DD-R-15(EN)

DANSK DEKOMMISSIONERING

Clearance Laboratory

Capability and measurement sensitivity

Per Hedemann Jensen, Bente Lauridsen, João Silva, Jens Søgaard-Hansen, Lisbeth Warming

Danish Decommissioning, Roskilde
September 2005

DD-R-15(EN)

Clearance Laboratory

Capability and measurement sensitivity

Per Hedemann Jensen, Bente Lauridsen, João Silva
Jens Søgaard-Hansen, Lisbeth Warming

Danish Decommissioning, Roskilde
September 2005

Abstract A new low-level Clearance Laboratory has been built at the Risø-site. Building materials with a low content of naturally occurring radionuclides have been used. To minimize transport of radon gas from soil into the laboratory the foundation has been supplied with a membrane.

The laboratory has been equipped with two high-efficiency germanium detectors. These detectors will be used for clearance measurements on the predicted amount of 15,000 - 18,000 tonnes of non-active or nearly non-active materials, which will originate from the decommissioning of all the nuclear facilities at the Risø-site. They will be used also for clearance measurements on buildings and land.

Objects and materials to be measured for clearance are placed on a rotation table that can carry up to one tonne and can rotate once a minute to simulate some averaging of inhomogeneously distributed activity. Sensitivity and background measurements reveal that measuring times of 20 - 50 minutes would normally be sufficient to detect radionuclide concentrations of only a small fraction of the nuclide-specific clearance levels with a sufficiently low uncertainty.

Probability calculations of the measurement capacity of the Clearance Laboratory indicate that the mean value of the total measuring time for all materials that potentially can be cleared would be 13 years with a 95% probability of being less than 25 years. The mean value of the annual amount of materials that can be measured in the laboratory is 600 tonnes with a 95% probability of being less than 1,200 tonnes. If needed, there is room for additional measuring systems to increase the capacity of the laboratory.

ISBN 87-7666-020-6; ISBN 87-7666-021-4 (Internet)

Print: Pitney Bowes Management Services Danmark A/S, 2005

Contents

1 Introduction

A new laboratory - the Clearance Laboratory - has been built at the Risø-site. The laboratory will be used for clearance measurements off materials originating from decommissioning the nuclear facilities at the site: the research reactors DR 1, DR 2 and DR 3, the Hot Cells, the Fuel Fabrication facility and the Waste Treatment Plant. The laboratory is placed close to Roskilde Fjord in a suitable distance from the nuclear facilities to minimize any disturbing γ-radiation from these facilities during their decommissioning. The Clearance Laboratory is classified as a 'low-level laboratory' and provided with changing room and bathroom facilities to minimize the probability of cross contamination from the nuclear facilities.

2 Clearance Laboratory

The Clearance Laboratory has three main sections: (a) an entrance section where the materials to be measured are received, (b) a hall for measurements, and (c) a control room in which the measurements can be controlled from computers. The laboratory is shown in Fig. 1.

Figure 1. Clearance Laboratory. The materials enter the laboratory via the entrance section at the front of the building.

Entrance section. Materials and objects to be measured are transported to the laboratory by a fork-lift truck or a small lorry that are used solely for transporting potential non-radioactive materials in order to minimize the probability of contaminating materials and measurement hall. The entrance section has four gates: entrance, exit and two gates to the measurement hall. The gates are interconnected so that only one gate can be open at a time. A crane or truck in the measurement hall carries the materials to the measuring equipment. Before being transported to the laboratory, it will be controlled that no materials and objects to be measured in the laboratory are externally contaminated with non-fixed activity or emits measurable γ-radiation, and even so, the objects will be wrapped up in plast to avoid contamination of the measuring hall.

Hall for measurement. During the clearance measurements in the hall no other materials or objects than those being measured are allowed to be stored in the building. The total measuring time for all materials and objects that potentially

can be cleared will depend on the total amount of materials from the decommissioning of the nuclear facilities and the number of measuring setups in the laboratory (see Section 6). The laboratory has from the onset been supplied with two measuring setups but has been built so it can contain one or two additional measuring setups.

