# Department of Technical Cooperation End-of-Mission Report

Report Title:	Methods for Localization of Radioactive Sources in a Large
•	Concrete Structure embedded into the entombment structure of
	the reactor compartments at Paldiski site
Project Number:	RER9146, Enhancing Capacities in Member States for the
	Planning and Implementation of Decommissioning Projects
Project Title:	Enhancing Capacities in Member States for the Planning and
	Implementation of Decommissioning Projects
Name of Expert:	Jaap Velthuis, Steven Slater, Michael Ojovan, Zoran Drace
Dates of Mission:	27-30 January, 2020, Tallinn Estonia
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#### Terms of reference:

Describe the specific objectives of the assignment and the duties to be performed by the expert as they relate to the objectives.

The purpose of the expert mission is to discuss/present suitable non-destructive methods for localization of radioactive sources embedded into the entombment structure of the reactor compartments at Paldiski site and to provide recommendations on the most suitable ones.

Provide recommendation on suitable method or complex of methods to be deployed and assist counterparts in drafting a job description for procurement documents.

#### **Background:**

Presentations by the host utility provided orientation of equipment, configuration and radiological characteristics of reactor stands 346a (Unit-1) and 346b (Unit-2) comprising the following:

Approximately 25 tonnes of radioactive waste was placed in the submarine reactor compartments (15 tonnes Unit-1 and 10 tonnes Unit-2<sup>1</sup>) prior to entombment with 72 m<sup>3</sup> of concrete during 1995. Lists of these wastes were compiled in September 1995 and given to the Estonian authorities when transferring ownership of the site from the Russian Federation to Estonia.

<sup>&</sup>lt;sup>1</sup> Task 2 Interim report collection of data and overview of national and international requirements, Pg57

#### Unit-1 (346A)

Has 15 tonnes of low-level radioactive wastes comprising; rags, metallic wastes, tools etc, with surface contamination. The wastes are located principally on the first and second floor of the 'non-pass through premises of the reactor compartment'. In addition to these wastes 100 radioactive sources (used for calibrating radiological measurement equipment) were also entombed in concrete poured into Unit-1. These sources are reportedly held in five containers within the concrete matrix. At the present the host utility is unable to accurately determine the exact location of these sources which comprise:

- neutron sources: Pu-238, Be-7, Cf-252
- γ-radiation sources: Co-60
- β-radiation sources: Na-22, Cl-36, Sr-90/Y-90, Cs-137, Tl-204
- α-radiation sources: Pu-239

Plutonium and caesium sources ranged from a few kBq to a few MBq. The total activity of the radioactive sources that were on site and might have been placed into Unit-1 was about 4.4 TBq in 1995 (mainly Co-60). All these sources are located inside shielding containers (list of disused sealed radioactive sources (DSRS) is provided in Tables 2-4, Attachment 1). For neutron sources and some  $\gamma$ -radiation sources, the container is constructed of special paraffin and/or lead. For  $\beta$ -radiation and  $\alpha$ -radiation sources, the container is plastic or wood.

It is documented that the sources were placed into the U-shaped first-floor room and or the reactor vessel lid before these spaces were grouted with concrete. A total of 30.75m<sup>3</sup> of concrete was used to isolate this material (mass 67,650Kg).

#### Unit-2 (346B)

It is understood Unit-2 has 10 tonnes of low-level radioactive wastes comprising tools and loading equipment entombed in 41.25M<sup>3</sup> of concrete with a mass of 90,700Kg. No radioactive sources are reported as present in this Unit.

## Duties performed by the expert:

Describe the work carried out to meet the terms of reference as set out above. Please include any technical, logistical, administrative and other problems encountered, and any other considerations of importance. Please include also the Agenda and List of persons met.

NOTE: Figures, tables and annexes should be mentioned in the body of the text and should be numbered in the order in which reference is made to them (e.g. Fig.1, Fig. 2, Table 1, Table 2, Annex 1, Annex 2, etc.). All attachments should be clearly labeled.

