Final radiological release of the Radiochemistry Hot Laboratory at the Joint Research Centre in Ispra

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Abstract

The Joint Research Centre of Ispra is one of the research Sites belonging to the European Commission, Directorate General JRC. It was created in the late '50s, in order to steer European research on nuclear industry. It currently hosts numerous nuclear facilities, some are still in operation, while the remainder are in a position of safe shutdown or in the process of decommissioning.

One of the first JRC-ISPRA's radiological facilities to be released from regulatory controls, in 2010, was the Radiochemistry Hot Laboratory (RCHL). This facility has been used, since the sixties, to perform radiochemistry separation and analyses on many radioactive samples obtained through irradiation in other JRC-ISPRA's nuclear facilities, including the two research reactors ISPRA-1 and ESSOR.

Radioisotopes found in RCHL during its decommissioning process were alpha, beta and gamma emitters, including the majority of the most interesting hard-to-measure radionuclides.

An extensive radiological characterisation campaign, composed of a first phase of (mostly) non-destructive analyses and a second phase, of more detailed both destructive and non-destructive analyses, was performed in RCHL between 2008 and 2009.
Homogeneous groups and nuclide vectors were then properly defined, according to the results of the characterisation campaigns.

A final decontamination campaign was performed before the facility's final radiological release.

A thorough final radiological survey was performed after RCHL final decontamination, examining hundreds of samples and direct analyses, with the aim of a thorough description of the facility's final radiological status.

At the end of this process, the RCHL was released from regulatory control, and was destined to non-radiological activities.

This report covers the pre-characterisation, characterisation and final radiological survey processes, which lead to the final radiological release of the facility.

Keywords: building release, characterisation, non-destructive analyses, final survey, nuclide vectors

1. Introduction

The Radio-Chemistry Hot Laboratory (RCHL) was constructed in the early 1960s at the Joint Research Centre of Ispra (JRC-ISPRA). The Laboratory was composed of many separate buildings, the main one (Building 46) built over three levels, was completed in 1966. In 1976, an annex to building 46 was built (Building 46d) and in 1978 a new area of laboratories, buildings 32b-32c – "*Stabularium*" was completed. Other adjacent buildings were used to perform research activities with several radionuclides, including heavy metals and short lived α and β-γ emitters.

The Radiochemistry Hot Laboratories covered an integrated surface of 2.652 m^2, operated under an Authorization from the Italian Ministry of Industry, and globally, hosted 50 scientific personnel, working in the following research fields:

1. environmental and biochemistry research using radioactive tracers;
2. neutron activation analyses;
3. extraction of actinides from radioactive liquid waste coming from nuclear fuel reprocessing plants;
4. analyses of U, Pu and Th in samples from the nuclear fuel cycle, in order to determine isotopic ratio and burn-up.

All research activities in the laboratory ended before 2005, and some preliminary decommissioning activities started, such as the removal and transport (to temporary storage) of radioactive waste, radioactive sources, other radioactive substances and compounds present in the facility, including frozen radioactively traced laboratory animals. Drain lines for liquids from the controlled areas were sectioned off from the corresponding collection tanks, sealed and removed; collection tanks were drained, original laboratory equipment was removed from the building and extraction lines for therein located glove-boxes were cut off and dismantled; accessible drainage piping was dismantled as well.

Finally, a preliminary radiological characterization campaign was performed, and some basic corrective measures (e.g. decontamination and removal of remaining "hot spots") were achieved and subsequently, the final radiological survey started.

This report covers the pre-characterisation, characterisation, and final radiological survey processes, which finally lead to the radiological release of the facility with no radiological constraint.

2. Materials and methods

Many different actors took part in the preparatory phases of the facility's decommissioning: all work activities were performed by external Companies under the coordination of a JRC-ISPRA Project Leader and the support of the JRC-ISPRA Radiation Protection Sector.

From 2005 to 2007, some preliminary measurement campaigns were performed, in order to ascertain the radiological status of the facility: they were organised by the JRC-ISPRA's Radiation Protection Sector, mostly with the use of operational radiometric equipment (contamination and dose rate meters) and with some basic laboratory radiometric instruments (fixed contamination monitors and spectrometers, used to assess removable contamination via smear tests). These preliminary results were the basis for planning the following steps, namely the radiological characterization of the facility and the subsequent decommissioning strategy.