Figure 2. Measuring hall with shielding walls separating the germanium detectors. The shielding walls and the floor have been made of a special concrete with a very low content of ^{40}K, ^{238}U and ^{232}Th. The windows between the measuring hall and the control room can be seen in the background.

The dimensions of measuring hall is approximately 9×24 m^2 and a height allowing a moveable crane in the ceiling to lift rather large objects to the measuring tables (maximum 1 tonne). The measuring setups can be separated in sections by moveable shielding walls as shown in Fig. 2. These moveable concrete walls have been constructed from materials with a low content of the naturally occurring radionuclides ^{40}K, ^{238}U and ^{232}Th. The floor in the hall has been made of the same sort of concrete and has been coated with epoxy so that potential surface contamination will be easy to remove. The foundation of the laboratory has been supplied with a membrane to minimize the transport of radon gas from the soil into the laboratory.

To avoid outside contamination entering the measuring hall, the access to the laboratory takes place through a changing room where the staff will change shoes (cover shoes).

Background spectra in the measurement hall will be measured frequently and surfaces will often be checked for contamination. These measurements will assure that the laboratory remains clean and maintains its classification as 'low-level laboratory'. In addition, the detector characterisation (basic calibration) will be checked regularly.

Control room. The staff will stay in the control room (shown in Fig. 3) during all measurements. All communication with the measuring setups takes place from computers in the control room. This include selection of measuring time, material composition and density of the 'source'-material, material-detector geometry and activity distribution (template) and preparation of analysis reports.

In conjunction with the control room there is a library/storage room supplied with movable shelves for filing paper copies of the analysis reports.

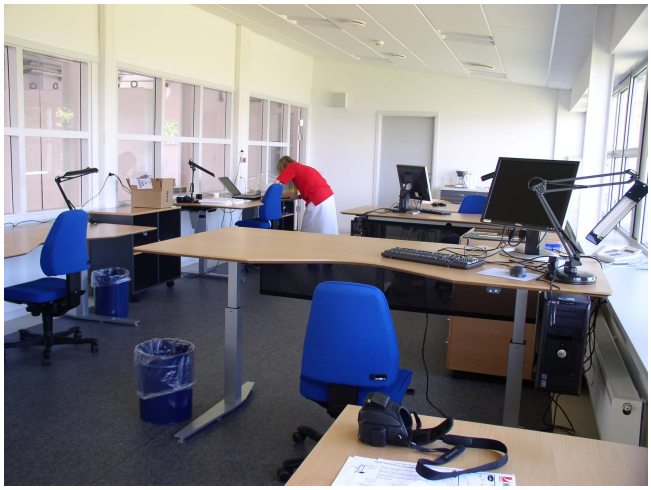

Figure 3. Control room at the Clearance Laboratory with computers connected to the germanium detectors in the measuring hall. Selection of measurement geometry, setting measuring time and all data processing of the measurements are carried out from the control room.

3 Measuring equipment

The γ-spectrometric measurements in the Clearance Laboratory are performed by two large germanium detectors with an efficiency of 100% relative to a 4×4 inch sodium iodide detector. The energy resolution (FWHM) is less than 2.2 keV at a photon energy of 1330 keV, and the detectors are applicable in the energy range of 10 keV to 7 MeV (the ISOCS software from 45 keV to 7 MeV). The detectors can be shielded with 2.5 cm and 5 cm circular lead shields, which can be arranged to give different apertures as shown in Fig. 4.

Figure 4. Germanium-detector with circular lead shield around the detector.

The analysis of the measured spectra and the determination of the activity content in the different objects is based on the characterisation of the germanium detector by the supplier. The detector response function has been calculated in 200 - 300 points around the detector up to 500 metres from the detector. For each point the response function has been calculated for 8 - 10 photon energies. The calculated values have been verified by the supplier using certified point sources.

