sub>e</sub> gas-cooled graphite-moderated reactor, twin to those of Saint Laurent des Eaux A-1 and A-2 in France. It was suddenly stopped on 19 October 1989 due to a fire in the high-pressure turbine (Figure 9), with no radiological impact but making its reparation uneconomically viable. This led to a ministerial order on 28 January 1990 to permanently shutdown the facility, after 17 years operation, by equally specifying the dismantling conditions (i.e., removing the spent fuel and conditioning the wastes produced during its operation). The accepted strategy also included dismantling of non-essential or redundant systems and peripheral structures located outside the reactor vessel. In 2003, the facility was decommissioned to what is known as the IAEA stage 2 (safe storage under surveillance of localized/isolated equipment). Since then, no further actions have been taken with respect to the vessel in which the unfuelled reactor remains now confined until completion of the waiting (dormancy) period of about 30 years.

In turn, the decommissioning of José Cabrera, which is a pressurized-water reactor with a nominal power unit of 150 MW<sub>e</sub> and almost 38 years of normal operation, has been initiated in 2010 and it is scheduled to be finished by 2018. The third nuclear power plant that recently started its decommissioning programme, after more than 46 years of normal operation, is Santa María de Garoña, which is a boiling-water reactor with a nominal power unit of 440 MW<sub>e</sub>. It was the oldest Spanish nuclear power plant in service.

In accordance with the current regulations, ENRESA (the National Radioactive Waste Company) is the sole responsible for the management of all activities arising from the decommissioning of nuclear facilities in Spain. Nowadays, the decommissioning strategy finally adopted in Spain involves immediate and complete dismantling (IAEA stage 3), starting 3 years after shutdown, with an expected duration of 10 years. In the case of Vandellós I, Level 3 is expected to be implemented in 2028.



**Figure 9: Picture showing the damage of the Vandellòs 1 turbine after the fire incident on 19 October 1989 (Source: *El País* newspaper)<sup>32</sup>.**

According to the questionnaire participant, the specific difficulties and obstacles during the above decommissioning programmes are basically the schedule, budget and resources involved. He likewise indicated that negotiation with authorities is sometimes another main issue.

The participant remarked that the main objectives to be achieved during the radiological and/or chemical characterisation of the considered nuclear facility and its nearby soils are:

- to develop a sampling process of the site/facility in order to have representative radionuclide vectors of the different places and natures of the waste,
- to determine the radionuclide vector of the soils,
- to quantify the scope of the remediation activities,
- to define the kind of decontamination to apply, if any is going to be performed,

In his opinion, characterisation of soils must include the radiological scanning analysis of the whole surface of the site (first 15 cm depth) to evaluate the potential contamination at every place. The corresponding results, in conjunction with the available historical data, allow to improve the areas of major concern as also to assess the places and the number of the boreholes to be performed.

He commented that the source term has to be defined during the licensing process with the goal to know the radionuclides to be measured. This source term should be defined from the type of installation, the historical data and the samples collected during initial characterisation. The main ETM radionuclides to quantify in contaminated soils are  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  in lesser extent, whereas  $^{90}\text{Sr}$  as a HTM radionuclide could be quantified as well. But in general the radionuclides to quantify in soils must be the ones explicitly defined in the soils' source term. Habitually, ETM radionuclides are firstly measured and, by applying the scaling factor approach, the HTM ones are determined.

<sup>32</sup> Available at: [https://elpais.com/ccaa/2014/10/18/catalunya/1413655337\\_690826.html](https://elpais.com/ccaa/2014/10/18/catalunya/1413655337_690826.html)



In line with Section 3.1.2, the participant insisted that for activated materials (metals and concrete), the usual way to proceed is to equally measure their chemical impurities, and knowing the irradiation history, the neutron energy spectra flux/energy to determine, as well as the corresponding  $(n,\gamma)$  cross-sections a theoretical model is developed. Normally, it is very difficult to have the information of chemical impurities for old components and then there could be several valid alternatives that could complement each other:

- measuring the chemical composition of virgin material, if available, and
- collecting, if possible, active samples for radiochemical analysis, including impurities.

In Spain, virgin concrete samples have collected for chemical analysis in addition to radiochemical analysis of activated concrete, with the objective of having a complete picture of the neutron activation process, and of comparing the obtained results with the theoretical calculations. In addition, chips resulting from the cutting process of the activated metals have been radiochemically analysed, just to verify the neutron activation calculations.

The participant said that the starting point for the uncertainty evaluation is always that associated to the measurement of the most representative ETM radionuclide (radio-tracer) and as scaling factor uncertainties are also assigned to the considered fingerprints, the complete uncertainty propagation is determined for all the declared radionuclides.

In the case of in-situ measurements, the best approach for obtaining a dynamic analysis for soils is the use of gamma spectrometers (based on NaI, LaBr, CZT and/or HPGe detectors) mounted on a robust vehicle, allowing an accurate surface mapping of the site. It is very recommended the use of high precision GPS (10 cm accuracy). For boreholes, the best approach is the gamma analysis of the vertical profile of the left hole from the surface, because it is very difficult to measure the material of the borehole itself due to the different matrixes involved, and that the radionuclide concentration is usually measured in the finest part and extrapolated to the rest, given a conservative quantity. For the facility measurements, inside buildings and so on, there are many possibilities of measurements, ranging from dose rate to gamma spectrometry. For large walls of difficult access, inside or outside buildings, one option is the use of gamma devices mounted in a drone that is able to position in the right place. Another alternative inside buildings is the use of gamma cameras that give a global vision of the whole room/wall radiological situation saving cost, time and unnecessary people dose exposure. Further measurements of the radiation dose rate and alpha/beta contamination are also carried out.

To verify the suitability of the above devices, they are calibrated by applying suitable procedures and reference sources to simulate as best as possible the actual measurement conditions.

Concerning radiochemical analytical methods, the participant provided almost the same response as in the Belgium case (see Section 4.1) but stressing that, to be properly accredited in this specific domain, the different specialized laboratories have to fulfil the corresponding ISO and/or national standards.

For the most part, quality assurance methodology is applied for assuring the proper implementation and tracking of the process, reports, personal qualifications, calibration verification of devices, etc.