## Day 1

Mission started with a presentation by the Host representative Mr. M. Varvas about "Paldiski Nuclear Training Center- Historical overview" (Attachment 2). This presentation provided information on the work done during Decommissioning Plan Phase 1 comprising:

- Update and renovation of infrastructure, to prevent spread of contamination recognising the changing situation and environment.
- Establishment of waste management system and interim storage for waste, in accordance with current safety criteria.
- Reduction of controlled areas by demolition and decontamination of unnecessary buildings.

The next presentation provided by the Host representative Mr. I. Tatrik and focussed on new development in A.L.A.R.A., Estonia (Attachment 3) stating that:

- From 09.2014 to 12.2015 the project 'Preliminary studies for the decommissioning of the reactor compartments of the former Paldiski military nuclear site and for the establishment of a radioactive waste repository' was implemented.
- The main contractor was UAB 'Eksortus' from Lithuania and subcontractors Federal Centre of Nuclear and Radiation Safety and 'Atomproject' from Russia.

Outcomes of project were:

- Selected decommissioning option
- Conceptual understanding what kind of repositories are needed in Estonia taking into account accumulated waste, arising waste from decommissioning of reactor compartments and future institutional waste.

IAEA Experts have been provided with the final report "PRELIMINARY STUDIES FOR THE DECOMMISSIONING OF THE REACTOR COMPARTMENTS OF THE FORMER PALDISKI MILITARY NUCLEAR SITE AND FOR THE ESTABLISHMENT OF A RADIOACTIVE WASTE REPOSITORY" (Attachment 4), and Task 2 Interim report – Collection of data and overview of national and international requirements (Attachment 5).

A short video presentation was provided on a concept of decommissioning of the reactor compartments and placement of decommissioned wastes in a near surface repository (low level wastes) or a silo style repository (intermediate level wastes) or free release of materials. It was

stated that start for completion of decommissioning of two the reactor compartment is circa 2040.



#### Figure 1, Unit-1

Figure 2, Unit-2

Figures 1 and 2 illustrate the orientation of the reactors and their auxiliary equipment. The reactors are located in the same building with approximately 15 metres separating the two-reactor compartment. Both units have their own sarcophaguses.

It was stated also that the environmental impact assessment (EIA) following studies must be conducted between 2019-2023 as follows:

- Engineering survey of the reactor's sarcophagus, reactor compartment structures, main building, waste storage facility;
- Radiological survey of the:
  a) reactor compartments without entering RC;
  b) main building;
  c) Paldiski site environment;
- The establishment of the equipment for "show through" concrete. Identification of radioactive waste and sources inside the concrete mass.

The afternoon was devoted to the tour of the two reactor compartments of Paldiski facility followed by in depth discussions on possible location of DSRS's and techniques that might be considered for their location.

#### Day 2

The morning was devoted to the discussion with Mr Valery Badyrkhanov, employee of A.L.A.R.A. who was eyewitness during grouting of two reactor compartments performed in 1994 by personnel of ex-Soviet Navy former operator of the Paldiski Military Training Centre.

Mr Valery Badyrkhanov provided the following background information

Estonia has a former Soviet Union military submarine training centre at Paldiski site with two reactor compartments (Unit-1 and Unit-2). The nuclear fuel was removed from reactors and reactors were prepared for safe storage. Among other safety improvement actions grout was poured inside reactor compartments which fixed low level radioactive materials cut from different parts of reactor, supporting facilities and some radioactive waste materials into the body of concrete. DSRS in shielding containers and boxes from the site laboratory were also fixed in the body of concrete. Via analysis of documents available and discussions held with Mr Valery Badyrkhanov it was established the most likely location of the sources was in a cement matrix on in the top of unit 1 - reactor closure plate (red box below - validated through eye witness accounts) entombed in storage boxes (5 off) - see Fig. 3. There is however some confusion in the written text of report "Collection and Analysis of Information Regarding the Design and Content of the Reactor Compartments of Russian Nuclear Submarines that are being stored in Estonia", Technicatome, 09.10.2001 which stated that "Most sources were placed into the U-shaped firstfloor room where the main equipment of the first loop is located, and in the second floor area containing the motors and pumps, before these spaces were grouted with concrete. However, some sources could also have been placed in concrete poured onto the reactor vessel lid". The eye witness account did not support this written text. The eye witness account also confirmed the placement of waste materials into Unit-2.