In 2007, the Italian company *Ansaldo Nucleare* published a first report ("**RCHL: Historical Site Assessment Report**") stating a general description of the facility including historical and up-to-date radiological data.

The characterisation campaign continued in 2008, when four studies, namely "**Executive Design Reports**" and "**Work Plans**", identifying the DA and NDA measurements for the radiological characterization, were finalised by the Spanish Company *Iberinco* and the consortium between the Czech Company *Envinet* and the British Company *VT Nuclear Services*. These reports were mainly based on previously obtained dose rate and activity data, and indicated the required measurements, procedures and methods to be employed to complete the characterisation plan.

Further on, in 2009, the First & Second Intermediate Reports were published together with the "**Plant Characterization Report**", prepared by the Spanish Company *Iberdrola* and in 2010, the Italian Company *Sogin* delivered the last characterization plan for the final release of the facility ("***Piano di caratterizzazione radiologica per il rilascio finale di RCHL***") taking into account all released documentation and data.

This huge documentary body and the activities that had already been achieved, gave a solid basic structure for the successful accomplishment of the Final Survey. In particular, the Final Survey was carried out in the second semester 2010, with the joint effort of the Radiation Protection Sector, the Laboratory for Radioactivity Measurements (LMR) Sector and the Decommissioning Sector of the Nuclear Decommissioning Unit of JRC-ISPRA. The Final Radiological Survey had, as its main scope, an accurate verification of the radiological condition after decontamination and dismantling, proposal of additional implementation measures, if necessary, and constituted the technical basis for the unconditional release of the laboratories.

Pre-characterisation and characterization: DA and NDA methodology and instrumentation

NDA and DA measurements were carried out from 2007 to 2010: all physical and radiological data obtained were incorporated into the database *Misure Radiologiche Ispra* (MiRadIs), containing all physical and radiological characterization results from all JRC-ISPRA Site facilities, including RCHL, for which, the initial programmed surveys were: physical and hazardous materials on site inspection and radiometric surveys.

The radiometric survey was based on the MARSSIM methodology [3] and other European and International recommendations ([4], [5] and [6]). It included direct measurements of gamma dose rate to identify the radiation field and total alpha, beta and gamma surveys to assess the existence of surface contamination, if any.

Non-Destructive Analyses (NDA) were performed according to the "Executive Design Reports" and the corresponding "Executive Work Plans", following NDA Contractors standard operating procedures and JRC-ISPRA specific indications.

All NDA and DA measurements were performed with off-the-shelf instrumentation, following specific quality procedures and JRC-ISPRA indications and Radiation Protection instructions.

Final Survey: DA and NDA methodology and instrumentation

The radiological Final Survey (FS) has a two-fold objective: an independent JRC-ISPRA verification of data collected from the contractors' reports; and the final release, without radiological constraints, of the entire RCHL facility. The Nuclear Decommissioning Unit gave the responsibility of this task to both the Radiation Protection Sector (RP) and the Laboratory for Measurements of Radioactivity (LMR), with the operational collaboration of the Decommissioning Sector, for final dismantling, clean-up and waste management activities.

Clearance and release levels

General material clearance and building release levels do not currently exist in Italy, and are therefore indicated by the Nuclear Authorities, case by case, in the decommissioning license of a specific facility.

JRC-ISPRA radioactive waste treatment and storage facility (*Stazione Gestione Rifiuti Radioattivi*) is operated on the basis of an authorisation [8] including a table for clearance of solid materials (**Table 1**) therein transferred from the JRC-ISPRA nuclear facilities.

This table has been used for very low level radioactive materials aimed to be recycled, reused or disposed of, and for dubious materials dismantled in RCHL.