Relating to the benchmarking exercises laboratory analysis of samples and *in-situ* measurements, a recommendation was made to take profit from what has been done in the past, such as ENTRAP in the EU and LABONET in IAEA inter-laboratory projects.

The correlation between HTM to ETM radionuclides is performed in Spain by collecting samples for radiochemical analyses and applying the same approach as that described in Section 3.2. At the same time, the participant referred to ISO 21238 (ISO, 2007) and proposed that if there is a predominant presence of HTM radionuclides their associated scaling factors must be determined.

With respect to no presence of HTM radionuclides, the participant insisted that there are always long-lived ones (such as  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ , etc.) to be reported in the repository of the wastes. In his appreciation, by establishing an appropriate cooling period (i.e.,  $\gg 10$  half-lives), all the short-lived HTM radionuclides will not be present in a meaningful amount, due to the radioactive decay process, and they could then be discarded from the list.

The main difficulty in defining a radionuclide vector for a given facility is the collection of representative samples for radiochemical analysis, which in turn is a very difficult process. In addition, the cost analysis has to be optimized through the sampling strategy, going in a progressive approach for obtaining composite samples and covering as much as possible the entire range of the activity concentrations. Such approach allows the scaling factors to be applicable for all types of the generated wastes.

In most cases, there is a need to virtually partition the nuclear facility into different subsystems with potential distinct radionuclide vectors, which have to be verified by the sampling and analysis process.

In matter of chemo-toxicity, low- and intermediate-level waste disposal are assigned for hazardous materials, being the radioactivity the main concern of this kind of risk. For very low-level waste repository, both radiological and toxic problems are taken into account with similar importance. Anyway toxics chemicals must also be declared for the very low-level waste repository. Of course, there is an evident need to follow (track) the chemical limits for specific elements, and to define what is considered as hazardous waste in order to develop the stabilization process that must be applied.

As mentioned above, only theoretical calculations of neutron activation are verified by taking samples or *in-situ* measurements. Till now, radioactive contaminated materials, due to the sampling process developed, did not required numerical simulations, instead constant monitoring allowed to control the corresponding time variation.

Yet again, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for but the participant shared the following case-by-case feedbacks:

- Subsequent clearance process: In the sense that all the obtained results are valid in the whole clearance process, disregard the cooling period if taken. Moreover, the participant agrees with idea that also the measurements taken during operational life of installation are useful for the dismantling period. For him, characterisation is a progressive task, starting since the facility is being commissioned till the end of dismantling activities.

- Waste classification and waste treatment process: The same approach as before is also valid here, namely by plotting the whole data of a specific HTM radionuclide along with the candidate ETM radionuclide and correcting all of them for their own radioactive decay, it is possible to detect correlations and to determine their associated scaling factors.
- Planning decontamination actions: There are many important aspects that can be well addressed if the process of pre-decommissioning facility characterisation is as detailed as possible in order to better define the actions to apply later on in this domain. To highlight such aspect, the example of Vandellòs 1 is given in where the initial characterisation has permit to fittingly vision its global radioactive contamination status and to firstly proceed in cleaning the primary circuit before starting the dismantling process. This action saved time and a lot of radiological dose to the workers. Other action taken was the decontamination of low-level waste (LLW), basically metals, with the objective of decreasing its classification to very low-level waste (VLLW) to be sent to other kind of repository.
- Planning site remediation actions: With the help of geostatistics, which is a useful tool to quantify the remediation work to be done, especially when experimental data are structured. Otherwise, classic statistic approaches (e.g., full nugget effect) would provide the same results.
- Performing the final site release process: Previous boreholes are useful during the site release phase when their depth are greater than the depth of the remediation finally done. In addition, during the remediation actions performed in Vandellòs 1 everything has been measured, for instance, by collecting soil samples to decide the next step (washing or clearance measurements) and the remaining holes were measured again in a detailed way for demonstrating the fulfilment with the approved limits.
- Estimating radiation doses to workers during the decommissioning activities: Characterisation also means the performance of a radiometric measurement of the plant in order to know the radiation dose rate to be faced by workers involved in the different tasks during the dismantling project.
- Estimating the environmental impact during the decommissioning activities: In Spain, pre-decommissioning facility characterisation is required during the licensing process with the objective of estimating and calculating those issues needed to obtain the license for the dismantling project.
- Estimating the amount of wastes produced during the decommissioning activities: Characterisation is used to determine the radionuclide inventory in different places and systems in order to quantify the amount of wastes to produce. It has to be focused to determine as better as possible the order of magnitude of the extension of the contamination/activation by means of sampling/radiometric measurements. Detailed diagrams of the plant are required to better quantify the mass involved.
- Estimating of decommissioning costs: Once both the extension of the radioactivity and the mass involved are estimated by initial characterisation, the next step is to develop the

schemes to apply in the different places (soils, systems, etc.). The radiological risk and the engineering aspects are the main challenges to combine for estimating the decommissioning costs.

Finally, the participant confirmed that operational characterisation is carried out during the dismantling process. He gave the example of the Radiation Protection Service from the same nuclear facility has to constantly know the means and resources to be used during the dismantling activities. This supposes that besides initial characterisation, additional measurements are taken continuously to know the evolution of the involved places, areas, etc.

#### **4.7 Ukraine**

On 26 April 1986, the Chernobyl nuclear power plant suffered the worst nuclear accident in history until then when an explosion took place in its 4<sup>th</sup> water-cooled graphite-moderated reactor due to combination of successive inherent failures and human errors (see Figure 10). The damaged reactor building was immediately sealed by means of a concrete shelter after dropping there thousands metric tons of sand, lead, clay, and boric acid. Meanwhile, as the three remaining reactor units were vital to Ukraine's electricity needs, they continued to operate for some years (unit 2 shutdown in 1991, unit 1 in 1996 and unit 3 in 2000). Later on, such physical protection became unstable and a new safe confinement (i.e., arch-shaped steel structure) was added on 29 November 2016. The decommissioning of units 1-3 is being carried out separately from that of the destroyed unit 4, which is expected to take many years longer to complete.