It was confirmed through discussion the Unit-1 reactor closure plate was sealed with a high integrity weld.



Figure 3, Unit-1 containing DSRS's

During the day experts provided following presentations:

- 1. An introduction to Steve Slater and Sellafield Ltd (Attachment 6)
- 2. A Case Study Characterisation Why so Important ? (Attachment 7)

These presentations and additional movies shown contained examples of characterisation where there is poor quality information; Characterisation examples using (a) Drones, (b) Laser and 3D characterisation; Importance of physical characterisation; Importance of 'cleansing data' to that which can be trusted.

The presentation of methods and technique of muon tomography has been provided (Attachment 8).

The rest of the day was devoted to drafting report for discussion during third day of the EM.

## Day 3

EM Report has been drafted and discussions were held with the Host. Photos taken during EM are presented in the Attachment 9.

DSRS's in containers are present within Unit-1 is of concern to the A.L.A.R.A. and reportedly their regulators. There is a concern the decommissioning will compromise the source container or the source itself resulting in:

- 1. Contamination.
- 2. Unnecessary radiation exposure.
- 3. Waste that may not be acceptable to the final disposal site.

During preliminary studies related to decommissioning project it was requested to identify techniques to identify the location of the sources with the help of some non-destructive methods or complex of methods in the reactor compartments.

In finalization of the EMR experts used variety of IAEA reference material to discuss techniques to locate embedded sources into the entombment structure of the reactor compartments at Paldiski site, such as shown in Fig. 4:



Figure 4. Characterisation methods used to localise sources of radiation.

Different imaging techniques were discussed since these are used to identify any variation in the distribution of density and/or attenuation coefficients in waste, which may indicate the presence

of sealed radioactive sources. Imaging techniques can be categorized according to the type of external radiation source (except autoradiography) used for the creation of the images (e.g. X rays, gamma rays and neutrons), measuring technique (e.g. film/converter, digital systems, gamma detectors) and processing of data (e.g. radiography, tomography), as presented on the Figure 16 from IAEA document on "Locating and Characterization of DSRS".



FIG. 16. Schematic description of commonly used imaging techniques. The radionuclides in the figure refer to the external sources.

As seen from this figure, many alternative characterization techniques exist. One can also note acoustic methods such defectoscopy ultrasonic testing as or (https://en.wikipedia.org/wiki/Ultrasonic\_testing), microscopy acoustic scanning (https://en.wikipedia.org/wiki/Scanning\_acoustic\_microscope), ground-penetrating radar technique (https://en.wikipedia.org/wiki/Ground-penetrating\_radar) However, the scale of the Paldiski reactor units makes it very challenging to apply them. For example, any type of X-ray scan relies on differences in beam attenuation. At the Paldiski site, reactor unit one has a diameter of 7 m and a length of 15m while unit two is 9m in diameter and is 12m long.



Figure 1. The photon mass attenuation length  $\lambda$  for various elemental absorbers as a function of photon energy.[1]

For X-ray characterization some of the X-rays need to make it across the unit. The intensity *I* remaining after traversal of thickness *t* (in mass/unit area) is given by

$$I = I_0 e^{-\frac{t}{\lambda}} \tag{1}$$

Where  $\lambda$  is the photon mass attenuation length [1]. The density of concrete varies but is typically around 2400 kg/m<sup>3</sup>. The chemical composition of concrete varies, but it is reasonable here to assume that it behaves roughly like silicon for the purposes of photon attenuation. Using the graph and equation (1) the fraction of remaining 1 MeV photons after 7m of concrete is  $3.3 \cdot 10^{-37}$ . The fraction of remaining photons after 7 m of concrete is at its maximum for 10 MeV and reaches  $5.7 \cdot 10^{-19}$ . This shows that it is very challenging to undertake X-ray radiography, simple because of the enormous numbers of photons required. Having said that, replacing 10cm of concrete with 10cm lead reduces the fraction to  $3.61 \cdot 10^{-21}$  so there is enough contrast if enough photons can be produced.