RN (i)	***Metal material (c_{li})***		***Building rubble (c_{li})***		***Other materials (c_{li})***
Symbol	***Mass ($Bq \cdot g^{-1}$)***	***Surface ($Bq \cdot cm^{-2}$)***	***Mass ($Bq \cdot g^{-1}$)***	***Surface ($Bq \cdot cm^{-2}$)***	***Mass ($Bq \cdot g^{-1}$)***
^{3}H	1	10000	1	10000	1
^{14}C	1	1000	1	1000	1
^{22}Na	1	1	0.1	10	0.1
^{36}Cl	1	100	1	100	1
^{54}Mn	1	10	0.1	1	0.1
^{55}Fe	1	1000	1	10000	1
^{59}Ni	1	1	1	10000	1
^{60}Co	1	1	0.1	1	0.1
^{63}Ni	1	1000	1	10000	1
^{90}Sr	1	1	1	100	1
^{125}Sb	1	10	1	1	1
^{134}Cs	0.1	1	0.1	1	0.1
^{137}Cs	1	10	1	1	1
^{152}Eu	1	1	0.1	1	0.1
^{154}Eu	1	1	0.1	1	0.1
^{235}U	1	1	1	1	1
^{238}U	1	1	1	1	1
α- emitters	0.1	0.1	0.1	0.1	0.01
^{241}Pu	1	1	1	10	1

Table 1. JRC-ISPRA radioactive waste treatment and storage facility clearance levels for materials.

No general building release levels being available, it was necessary to adopt and apply shared international standards ([3], [4], [5], [6]).

Implementation of MARSSIM strategy at RCHL

According to the MARSSIM methodology [3], and using collected information from previous reports and the history of the facility, RCHL (**Figure 1**) was divided into four homogeneous groups (HG):

1. HG I: Bld. 46
2. HG II: Bld. 46d
3. HG III: Bld.s 32b & 32c
4. HG IV: Bld.s 46f, 46g, 46h & 32e

Each room of the HG represented a survey unit (SU) and each one of them was subdivided into three different classes of measurements to be performed:

- *Class 1*: areas potentially contaminated with values above the release levels;
- *Class 2*: areas where radioactive residual contamination measured is below the release levels;
- *Class 3*: areas where there is no residual radioactivity or, at most, the amount is equal to some fraction of the release levels.

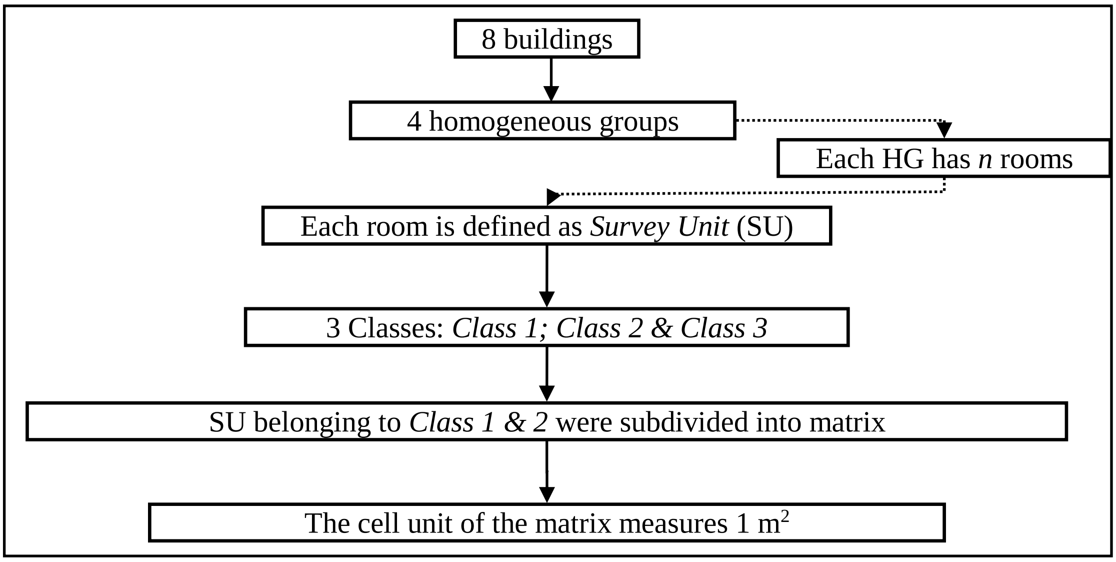

Figure 1: Structure of programmed measurements

Non Destructive Analyses (NDA)

A complete manual scan of surfaces in RCHL, according to the measurements plan, was performed by the Radiation Protection Sector, using the Berthold LB124 SCINT contamination meter (**Figure 2**). This instrument is battery-powered, comprised of a display unit, a signal processing electronics and a ZnS- scintillator with photomultiplier, with an active measurement area of 170 cm^2, and is suitable for measuring alpha and beta-gamma contamination.