The current decommissioning programme is planned to accomplish different stages before 2028. The first stage is to refurbish the water supply system for the plant's fire protection system. The second stage will involve the dismantling of the pressure tubes and other equipment of units 1 to 3. The reactors of units 1 and 2 will then be put into a safe and controlled state. In the fourth stage, the roof of each reactor hall of units 1 and 2 will be refurbished while their fuel handling machines will be dismantled. The same intervention steps will afterwards be applied to the third unit. At the end, the contaminated equipment will be removed from the three units during the period between 2028 and 2046, while the reactors themselves will be dismantled between 2046 and 2064.

The above decommissioning programme will also include the wet storage facility of spent fuel (ISF-1) and solid radioactive wastes (HTO-1), both objects located on the same Chernobyl site. Right now, there are 21217 fuel assemblies in ISF-1 removed from the above undamaged units (WNA, 2016). These fuel assemblies will be suitably cut and packaged within double-walled canisters, which in turn will be welded after being filled with inert gas and transferred into the new ISF-2 dry storage facility to be stored over there for at least 100 years. HTO-1 was put into operation after the accident and consists of a concrete building divided into 6 sections filled with low, medium and high level wastes (LLW, LMW and HLW). The filling process in each of section was disorderly executed by throwing down directly the radioactive contaminated materials and pouring concrete thereafter. Such wastes must be retrieved, conditioned and moved to new surface depository facility planned for long time storage for low and medium level wastes as well as a temporary storage for high level waste.



**Figure 10: Chernobyl reactor 4 the accident (source: Wikipedia).**

Approximate estimations about the physical nature, composition and estimated volumes of radioactive wastes generated in the above decommissioning programmes are:

4. In the HTO-1 storage facility<sup>33</sup>:

- Group I<sup>34</sup> waste – 1069 m<sup>3</sup> and 0.11 TBq
- Group II waste – 926.5 m<sup>3</sup> and 4.11 TBq
- Group III waste – 417.65 m<sup>3</sup> and 95.67 TBq

5. The Chernobyl NPP site and object “Shelter” (Panaciuk et al., 1999; Panaciuk et al., 2002):

- The volumes of solid radioactive waste that located in the ChNPP technogenic layer (without the Shelter territory) assessed as 800000 m<sup>3</sup>.
- In the technogenic layer of “Shelter” there are 371000 m<sup>3</sup> of radioactive waste including 25000 m<sup>3</sup> of high level waste.

<sup>33</sup> Waste stored in the HTO consist of the protective clothing, metal, heat-insulation, building materials, plastic, wood, paper and so on. There are several hatches for access in compartments.

<sup>34</sup> The type and classification of Ukrainian radioactive wastes are given in Poyarkov (2013).



- On the area of West zone of “Shelter” by zones I and II located:
  - Low level waste – 9668 t or 4910 m<sup>3</sup>
  - Medium level waste – 215 t or 112 m<sup>3</sup>
  - High level waste – 185 t or 100 m<sup>3</sup>
- On the areas of credible device of foundation pits on the West from pioneer wall (areas 4A-1, 4A-3, west site 4E) by zones I and II located:
  - Low level waste – 3339 t or 1711 m<sup>3</sup>
  - Medium level waste – 49 t or 26 m<sup>3</sup>
  - High level waste – 109 t or 60 m<sup>3</sup>

It is a well-known fact, that the above decommissioning programmes have been faced with several problems related to the spent fuel storage and radioactive waste management. There was likewise no way in the beginning to have an access to the facility historical documentation or the accident reports to accurately estimate the amount of radioactive contaminated materials. Moreover, some construction defaults during construction of the plant, such as splitting of concrete layers, were never acted upon.

According to the questionnaire participant, the main objective to be achieved during the radiological and/or chemical characterisation of the considered nuclear facility is to determine the list the radionuclides of interest and their activity concentration. This allows the determination of the optimal decommissioning scenario as also the minimization of occupational dose and volume of generated radioactive wastes.

The most part of contaminated soils are characterized as low level waste and should be stored in a surface disposal of trench type. More often, corroboration of the presence of transuranic elements (TUE) is also necessary. In addition, analysis of chemical properties may considerably help defining possible procedures of decontamination and treatment methods.

The current Ukrainian Regulation classifies the radioactive wastes by groups (Poyarkov, 2013) and for which most of the sought radionuclides are already listed in Sections 3.1.2 and 3.1.3. These radionuclides are only measured on surfaces without accounting for chemical compositions of contaminated materials. In a case of necessity of more detailed characterisation, enhanced sampling and investigation of radioactive waste volume are carried out.

Regarding the protection levels to be targeted in terms of activity concentration and associated uncertainty for each of the declared radionuclide, the participant answered that the following radiation dose limits must be implemented for both the personnel and the general public during the radioactive waste treatment, storage and release from regulatory control:

6. No more than 40 mZv (i.e., 400  $\mu$ Sv/h) per year, or 10 mZv (i.e., 100  $\mu$ Sv/h) in condition of radiation accident, for the general public.
7. No more than 14 mZv (i.e., 14 mSv) per year for professionally exposed workers.



The main radionuclides that typically generate radiation dose are:  $^{90}\text{Sr}/^{90}\text{Y}$ ,  $^{137}\text{Cs}$ ,  $^{241}\text{Am}$  and  $^{239}\text{Pu}$ .

Criteria have been established for levels of seizures from regulatory control in terms of the activity concentration for every radionuclide group: alpha-emitting transuranic elements; alpha-emitting excluding transuranic elements; beta and gamma-emitting except for the following:  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{36}\text{Cl}$ ,  $^{45}\text{Ca}$ ,  $^{53}\text{Mn}$ ,  $^{55}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{93}\text{Nb}^m$ ,  $^{99}\text{Tc}$ ,  $^{109}\text{Cd}$ ,  $^{135}\text{Cs}$ ,  $^{147}\text{Pm}$ ,  $^{151}\text{Sm}$ ,  $^{171}\text{Tm}$ ,  $^{204}\text{Tl}$ .