References

[1] C. Patrignani et al. (Particle Data Group), Chin. Phys. C, 40, 100001 (2016).

After discussion experts agreed that whilst there is some merit in undertaking this activity the experts believe the level of accuracy attached to this limited with the most promising option being the deployment of muon tomography to assist in locating sources in the upper most part of Unit-1 i.e. top of unit 1 – reactor closure plate

#### Conclusions:

An assessment of the results and impact of the expert's mission, relevant conclusions, including an evaluation of the degree of success in solving the problems encountered. Provide an analysis and description of any additional training, expert services and equipment that are considered to be necessary if the project's objectives are to be met. Suggestions or recommendations made concerning future work should take into account the advisory role of the IAEA and the limitation on funds that may exist.

The following are recommendations for localization of DSRS's in containers for Paldiski.

Given the dimensions of the two reactor units and the amount of concrete suspected to be inside, muon tomography seems the only feasible technology to make a 3D image of the inside of the reactor units. Muon tomography is still in an early phase: many university groups around the world are working on it using many different detector types and developing many different data analysis algorithms. Commercial offering of the technology is still limited.

Muon tomography relies on multiple scattering. Muons are electrically charged and will undergo random scattering inside material due to the Coulomb interactions with the electron clouds and nuclei. Hence, muons will exit material under an angle. The standard deviation of the angular distribution scales approximately with  $Z^2$ , making the technique very suitable to look for heavy elements. By measuring the incoming and outgoing trajectory of a large set of muons, conclusions can be drawn on the material traversed. The muon flux depends on the angle. The maximum flux at sea level is approximately 100 Hz/m<sup>2</sup>. It falls roughly as  $1/\cos^2$  of the zenith angle. This is shown in Figure 1.



Figure 1. Muon intensity versus muon momentum for different zenith angles [1].

The muons trajectories are measured by placing a stack of detectors on either side of the volume under study. To measure the muon direction accurately, it is beneficial to have a large spacing between the detectors in a stack. At Paldiski, it is possible for both reactor units to place a stack of detectors above and a stack of detectors below the reactor vessels. Above the vessels, space is limited but detector layers can also be installed on top of the sarcophagus. Below the space is limited, which will limit the angular resolution of the outgoing muon track direction. However, the available space is enough to achieve a reasonable resolution on the muon direction, see figure 2.



Figure 2. Top of reactor unit 1 (left) and bottom of reactor unit 1 (right)

At Paldiski the radioactive sources have been placed in 5 boxes: a 400 kg paraffin box, a 1200 kg steel box with lead lining, a 350kg metallic box, 60kg wooden box and a 25kg wooden box. These boxes are quite large (in the order of 1 by 1 by 1  $m^3$ ) and their materials are very different from concrete. As such they should be relatively easy to locate using muon tomography. Furthermore, verbal evidence suggests that these boxes are entombed in concrete on top of the reactor vessel This information has to be verified. Using muon tomography to exclude the presence of these boxes next to the reactor vessel will not require much data and can thus be achieved quite quickly. Without a detailed study, a time estimate is difficult but from experience it is most likely well below one week for the area covered by the two detectors. The 6 months estimate is for a full 3D image of the vessels. To confirm or exclude the presence of these large boxes would take around 1 week of data taking per detector required location. Confirming that the boxes are present above the reactor vessel will take a similar amount of time.

Wooden boxes are difficult to detect as wood because they are filled with metallic material but as they contain relatively small amount of activity ( $\sim 10^5$  Bq) consequences of the worst case scenario (crushing of sources) are relatively low. Please note that several groups have been working successfully on muon tomography for cargo container inspection. In that application the presence of  $\sim 1$  litre sized uranium objects inside cargo containers that can potentially be filled with lead-acid batteries, engine parts and so on needs to be established on the time scale of minutes [2-6]. Although the challenge at Paldiski is different (the reactor unit is much larger than a cargo container) the challenge is actually easier as the background is a reasonably well-known

steel structure and concrete. The paraffin box challenge is similar to the detection of voids inside waste drums, which was successfully addressed in e.g. [7]. Material identification using muon tomography for encapsulated materials is possible as well, see e.g. [8,9], but requires long(er) data taking times.