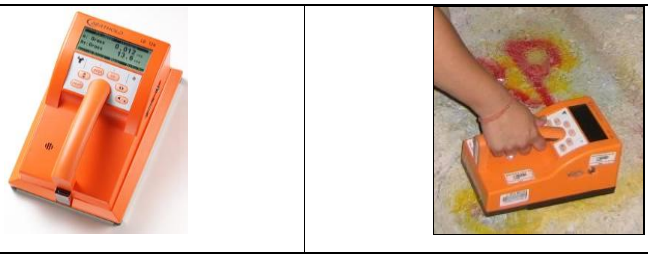

Figure 2: Berthold LB124 SCINT instrument (left) and operating on-site (right)

Other NDA analyses were performed by the LMR, using two ISOCS (IGSS Canberra), self-calibrating high resolution gamma-spectrometry assay systems, designed for performing non destructive radiological assessments by near real-time on-site measurements (**Figure 3**):

Figure 3: The ISOCS system with all its available components (left) and operating on-site (right)

<u>Destructive Analyses (DA)</u>

Furthermore, as for destructive analyses (DA), thirty samples (dubious and "hot spots") were sent off-site to *ENVINET Inc. - WERT Ltd.* laboratories for α, β, γ measurements, for results intercomparison with JRC-ISPRA's measurements.

In detail, the *ENVINET Inc. - WERT Ltd.* laboratories were in charge of the DA of different matrixes samples (**Figure 4**) measurements, including hard-to-measure radionuclides (such as ^{238}Pu, ^{241}Pu, ^{235}U, ^{238}U) and also gamma emitters' nuclides (i.e. ^{137}Cs, ^{60}Co, ^{133}Ba, ^{241}Am, ^{152}Eu, ^{154}Eu), with liquid scintillation, alpha spectrometry, gamma spectrometry and low level alpha/beta counting techniques.

Figure 4. Sampling for DA in different matrixes

Measurements' work plan and methodology

Once the division of the RCHL complex was defined through HGs and SUs, a work plan consisting of seven phases was devised.

1. Preparation of the matrixes in each SU

Radiation Protection technicians prepared 7.363 matrix unit drawing areas of 1 m^2: they were differently marked, depending on the surface (paint spray for concrete, chalk for sand and adhesive tape for linoleum floors or surfaces) as shown in **Figure 5**.

Figure 5: Different marked matrixes

2. Manual scanning of surfaces, identification of "hot spots" and nuclide; verification of activity

The manual scanning of floors' and walls' surfaces was performed with the Berthold LB124 SCINT by Radiation Protection Technicians (**Figure 6**). The floor was covered by a 1 m^2 matrix which extended on the walls up to 2 meters high for *Class 1 areas* (100% of the surface) and for *Class 2 areas* (70% of the surface).

Measurement procedure was:

1. Background measurement in the SU to be scanned;
2. Scanning of the 1 m^2 matrixes in an average timing of six minutes and further marking the measuring point highest value (measured again for additional 60 seconds);
3. Locating and identifying any result above reference surface contamination values (indicated as "hot spots").

Figure 6: RP technicians scanning matrixes

LMR was responsible for the detection of any average residual radioactivity in all the selected rooms and "hot spots'" activity characterization, *via* near surface measurements (**Figure 7**) using IGSS detector appropriately collimated at a distance of 30 cm from the measuring point, for *Class 1&2 areas* (all "hot spots" have been measured before and after contamination removal).

Figure 7: Near surface measurement with in situ gamma spectrometry

In addition, the two IGSS were used, with the detector collimated for large surfaces measurements (**Figure 8**), in *Class 3 areas* taking one measurement point every 20 m^2 of ceiling, walls and, where applicable, floors.