Moreover, the participant sent a quite extended response, which is transcribed below, about how nuclear facilities are characterized in Ukraine. He specified that guidelines, adapted from the IAEA technical document IAEA-TECDOC-1092 (IAEA, 1999), are used to perform measurements aiming at assessing the radiation situation on the ground. For the purposes of gamma survey, equipment of mobile radiological laboratories is used, portable instruments with the possibility of binding the results of measurements to GPS coordinates.

### Engineering investigation

1. Determination the scope of works on conservation, decontamination and demounting of equipment.
2. Planning and carrying out the works on conservation, decontamination and demounting of equipment.
3. Carrying out of the prognosis estimations on determination of the amount and type of generated radioactive wastes.
4. Determination of equipment nomenclature that requires extension of resource.
5. Reducing the fire danger (removing of combustible and aggressive substances, deenergizing of electrical equipment), avoiding the influence of low temperatures on equipment catchment and chopping off of the systems, setting of the additional heating and so on.
6. Carrying out the calculations to estimate volumes of radioactive wastes.

Scope of engineering investigation should be minimal from the point of view of dose, financial and working costs, but sufficient for actualization of KIRO. In the framework of KIRO actualization the thermo-mechanical, electrical engineering equipment and construction elements, removed objects and materials that require additional measurements and calculations – should be investigated at the places of location.

### Methods of engineering investigation

1. As basis of engineering investigation in part of preparing the initial maps accepted the “systematic” method of gathering and forming the information by systems according to the list of the object technological systems.
2. Engineering investigation is account for analyses of existing data by results of previous investigations with following collation by the design, operational, technical documentation and with practical determination of other parameters at the places of equipment location.

3. The practical determination of needed parameters carry out at the places of equipment location taking into account:
  - the real composition of equipment;
  - failing technical parameters by tallies and brands;
  - overall sizes;
  - composition and volume of heat-insulation, combustible and aggressive materials and environments.

### **Procedure of preparation and fulfilment of engineering investigation**

1. Engineering investigation should be carried out in close compliance with the corresponding program by single dress-admittances, single order, removable tasks and so on.
2. By the results of documentation and data analysis obtained in previous inspections, should be filled columns of "Inspection carts" without any measurements in the places.
3. If it is necessary to carry out the preparatory operations at the places of equipment location (opening of thermal-mechanical equipment, illumination setting, so on) workshop-proprietor defines the possibility of given work carrying out by own forces or by personnel of other subdivisions.
4. During engineering inspection:
  - determine the engineering parameters of equipment and construction elements that have not been accounted for in the previous inspection and in operational documents;
  - determine the engineering parameters of equipment and construction elements that were changed after the previous inspections of elements and systems (reconstruction and dismantling);
  - determine the engineering parameters of equipment and construction elements in accordance with requirements of changed methods of calculation and classification;
  - inspection of compartments on the presence of removed objects and materials.
5. Determination of parameters of engineering state in accordance with what has been pointed out in "Investigation carts".
6. Once the investigation is finished in a given place, the "Investigation carts" should be supplemented with engineering parameters, which require additional calculations.
7. By the results of analyses of operational documentation and gathering of failed parameters the "System Investigation carts" are filled with inventory numbers.
8. These filled "System Investigation carts" are passed on a network by Email in accordance with "Chart of realization of Engineering inspection".
9. In accordance with initial data of "System Engineering Investigation carts" the "System Radiation Investigation Carts" are filled.

## Chemical investigation

Purpose of chemical investigation is the analysis of inner and exteriority equipment surfaces state relative to corrosion, amount and composition of sedimentations to obtain the results on prognostication of NPP Unit equipment state at the stage of operation stopping and on following decommissioning stages.

List of thermal-mechanical equipment (TME) inner surfaces that should be investigated determined by the following principles:

8. scope of chemical investigation of equipment inner surfaces should be as minimal from point of view of dose, financial and labour expenses, but should be sufficient for gathering of information needed for planning decommissioning activities;
9. to obtain the results of Unit equipment state prognostication the chemical investigation should be carried out for systems and piping that were decommissioned and emptied during the period beginning from Unit stopping of operation;
10. for analysis relative to corrosion, amount and composition of sedimentations the equipment should be investigated that have the worst indexes relative to amount of sedimentations;
11. chemical investigation of equipment inner surfaces is carried out jointly with radiation investigation, determination of radionuclide's composition realize by the NPP central laboratory (CRL).

Data of the radio-isotopic composition filled into the "Carts of system chemical investigations".

Under chemical investigation, determine the presence of sedimentations on the equipment inner surfaces, their chemical composition and thickness, together with the corrosion state of the equipment and piping inner surfaces.

By the results of Central Laboratory (CORA) designs "Protocol of chemical investigation" included into the "System Radiation Investigation Carts".

Information obtained as a results of joint chemical and radioisotope investigations is needed for following:

12. decision making relative to equipment preservation or passivation during the long time storage;
13. necessity estimation and methods define of equipment decontamination;
14. prognostication of the radioactive waste volumes during the equipment decontamination.

By the results of chemical investigations of equipment inner surfaces the "System Chemical Investigation Carts" are filled taking into account for data on radioisotope composition of sedimentations submitted by the Central Radiation Laboratory (CRL). Protocol of radioisotope composition of sedimentations.

## Radiation investigation

The purpose of radiation investigation data actualization is development of database relative to the Unit radiation state taking into account for:

15. levels of radioactive contamination and estimation of equipment and building constructions activities;
16. boundaries of radioactive contamination and degree of contamination by radionuclides of compartments, buildings and territory;
17. volumes and activity estimation of solid radioactive wastes presented in Unit at the moment of investigation;
18. radionuclide inventory of radioactive contamination of equipment and Unit building constructions.

Obtained results will be the basis for:

19. analysis of radiation state changes;
20. analysis of radiation state changes of equipment and systems which were decommissioned and emptied during the period from the previous investigation;
21. making of suggestions on ensuring the radiation safety under works relative to stopping operation and following stages of decommissioning;
22. making of suggestions on organization of Unit radiation defence (decontamination, equipment dismantling, establishment of sanitary barriers, developing of additional ventilation, limited access into compartments) to reduce the occupational dose and avoiding propagation of contamination for the set boundaries.
23. changes of Unit compartments zones taking into account for real state and changes of technological process at stage of operation stopping and following stages of decommissioning.