The activity in the measured objects is determined from the software system ISOCS [1] in which some twenty source geometries (templates) and a radionuclide library are integrated. The sensitivity of the system depends on the object-detector distance, measuring time and size and density of the object. For most objects, an activity concentration significant below the clearance level can be determined with a measuring time of 15 - 30 minutes (see Section 5).

Figure 5. Germanium detector with a 100% efficiency relative to a 4×4 inch sodium iodide detector. The detector is surrounded by a circular lead shield of 50 mm thickness. The objects are placed on the rotation table in front of the detector.

The objects are placed on a rotation table that will simulate an averaging of inhomogeneously distributed activity. The table can rotate once a minute and is designed to carry objects with a weight up to one tonne.

The germanium detectors will be applied also for clearance measurements on buildings and land [2].

4 Background measurements

Background measurements have been made in the measuring hall, both with unshielded and shielded detectors. As the orientation of the detectors during the measurements will always be horizontal, the detectors were placed horizontally at different locations in the hall and pointing in different directions. For naturally occurring radionuclides, no significant differences were observed between the measured background spectra at these locations and orientations. For ^{137}Cs, the variation was somewhat higher, about a factor of 2 - 3, with the lowest values, when the detector was pointing towards Roskilde Fjord.

One of the background spectra, which was obtained by measuring over 65 hours with an unshielded detector, is shown in Fig. 6.

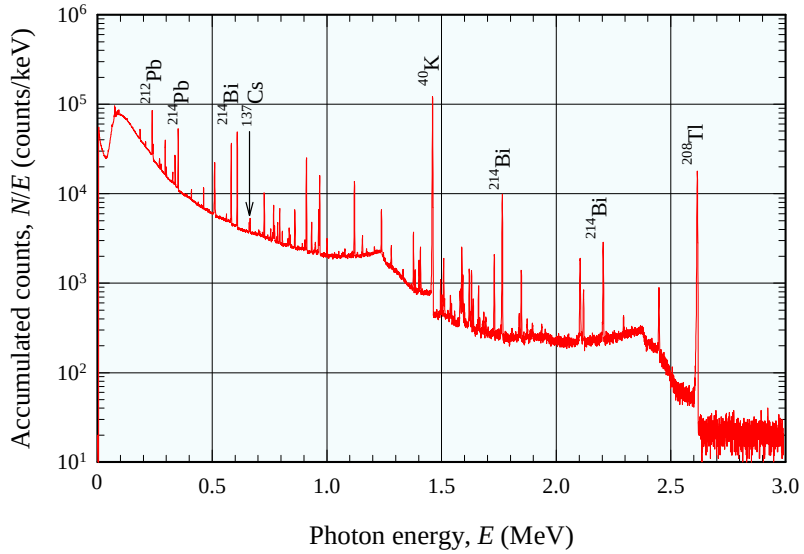

Figure 6. Background spectrum for a measurement time of 65 hours with no lead shield around the detector.

One of the background spectra, which was obtained by measuring over 50 hours with a detector shielded by 5 cm lead, is shown in Fig. 7.