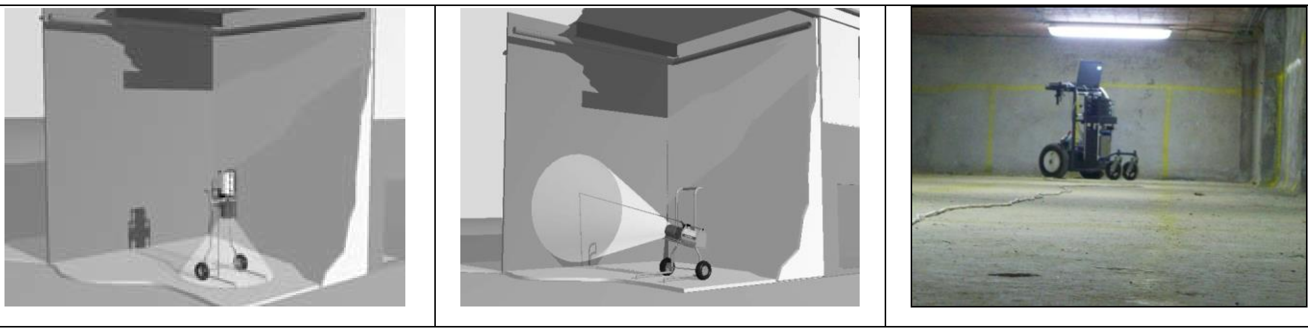

Figure 8: The ISOCS system properly collimated for wide areas surface measuring
(CANBERRA ISOCS documentation and RCHL on-site measurement)

Parameters for IGSS measurements were:

1. *Background:* surface/near surface mode of 60.000 s, 10.800 s and 5.400 s, with the detector properly oriented and collimated to the blank measuring point;
2. *Library:* including all gamma emitting radionuclides from the Italian Safety Authority clearance levels list;
3. *Geometry:* for surface measurements, a circular plane geometry with lead collimators of 25 mm/90° for 10 800 s and a rectangular plane geometry with lead collimators of 25 mm/180° to ensure better accuracy on larger surfaces, in order to adequately monitor ceilings, walls and floors; for near surface measurements, a circular plane type and lead collimators of 25 mm/90° to minimize interfering radiation and limit the field of view;
4. *Time:* surface measurements were programmed for 10.800 s and 55.000 s overnight; all near surface measurements were programmed for 10.800 s;
5. *Measurement accuracy:* all near surface measurements were performed at 30 cm distance with three different depth models according to the kind of surface to be measured (concrete, sand/earth or other specific surface characteristics) from 10 μm to 1 cm.

All NDA measurements have been achieved with a high level of accuracy and traceability following the quality standard procedures in force at JRC-ISPRA.

3. Identification of the DA sampling points and assistance during sampling

Once a "hot spot" had been localised and marked by the Radiation Protection Technician (**Figure 10**), a gamma spectrometry measurement with the IGSS was performed (**Figure 9**) before taking the DA sample that was immediately prepared and sent to the LMR and the external Laboratories.

Figure 9: The ISOCS system properly collimated for "hot spot" characterisation

The DA sampling points were chosen following these criteria:

a. If manual scanning measurements resulted in a "hot spot" identification, or;
b. If only α–emitting radioisotopes were detected, making it impossible to assign a SF and obtain RNs, or;
c. If, on the contrary, it was possible to identify the SF but the α–emitting findings were considered not reliable, using existing historical data of the SU, or;
d. If the room access made difficult either surface scanning or gamma spectrometry via IGSS, or;
e. For all buildings belonging to HG IV.

Figure 10: Identification and marking of "hot spots"

The DA measurements results have been certified by ISO international standards, with expanded standard uncertainties with a coverage factor *k=2*, corresponding to a normal distribution to a level of confidence of approximately 95% and the detection limits have been calculated according to ISO-11929. Moreover, five DA measurement results from the external Laboratories were also measured by LMR laboratory at JRC-ISPRA, which has nine accredited test methods ISO/IEC 17025:2005, awarded by the Italian body ACCREDIA.

4. Assistance to the surfaces final cleaning and verification of residual activity.

All the NDA and DA results were carefully analysed and implementation measures taken. In all cases, where contamination had been detected, it had to be removed (**Figure 10**).

Figure 10. Contamination removal on a laboratory wall and floor

Of course, after contamination removal, additional IGSS NDA measurements were performed, in order to verify that no residual activity remained (**Figure 11**).