Scope of radiation investigation is determined by the Central Laboratory personnel under conditions of maximal information plenitude and authenticity.

Radiation investigation is carry out by the working commissions according to the Program requirements and ratified chart.

Under estimations of Unit radiation state using the presence methodologies should be carried out sampling and investigations of samples to check the authenticity of obtained results.

Under estimation of Unit radiation state using the calculation method should be taken into account for all known incidents at given object, related to the exit of radioactivity beyond the set boundaries.

Under estimation of Unit radiation state using the calculation method should be carried out sampling and investigations of samples to check the authenticity of obtained results.

Under measurements realization should be used the approved devices of radiation control, which should be metrological checked and verified. Devices and equipment that were not verified or checked, can be used only in a case of absence of verified devices in the indication mode.

Measurements of parameters of radiation state carry out in accordance with appropriate developed methodologies

### **Radiation investigation of equipment and compartments**

To improve the investigation performance and avoid the duplication, the radiation investigation should be carry out only after full completion of analysis:

24. data of results of previous investigations and preparation of carts for radiation investigation;
25. results of all dosimetric, radiometric and spectrometric investigations of given equipment and in given compartments for the last year;
26. data of incidents for given equipment related with exit of radioactivity beyond the design set boundaries.

As the results of analyses the scope of radiation investigation of equipment and compartments is determined. All compartments should be investigated independently from the zones.

Radiation investigation of compartments should be carried out jointly with radiation investigation of equipment located in given compartment.

Before the system's radiation investigations of equipment and compartment is necessary:

27. prepare the drawings of compartment's plans;
28. using data of "Engineering investigation carts" prepare in electronic format "Radiation investigations carts" for all investigated equipment, technological systems and compartments, with pointing of the equipment squares, construction elements and building constructions, removed objects;
29. analysis of sufficiency of presented equipment technical specifications to determine and calculate the needed parameters of radiation state according to the "Program" requirements;
30. forming of summary packages of "Radiation investigation Carts" for all equipment in all technological objects;
31. prepare chart of radiation investigation realization in the compartment determining the personnel group needed for investigation of given compartment, and submit this chart before the investigation beginning.

As a base of the radiation investigations of equipment and compartments approved method of commission investigation by the compartments (with participation of representatives of all subdivisions, whose equipment located in given compartment).

Under the radiation investigation realization the following parameters are measured and calculated:

32. dose level from gamma-irradiation in compartment and from equipment;
33. levels of radioactive contamination of inner and exterior surfaces of equipment, surfaces of the floor, walls, ceilings, service sites and so on;

34. amount of the removed radioactive contamination of inner and exterior surfaces of equipment, building constructions and so on;
35. spectral analysis of the radioactive contamination of surfaces;
36. specific and summary activity of equipment, building constructions;
37. activity and location of sources with increased dose level;
38. activity concentration and radionuclide composition of substances located inwardly of equipment and piping.

Under measurements realization in a case of homogeneity of investigated parameter (no more 50% difference of maximal value at 5 different points equipartitioned on the object square) – assumed using of mean parameter measured at the part of object and its distribution by all square. Otherwise calculation made by the single parts of contamination.

Investigation of compartments should begin from the dose field distribution scanning (determination of mean for given compartment dose level of gamma-irradiation) and determination of sources with increased dose level (more than two times larger than mean value for given compartment).

Systems investigations located in compartments and preparing of working places for investigations (including works on equipment opening) made by the “single dress-admittances” and “single orders”.

Under radiation investigations on the compartment plans drawings should be signed point with coordinates having an increased control level values and data submitted into the Central Laboratory.

During the process of actualization of radiation parameters of technological system's equipment the Central Laboratory personnel mark equipment having the (maximal exposure dose) levels higher than 10 mBer/h (i.e., 100  $\mu$ Sv/h).

### **Radiation investigations of buildings and structures**

Radiation investigation is made by the method of direct measures of parameters of radiation state of given object.

Before the radiation investigation it is necessary:

39. to become familiar with results of all dosimetric, radiometric and spectrometric measurements made in given building, structure or on the Unit territory during last two years;
40. prepare the drawings of external buildings and structures plans.

Under radiation investigation of buildings and structures should be measured and calculated:

41. dose level from gamma-irradiation in compartment and from equipment;
42. levels of radioactive contamination of accessible surfaces;
43. amount of the removed radioactive contamination of accessible surfaces (for surfaces and materials assumed obtaining of representative data);
44. spectral composition of the radioactive contamination of surfaces;



- 45. specific and summary activity of controlled objects;
- 46. activity and location of sources with increased dose level.

Scope of radiation control (number of measured points) in investigated building is chosen in according to the Program of radiation investigation.

Under measuring the radiation state parameters of buildings and structures it is strongly necessary to investigate all places of possible accumulation of radionuclides (rain flows, places of failed roof), and also places of possible propagation of radionuclides into the building constructions (places of radioactive liquids propagation on the surfaces, places with failed preventive coverage, places of accumulated rain water).

If the measured dose level is more than two times that on the nearby areas, estimation of local source activity should be performed.

As a result of radiation investigation fill the “Radiation investigation carts” and also clarified the previous results.

Under radiation investigations on the building's and structures plans drawings should be signed point with coordinates having an increased control level values and data submitted into the Central Laboratory.

Results of the spectrometric measurements after recalculation into summary activity of all meaningful radionuclides are described into the “Radiation investigation carts” signing the places of given radionuclide location and their percentile amount.

“Radiation investigation carts” developed by the systems and compartments, supplemented with data of summary and specific activity, jointly with clarified drawings of building plans and “Jointly table of radiation parameters” – are the initial data for Jointly Report preparing, and for subdivision of Summary Report by the results of radiation investigations of buildings and structures.

In addition to “Joint Table” and “Radiation investigation carts” the following are included into the Joint Report:

- 47. insights on the radiation state of technological systems;
- 48. insights on the radiation state of buildings taking into account for technological systems of all departments and workshops located in given compartment;
- 49. suggestions on ensuring the radiation safety (decontamination realization, equipment dismantling, sanitary barriers organization, additional ventilation development);
- 50. suggestions on changing of the building's zones, that take into account for presence radiation state and changes in technological process.