A key remaining question is whether or not additional small sources have been entombed in unit 1. Without information on their size it is impossible to estimate the time required for their detection. Measuring longer will reveal more structural details. In principle it is possible to construct a full 3D map of the internal structure of the units, however in practise this might take an unfeasibly long time. Given the reliability of the verbal evidence, a muon tomography study should start with the aim of confirming the presence of the five boxes on top of the reactor lid and subsequently demonstrate the absence of larger objects next to the reactor. This will minimize the required data taking time and hence the cost. Muon tomography is a well established technique for scientific applications. However, there are not yet many experienced companies offering it as a service. That is why we recommend to include a certification test in the tendering process. More details on this can be found in Appendix B.

A balance needs to be struck between minimising the data taking time and locating the boxes as quick, and thus cheap, as possible and taking more data to reveal more details of the structure inside the reactor vessel. Knowledge of the exact structure could be of high value to the Estonian authorities.

Given the large sizes of the boxes, the imaging challenge could also be addressed using muon radiography. Radiography relies on the absorption of muons in the material. By comparing the muon rate in a small solid angle of muons that travelled through the reactor vessel with the muon rate in the same solid angle yields the fraction of stopped muons. This depends on the amount of material traversed in the reactor vessel. Repeating the measurement from a different location, allows to reconstruct a full 3D density map. Radiography has the advantage that only a single detector stack is required. This stack will need to be mounted at a distance from the reactor vessel, e.g. on the grass outside the building. The key advantages are that only one, smaller detector is needed, which lowers cost, it can/needs to be done from a distance, so it can be done while the decommissioning is starting. The key disadvantages are that radiography exploits muons with a much larger zenith angle, which have a much lower rate and thus requires much longer data taking, and that the resolution is much worse. However, the boxes are so big that they will be found with radiography as well. Moreover, for more precise localization of boxes and possible small sources entombed in concrete, in the future when opening RC standard techniques as presented on the Figure 16 from IAEA document on "Locating and Characterization of DSRS" could be utilised for relative small (about 0.5 - 1 m) pieces of concrete when actual decommissioning has been starting.

In Appendix A a typical project structure and costing is discussed while Appendix B comments on issues with a potential tender.

#### References

[1] D Reyna. A simple parameterization of the cosmic–ray muon momentum spectra at the surface as a function of zenith angle. arXiv preprint hep-ph/0604145, 2006.

[2] L. J. Schultz et al., "Statistical reconstruction for cosmic ray muon tomography, IEEE Trans. Image Process., vol. 16, no. 8, pp. 1985–1993, 2007. [3] C. Thomay et al. A novel Markov random field-based clustering algorithm to detect High-Z Objects with cosmic rays, IEEE transactions on nuclear science, vol. 62, no. 4, august 2015
[4] K. Gnanvo et al., "Imaging of high-z material for nuclear contraband detection with a minimal prototype of a muon tomography station based on gem detectors," Nucl. Instrum. Methods Phys. Res. A, vol. 652, no. 1, pp. 16–20, 2011.

[5] K. Borozdin et al., Cosmic-ray muon tomography and its application to the detection of high-Z materials , in proceedings of 46th Annual Meeting , Institute of Nuclear Materials Management, Phoenix, AZ, U.S.A. (2005).

[6] C. Thomay et al. A binned clustering algorithm to detect high-Z material using cosmic muons, 2013 JINST 8 P10013

[7] M. Dobrowolska et al. A novel technique for finding gas bubbles in the nuclear waste containers using Muon Scattering Tomography, 2018 JINST 13 P05015

[8] L. Frazão et al, Discrimination of high-Z materials in concrete-filled containers using muon scattering tomography, 2016 JINST 11 P07020

[9] L.J. Schultz et al., Image reconstruction and material Z discrimination via cosmic ray muon radiography, Nucl. Instrum. Meth. A 519 (2004) 687

#### Appendix A: A typical project structure and cost indication

To guide the discussion and assist ALARA, here an outline of the project is given which gives an indication of the expected timeline and cost in an optimistic scenario.