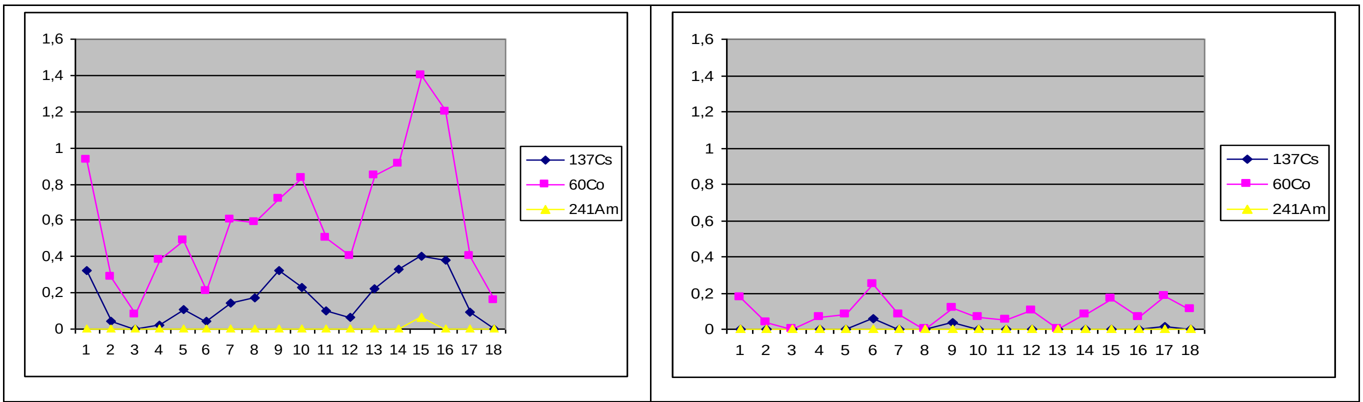

Figure 11: Gamma spectrometry after contamination removal on a wide area (floor) – Bq/cm^2

5. Verification of Clearance levels compliancy

Specific *scaling factors* [7] were identified from the abovementioned activities' results, and for each HG so as to trace hard-to-measure radionuclides concentrations once the gamma emitting nuclides had been confirmed (with IGSS). Subsequently, a list of *nuclide vectors* (NVs) was defined. The computation of NVs represented the very first difficulty because the preliminary characterisation didn't provide a sufficient amount of data for an accurate computation. Within a given HG, the same RN could be found with different orders of magnitude and with a non-homogeneous distribution of RN in each HG: it was therefore decided to choose the most conservative option.

The application of the clearance criteria proposed a series of different steps depending on the chosen type of analysis. For *in situ* measurement results (i.e. gamma spectrometry), the following process was used:

- collection of RN concentrations obtained for every SU of each HG;
- application, where possible, of the scaling factors for the determination of HTM radionuclides;
- verification, with the use of spreadsheets, of the possibility of the actual release.

Regarding DA samples analysis, direct verification with the clearance levels was done:

- directly, through surface activity concentrations (i.e. Bq cm^{-2}) for most of the RNs;

- indirectly, through the mass activity concentrations (i.e. Bq g^{-1}), given the density of the analysed material.

It should be noted that the levels shown in **Table 1** have been applied to the total activity expressed in $Bq \cdot g^{-1}$ (*i.e.* total activity divided by the analysed sample mass) or in $Bq \cdot cm^{-2}$ (*i.e.* total fixed or removable activity divided by the analysed sample surface). In the latter case, the area from which the mean value had been extrapolated was not greater than 1 m^2, the criterion that was always observed in the case of manual scanning measurements. Usually, total activity was the contribution of a group of RNs and not a single one: therefore, the following condition had to be verified, in order to ensure that activity generated by a mixture of RNs was below the clearance levels:

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

where: c_i is the total surface activity ($Bq \cdot cm^{-2}$) of RN *i*-th,

c_{li} is the clearance level for the RN *i*-th ($Bq \cdot cm^{-2}$),

n is the number of RN composing the mixture.

In this expression, if the sum of the concentration ratios for each RN with the corresponding levels of clearance is less than 1, then the material meets all the requirements for clearance.
Finally, activities resulting from natural occurring RNs have been ignored.

6. Decommissioning and waste management activities

In Figure 12, some examples of dismantling activities in the Laboratory are shown, including scarification of concrete floors and walls.