The participant stated that the methods of sample preparation, laboratory analysis and processing of results are approved by the accredited body for metrology and standardization of Ukraine (i.e., each laboratory has its own methods of laboratory control with reference to the instrument park).

The different techniques used for in-situ measurements, already described in the report No. 7305 of the Nuclear Energy Agency (NEA, 2016), are also qualified in a metrological laboratory by means of reference radioactive samples.

The corresponding program of quality assurance should ensure implementation of requirements to personnel, working procedures, technical means, control organization, corrective measures and data reporting.

The correlation between HTM to ETM radionuclides is performed by collecting samples for radiochemical analyses and applying the same approach as that described in Section 3.2.

According to the current Ukrainian Regulation, high-level radioactive wastes are that with a power generation more than 2 kVt/m<sup>3</sup>. These wastes may be disposed only in stable geological formations. Under treatment with high-level radioactive wastes is necessary the preliminary delay, which allows to reduce the power generation.

Under storage of high-level radioactive wastes it is necessary:

51. control of the hydrogen concentration in gas pillow;
52. blowing out of reservoirs of high level waste by air to remove hydrogen and reduce it's concentration;
53. control of gas pillow temperature (no more than 50-60 °C);
54. force cooling of reservoirs;
55. use of corrosion protected alloys and stainless steel in the evaporated facilities and reservoirs of high-level radioactive wastes;
56. location of reservoirs with concentrated high-level radioactive wastes below of the earth level on concrete pallets;
57. adding into the high-level radioactive wastes reagents which delay corrosion.

After the preliminary delay stage the immobilization of high-level radioactive wastes is performed into the refractory matrix (vitrification).

The major difficulties in defining a radionuclide vector are the following:

58. Several ways of radionuclides propagation with different accumulating characteristics.
59. Complicate source of radioactivity including the main source and additional ones (e.g. from the treatment line) especially of periodic operation that may cause a local distortion of vector.

Radionuclide vectors are made for every group of wastes having the same source (e.g., nuclear fuel, ventilation systems, coolant, etc.).

The participant remarked that there is not a clear correlation between chemo-toxic and radiological materials or substances. He furthermore assured that, concerning the radionuclides of concern, numerical simulations of their migration and accumulation are widely used on the basis of their characteristics (half-decay period, velocity of besieging, percent of transfer into steam, coefficients

of repeated wind rise). However, in his opinion, all the developed analytical model should be validated with realistic experimental data.

All over again, there are no available documents detailing how initial characterisation, conjointly with cartography, are used for:

- subsequent clearance process,
- waste classification and waste treatment process,
- planning decontamination actions,
- planning site remediation actions,
- performing the final site release process,
- estimating radiation doses to workers during the decommissioning activities,
- estimating the environmental impact during the decommissioning activities,
- estimating the amount of wastes produced during the decommissioning activities, and
- estimating of decommissioning costs.

However, the participant felt that initial characterisation allows to choose more effective technology and to estimate the time needed for carry out the needed procedures. Depending on the radioactive waste characteristics, selected treatment/decontamination methods and associated deadline make planning of means in order to ensure the radiation safety as well as the needed number of personnel to execute the required work without exceeding the corresponding irradiation limits. These means include:

- automation of technological process;
- using containers with the optimum biologic shield;
- any other protective actions (air cleaning, protective screens, distance protection, etc.).

Initial characterisation also allow to well analyse the dynamic of radioactive substances release into environment under normal conditions and in a case of accident. Analysis of the subsequent environmental impact is achieved in the framework of Safety Analysis Report for decommissioning, which should be approved by the state regulatory body as a necessary request to obtain the corresponding license.

Finally, the participant confirmed that operational characterisation is carried out during the dismantling process, but without providing any further precision about how it is used for estimating radiation doses to workers during the decommissioning activities.

## 5 General conclusions and recommendations

We have described here the whole steps that must be followed during the initial characterisation of a nuclear facility, subject to a decommissioning programme, for an accurate evaluation of its up-to-date physical, chemical and radiological properties. The document is based on a literature review of

several reference publications as well as on the information gathered by means of a questionnaire that has been submitted to several partners, experts and end-users from different EU member states, plus Japan and Ukraine, with a consolidated experience in the domain. We have equally explained the common sources of the radionuclides likely to be present in different nuclear facilities and how the associated scaling factors are derived. At the same time, the problematic of other hazardous substances exhibiting chemical toxicity have been briefly addressed.

In order to guarantee a successful accomplishment of the pre-decommissioning characterisation of a given nuclear facility the following recommendations must be duly adopted.

**- Regarding the characterisation objectives and the sampling process:**

1. It is necessary to define which radionuclides, non-radioactive elements and other properties will be measured and how they will be measured (NDA and/or DA measurements). It is also critical that representative samples are taken over the entire range of expected radioactive materials.
2. The sampling plan needs to detail the analytical procedures to be applied, including the necessary quality assurance (QA), quality control (QC) and quality management (QM) requirements. It should also ensure that the procedures meet all applicable levels of accuracy, reliability and precision needed to control and monitor the treatment or conditioning processes adequately. Without neglecting the time that samples will be gathered and analysed because it will have a direct impact on the throughput of such processes.
3. A minimum number of replicates have to be established in order to have sufficiently solid criterion for the statistical assessments of the proficiency test. Typically a minimum of three replicates per sample are required for destructive analysis (with similar amounts of mass or volume); two of which are used (mineralized or prepared for measurement) for the required analyses and the other one is kept as a reserve aliquot to cover any accidental event that may result in a loss of information.
4. Extrapolating information from in-situ measurements and samplings to the whole zone under study should be done very carefully, requiring a suitable strategy to be beforehand planned based on the study of the historical documentation of the nuclear facility and advanced statistical approaches.
5. Representative samples depend on the physico-chemical nature of the considered radioactive materials and on the chemical properties of the elements to isolate. For example, if a volatile material is to be determined, the sample treatment has to avoid exhausting the gases from the system.

**- Regarding in-situ measurements and laboratory analysis of samples:**

6. Both the in-situ measurement and radio-analytical methodologies, along with the uncertainty assessment and calibration procedures, must be previously tested by internal and/or external protocols.