A typical muon tomography project for a challenge like scanning the Paldiski reactor units would start with a short Monte Carlo study to determine the required data taking times and optimisation of the mechanical structure for the muon detector stack above the unit. Such a study would typically take 3 months.

Given the scale of the reactor units, a detector system covering several square meters area ( $\sim 10m^2$  or larger) seems required to minimize the required movement of detectors to cover the total area to be scanned. Several university groups and companies have the knowledge and expertise to deliver such a system. A typical time from project start to full operation is between 6 months and a year. To cover the relevant area, the detectors probably have to be moved several times. The measurement time per location is difficult to estimate but will be less than one month. Assuming 6 locations per unit yields a total measurement time per unit. Please note that this depends very strongly on the exact question asked: if the verbal evidence is correct and the question is to confirm that indeed the 5 boxes are where they are expected to be and to confirm that there are no other containers of significant size (coffee mug or larger) present, the measurement time can be a lot shorter.

Finally, all data will need to be analysed and a report written. This is estimated to take overall about 4 months.

A reasonable time scale for completion from scratch of a project like the scanning of both Paldiski units, would be around 2 years and in an optimistic, non-commercial scenario be around  $\pm 1.2M$ .

#### Appendix B: Tendering for the scanning of the Paldiski reactor units

An inherent challenge when tendering for the work to locate the sources is that it is impossible to verify the results until the reactor units are fully decommissioned. As muon tomography is not yet a fully established commercial technology that has routinely been applied to projects of this scale and nature, it is difficult for contractors to produce a range of references demonstrating that similar work was delivered successfully before.

As such, we suggest to ALARA to undertake a certification exercise for potential contractors. It is possible using standard simulation tools for muon tomography (e.g. CRY library to generate a realistic muon flux and GEANT4 to simulate the passage of the muons through material) to simulate real data appropriate for the contractors proposal detector option without revealing the exact geometry of the inside of the reactor unit. The contractors would receive a set of event data, i.e. hit locations in their detectors due to muons and background radiation, fully equivalent to the data they would obtain. The challenge for the contractors would be to locate a set of source boxes. ALARA can then check whether the correct locations are returned.

[B1] C. Hagmann, D. Lange, and D. Wright, "Cosmic-ray shower generator (CRY) for monte carlo transport codes," in Proc. IEEE Nuclear Science Symp. Conf. Rec., 2007, vol. 2, pp. 1143–1146 [Online]. Available: http://dx.doi.org/10.1109/NSSMIC.2007.4437209, ISSN 1095-7863
[B2] S. Agostinelli et al., "Geant4-a simulation toolkit," Nucl. Instrum. Methods Phys. Res. A, vol. 506, no. 3, pp. 250–303, 2003.

### **Recommendations:**

NOTE: Each group of recommendations is a separate table. Please enter each recommendation in a separate row in the table. To enter a new row within each table, press the "TAB" key.

#### **Recommendations to the Counterpart Institution and National Counterpart:**

Discussions were held re the relative merits of undertaking the decommissioning post recovery of the 'high hazard sources' i.e. those on top of the closure plate unit 1 and the relative risks associated with any remaining sources in the body of unit 1.

- 1. Muon tomography provides the best opportunity to give internal images of the reactor compartments given the concrete cover.
- 2. It is recommended a certification trial is undertaken prior to full scale deployment of Muon Tomography.
- 3. It should be noted that variety of imaging techniques for DSRS location could be utilized after decommissioning of sarcophagus starts and access to top of Unit 1 at reactor compartment becomes available.
- 4. It is the view of the experts that the risks associated with these sources are minimal and should not constrain the wider options for decommissioning of both units.

## **Recommendations to the Government:**

Consideration should be given to propose to IAEA national technical cooperation project to utilize muon tomography for Paldiski for full imaging of structures of the units 1 and 2 and also localization of DSRS boxes in embedded concrete of the Unit 1 reactor compartment.

## **Recommendations to the Agency:**

National technical cooperation project should be considered to assist A.L.A.R.A. in their decommissioning efforts for Paldiski including utilisation of muon tomography to localise internal structure of reactor compartment units.