Figure 12. Before and after dismantling contaminated and cold components and materials

In Figure 13, some examples of management of radioactive wastes resulting from the activities undertaken in the Laboratory are shown, including packaging and transport of liquid tanks.

Figure 13. Waste packaging and management

3. Discussion

The Radiation Protection Sector performed manual NDAs for a total of 5.494 matrix units in 94 rooms of the RCHL complex. The LMR carried out a total of 523 NDAs measurements, of 8.962 m^2, using two IGSS in 111 rooms, namely 448 measurements in surface mode, 28 Class 1 and 2 "hot spots" in near surface mode before contamination removal and 47 Class 1 and 2 "hot spots" in near surface mode after contamination removal.
DA samples were processed from a variety of matrix types, taken from different SUs, depending on the retrieved historical and radiological data. In particular, 26 concrete and 4 sand samples were measured at external Laboratories; 15 concrete samples analysed in parallel at LMR for results intercomparison with external Laboratories; and 11 concrete, 8 sand and 3 samples from surrounding trees at LMR.

Even though a pre- characterisation campaign had been successfully completed in 2010, during the final survey, results obtained both from NDA and DA still indicated the presence of slightly contaminated surfaces and materials: hence, some corrective actions had to be implemented.

The original cost for the release of the facility was estimated in the Project Plan at 2 383 600 €$_{2010}$, and the final cost amounted to 2 610 805 €$_{2010}$, which is within the 10% committed contingencies.

Costs are split as shown in **Table 2**.

Activities	**[€$_{2010}$]**	**[%]**
Underground Pneumatic Transfer Systems	277 926	11
Decommissioning (Dismantling-Radioprotection-Transports)	917 845	35
Plant characterization	532 000	20
Final Survey	314 455	12
Basic design and licensing	463 941	18
Supply	104 638	4
TOTAL	**2 610 805**	**100**

Table 2. Costs in RCHL release.

About 75 t of low activity waste was produced and disposed of in 220 L drums and 0,6 m^3 containers. About 220 t of clearable material was produced and deposited in dedicated storage areas.

Some technical difficulties were found during the final survey campaign with building structural limitations giving difficult access to some rooms, considering that IGSS weights 100 kg and is positioned on a wheeled cart (stairs, differently levelled rooms, sloping floors, irregular soil, sandy or gravelled pavement, etc. were present). Also, in potentially contaminated soil or areas, the instruments had to be appropriately protected (covering the wheels, avoiding spread of contamination). Additionally, presence of contamination in a single spot, that had permeated the floor, reaching a depth of almost 1 m required scarification, limited excavation and new DAs to ensure complete decontamination. Finally, the presence of unexpected contamination spread on sandy floors and low ceiling rooms in the basement required vacuum removal and new DAs were necessary to ensure complete decontamination.

4. Conclusions

The final release of the RCHL Laboratory has been the first building release experience at the JRC-ISPRA.
All the measurements and instrumentation were chosen in order to guarantee that the survey results will eventually reach the degree of accuracy needed for the facility final release. This process needed a significant amount of time and resources, and finally resulted in the expected unconditional release of the facility.

Final release costs (including operational decontamination activities and waste management) were in line with the forecasted budget.

Usually, in most nuclear facilities, decommissioning activities produce large amounts of waste, but in the RCHL release process (which did not include demolition of the building itself!) the on-site decontamination activities granted a significant reduction in the amount of final waste initially estimated (**Table 3**) and a significant increase in the volumes of clearable materials.

	Amount estimated in Project Plan	*Actual Waste produced*
Waste [kg]	101 600	74 615
Clearable material [kg]	153 100	218 300

Table 3. Waste production prevision and final amount generated.

Finally, from the radiation protection point of view, it is important to highlight that, considering time spent during the 5 years of this intense work activity, integrated doses to the staff were negligible.

5. Acknowledgments

The Nuclear Decommissioning Unit wishes to acknowledge the excellent work of many professionals who contributed to the final release of the RCHL facility, namely Marek FUTAS, Francesco ROSSI, Elisa PERSICO and Andrea RAVAZZANI.

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- Nucleco for the *in situ* DA and NDA measurements;
- Envinet for the off-site certified DA measurements;
- Salvarem for sampling materials and components and partial facility dismantling.

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