7. The radioactivity distribution can be checked by measuring at different locations one or more easy-to-measure (ETM) radionuclides and all the collected samples must be as homogeneous as possible, if not, any heterogeneity should be corrected for a good comparison of the obtained results.
8. Development and use of reference samples, both simulating non-radioactive and radioactive materials, is of prime importance to allow comparing the quality of their analytical procedures using different techniques and instrumentation, to provide a means to improve accuracy and precision of methods and techniques, to facilitate direct comparison of analytical results, both intra- and inter-laboratory, when used as blind samples, and to allow for bias correcting of analytical results when processed alongside actual waste samples.
9. An inactive matrix with the same physico-chemical properties as the considered family of radioactive materials to be measured must be supplied to subtract the blank contribution for each measurement and to perform a good estimation of the minimum detectable activities.
10. It is preferable to perform inter-comparison exercises with real samples representative of radioactive contaminated concrete and/or soil. Although it is difficult to know *a priori* the "true values" of such samples, this kind of exercises will consolidate, harmonize and identify the actual limits the different methodologies applied.
11. It is moreover recommended performing a common benchmarking exercise for both in-situ measurements and destructive analysis. By considering for example a piece of concrete that can be measured in-situ before taking representative samples for laboratory analysis.

**- Regarding other practical considerations:**

12. It is preferable to conduct the whole characterisation steps in cooperation with the facility experienced staff, as these personnel use to have the necessary technical skills and are most familiar with the site history and particularities.
13. Although the historical information is a valuable asset, it should be continuously viewed with some scepticism and an intuitive sense of criticism needing to be confirm, whenever possible, by experimental data, numerical simulations or both.
14. Characterisation activities should not endanger safety and integrity of the item under study. For example, cutting of pipes for sampling purposes should not result in loss of containment or uncontrolled leakage of contaminated fluids.
15. Because of several unknown factors, such as uncertainties in confinement conditions and possible migration of radionuclides, there is a potential for exposure to high radiation doses, contamination levels or both during in-situ measurements or samplings. Caution must therefore be taken in order to ensure that all safety concerns are addressed. In any case, occupational dose uptake to both plant operators and laboratory personnel must always satisfy ALARA principles and the own radiation protection regulation of the facility.
16. The assigned code, exact location and date of each sample must be correctly recorded in a unified computerized database to be shared between the different stakeholders. The same

in the case of each in-situ measurement together with the information about identification of the instrument used, its calibration file and associated set-up configurations (supply voltage, signal parameters, measurement time, background, etc.). All the in-situ measurements and sampling results must be systematically transferred to the unified computerized database as well.

17. Periodical reviews of the above results can be used to rethink the characterisation plan or the sampling strategy (e.g., when radioactive contamination is more extensive than originally anticipated), to generate the most accurate testing simulants as well as to redefine the optimum sample treatment and conditioning processes.
18. Consistency checks are necessary between in-situ measurement or sampling locations and the expected contamination spaces. The team responsible of the characterisation activities should have enough flexibility to remediate promptly any newly identified radioactive contamination.
19. The obtained results of both in-situ measurements and laboratory analysis of samples must also be considered to review/reinforce the historical assessment and/or to validate the numerical simulations, namely the neutron activation ones, to increase the confidence of these latter for future decommissioning programmes.

The benchmarking exercise under realistic conditions, as the one proposed by the INSIDER project, will give the participants the possibility to evaluate the strengths and weaknesses of their in-situ measurement and/or radio-analytical methodologies. This will furthermore provide them a unique opportunity to share basic information and to gain a consolidated know-how in this domain, through a spirit of cooperation, learning, improvement and harmonization.

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## **Appendix 1**



**Improved Nuclear Site characterization for waste minimization  
in DD operations under constrained Environment**

Research and Innovation action  
NFRP-2016-2017-1

# **WP2 Questionnaire**

Final Version

## Information

Grant Agreement #: 755554

Project Title: Improved Nuclear Site characterization for waste minimization in DD operations under constrained Environment

Project Acronym: INSIDER

Project Start Date: 01 June 2017

Related work package: WP 2: Requirements and Validation

Lead Organisation: JRC

Dissemination Level: Confidential

## Questionnaire for the EUG on tasks T2.1 & T2.2

The present questionnaire has been designed to gather as much information as possible about the important decommissioning programmes of nuclear facilities that are currently being undergone in the EU states. It is aimed to give a good overview of end-users requirements and specific objectives for characterisation and cartography of contaminated sites and structures in constrained environment aiming at identifying key parameters for decommissioning operations orientation as well as scenario improvement and documentation.

First of all, please fill the following fields and also indicate the preferred transparency level your answers must have by clicking directly on the corresponding boxes below:

<b>You name and surname:</b>	
<b>Name of your organization:</b>	
<b>Your role in the organization</b>	
<b>Postal address:</b>	
<b>E-mail address:</b>	

- ☐ Fine to publish the whole questionnaire with name
- ☐ Fine to publish the whole questionnaire anonymously
- ☐ Just use the answers as part of a general overview / statistical analysis

Please specify below to which country the present questionnaire must be related?

Once filled, please return the present questionnaire before **August 31, 2017** to:

Paolo PEERANI ([paolo.peerani@ec.europa.eu](mailto:paolo.peerani@ec.europa.eu))

Danielle ROUDIL ([danielle.roudil@cea.fr](mailto:danielle.roudil@cea.fr))

Jeremy BUTTIN ([jeremy.buttin@edf.fr](mailto:jeremy.buttin@edf.fr))

Khalil AMGAROU ([Khalil.AMGAROU@cea.fr](mailto:Khalil.AMGAROU@cea.fr))

**T2.1: End users requirements for initial characterisations of contaminated sites and structures in constrained environment**

*In this task, partners will conduct a review on how characterization processes are implemented by different end-users, identifying the related (prevailing) regulatory requirements and depicting the major constraints found in the process*

1.1) Describe synthetically the process for a site/plant characterisation process in your country/organisation

1.2) Describe the regulatory framework for decommissioning and dismantling (D&D) activities in your country

- Applicable laws and regulations
- Did these laws preach for immediate dismantling or deferred dismantling? Please develop more about the followed strategy (waiting period, safe enclosure, entombment, etc.) in the case of deferred dismantling.
- Please specify, by clicking directly on the corresponding boxes below, the main target followed for D&D activities in your country.
  - ☐ Deconstruction, remediation and release of the site (green field)
  - ☐ Release of existing buildings and plants for other industrial uses
  - ☐ Further nuclear use
- Which authorities are involved?
- How is the licensing process?
- How long it takes to get an authorisation?
- How collaborative is the regulatory body?
- Please specify the existing infrastructure for radioactive waste disposal in your country and at your sites, their status (intermediate or definitive) and their capacity?

- 1.3) How are defined the goals for the characterisation?
- 1.4) What is your strategy about the soils?
- 1.5) How the sampling plan is determined?
- Sampling strategy
  - Balance between destructive and in-situ analyses
  - Cost/benefit analysis
- 1.6) Which are the key points in a characterisation plan?
- 1.7) What is the target level of uncertainties and how are they assessed?
- 1.8) Which are the major difficulties/obstacles/problems that you face in characterising a plant?
- 1.9) Do you use software (and/or mathematical approach) to operate the results of the measures?
- 1.10) Do you identify tools that you would wish to have in order to improve the characterisation?
- 1.11) Do you see benefits from a potential homologation on international standards (to be developed) in the field of site characterisation?



## T2.2: Specific objectives for characterisation and cartography

*This task will establish specific objectives for characterisation and cartography and identify key parameters for decommissioning operations orientation & scenario improvement and documentation*

2.1) How many nuclear power plants are currently undergoing a decommissioning programme in your country?

Reactor type	Number	Power range (MW)	Fuel	Coolant	Moderator
Pressurised water reactors					
Boiling water reactors					
Gas-cooled reactors					
Light water reactors					
Fast breeder reactors					
Heavy water reactors					
Accelerator based and sub-critical facility					
Research reactor					
Other ( <i>please specify</i> ):					

2.2) Please specify below, by providing as much details as possible, if there are other nuclear facilities originally planned for either spent fuel processing and recycling or radioactive waste conditioning that are currently undergoing a decommissioning programme in your country?

2.3) Please specify if there are Linac or Cyclotron installations originally planned for research or medical applications that are currently undergoing a decommissioning programme in your country? In such case, please, also give details about their primary beam current as well as their maximum energy and current.

2.4) Please give as much details as possible about the physical nature, composition estimated volume of radioactive waste and other contaminated materials (metals, liquids, plastics, concretes, soils, graphite, bitumen, etc.) commonly generated in the above decommissioning programmes.

2.5) Please give as much details as possible about specific difficulties and obstacles in the process of the decommissioning programmes.

2.6) Which are the main targets you aim to reach in:

- Radiological characterisation of an installation?
- Radiological (and/or chemical) characterisation of the soils?
- Radiological (and/or chemical) mapping of a (potentially) contaminated site?

2.7) Which are the radionuclides you look for?

2.8) Do you measure impurities (i.e. chemical compositions) to explain the level of the radionuclides measured? If “yes”, do you made sampling on irradiated or no irradiated item ? Why ?

2.9) Which protection level do you target, i.e. what activity levels and uncertainties are to be met for the key radionuclides you look for? How do you summarize remaining uncertainties and other than key nuclides?

2.10) Please specify and give as much details as possible (or provide the corresponding reference documents if any) about the measurement techniques do you apply in plant/site mapping?

2.11) Please specify and give as much details as possible (or provide the corresponding reference documents if any) about other *in-situ* measurement techniques used.

2.12) Please specify and give as much details as possible (or provide the corresponding reference documents if any) about the different laboratory analytical techniques commonly used.

2.13) Please explain (or provide the corresponding reference documents if any) the experimental procedures if any used to qualify each one of the different laboratory analytical techniques used.

2.14) Please explain (or provide the corresponding reference documents if any) the experimental procedures if any used to qualify each one of the different *in-situ* measurement techniques used.

2.15) Please explain (or provide the corresponding reference documents if any) the objectives and principle implementation of any quality assurance plan. What QA/QM guideline are to be followed.

2.16) In your opinion, what should be the specific objectives of a benchmarking exercise regarding laboratory analytical measurements?

2.17) In your opinion, what should be the specific objectives of a benchmarking exercise regarding *in-situ* measurements?

2.18) How do you correlate HTM (Hard-to-Measure) to ETM (Easy-to-Measure) nuclides?

2.19) How do you correlate HTM (Hard-to-Measure) to ETM (Easy-to-Measure) nuclides in the case of many HTM nuclides (i.e.  $^{60}\text{Co}$  +  $^{137}\text{Cs}$  for example)?

2.20) How do manage the case with no HTM nuclide (i.e. cooling time  $\gg$  10 half-lives)?

2.21) Which are the main difficulties you encounter in defining a nuclide vector for a facility?

2.22) Do you target a global nuclide vector for the entire facility or specific vector for individual subsystems?

2.23) How do you assess chemo-toxic material or substances? Do you correlate radio-toxicity with chemo-toxicity? Do you request material classes and chemo-toxic compound vectors? What are the main difficulties with chemo-toxic and radio-toxic standardised vectors? How do you deal with uncertainties?

2.24) Do you complement your measurement data with modelling/calculations? How?

- 2.25) How preliminary characterisation is used for subsequent clearance process?
- 2.26) How preliminary characterisation is used for subsequent classification of waste and waste treatment process?
- 2.27) How preliminary characterisation is used for planning decontamination actions?
- 2.28) How cartography is used for planning site remediation actions?
- 2.29) How initial cartography is used for the final site release process?
- 2.30) How preliminary characterisation is used to estimate radiation doses to workers during the decommissioning activities?
- 2.31) How preliminary characterisation is used to estimate environmental impact during the decommissioning activities?
- 2.32) How preliminary characterisation is used for estimation of amount of waste produced?
- 2.33) How preliminary characterisation is used for estimation of decommissioning cost?
- 2.34) Do you apply operational characterisation during the dismantling process? If yes, how it is used to estimate radiation doses to workers during the decommissioning activities?