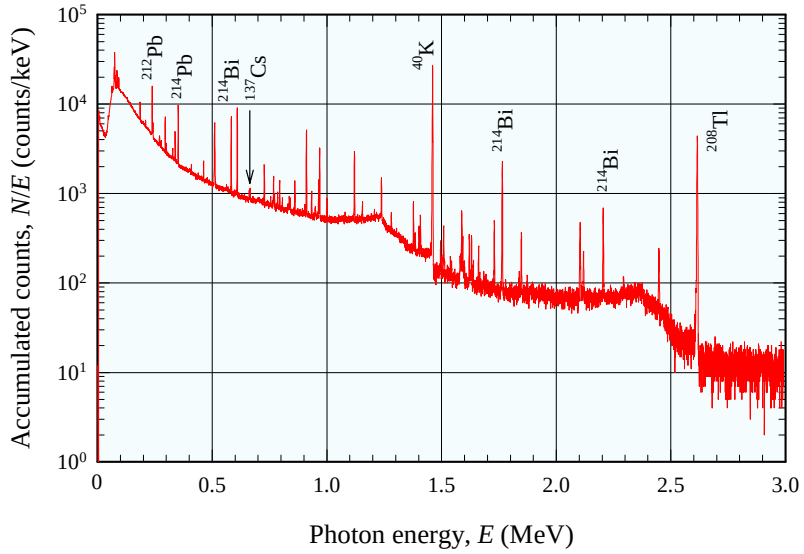

Figure 7. Background spectrum for a measurement time of 50 hours with the detector shielded by a circular lead shield of 50 mm thickness as shown in Fig. 5.

The major radionuclides detected in the two background spectra are radionuclides from the ^{238}U- and ^{232}Th-decay chains and ^{40}K. The detected ^{137}Cs in the background spectra is the remnant of fallout from the Chernobyl accident and from the nuclear weapons testing in the atmosphere in the 1950's and 1960's.

The net background count rates for the dominant peaks in the spectra are shown in Table 1.

Table 1. Net count rates for the dominating radionuclides in the background spectrum measured with a shielded and an unshielded detector.

Radionuclide	Energy [MeV]	Net count rate [cps]	
		Unshielded	Shielded
^{40}K	1.461	$3.2{\cdot}10^{0}$	$9.2{\cdot}10^{-1}$
^{212}Pb	0.238	$9.5{\cdot}10^{-1}$	$2.3{\cdot}10^{-1}$
^{208}Tl	2.616	$6.1{\cdot}10^{-1}$	$1.9{\cdot}10^{-1}$
^{214}Bi	0.609	$8.5{\cdot}10^{-1}$	$2.1{\cdot}10^{-1}$
	1.764	$2.7{\cdot}10^{-1}$	$8.0{\cdot}10^{-2}$
	2.204	$8.2{\cdot}10^{-2}$	$2.4{\cdot}10^{-2}$
^{214}Pb	0.352	$7.5{\cdot}10^{-1}$	$1.7{\cdot}10^{-1}$
^{137}Cs	0.662	$2.1{\cdot}10^{-2}$	$5.8{\cdot}10^{-3}$

It appears from the background measurements that, depending on the photon energy, the net count rates in the full energy peaks will be reduced by a factor of 3 - 4 by the 50 mm lead shield.

5 Sensitivity of clearance measurements

The sensitivity of the germanium detectors can for different radionuclides be expressed in terms of a minimum detectable activity, *MDA*, for given geometries and measuring times. The *MDA* is the smallest level of activity that can be detected with 95% confidence, while also having 95% confidence that "activity" is not detected from a null sample (L.A. Currie [3]).

5.1 Minimum detectable activity

The minimum detectable activity, *MDA*, for a given object can be lowered in three ways:

- ◇ increasing the efficiency of detection, *i.e.* by decreasing the detector to object distance
- ◇ decreasing the background using shielding of the detector
- ◇ increasing the measuring time

The energy-integrated gross counts, N, is the sum of the background counts, N_B, and the source-related net counts in the full energy peak, ΔN_S, as indicated in Fig. 8, and it is calculated as:

$$N = \int_{E_1}^{E_2} n(E)\ \mathrm{d}E = N_B + \Delta N_S \tag{1}$$

where $n(E)$ is the time-accumulated counts per unit energy (see Fig. 8) and $[E_1; E_2]$ is the energy range of the full energy peak.

The energy-integrated source-related net counts in the full energy peak, ΔN_S, is proportional to the activity, Q, in the source and the measuring time, T:

$$\Delta N_S = k_1 \cdot Q \cdot T \tag{2}$$

The energy-integrated background counts, N_B, is proportional to the measuring time, T:

$$N_B = k_2 \cdot T \tag{3}$$

The energy-integrated source-related net counts in the full energy peak, ΔN_S, is just significant at the 95% confidence level when:

$$\begin{aligned} \Delta N_{S,\,\mathrm{sign}} &\approx 2 \cdot \sqrt{N_B} \\ &= 2 \cdot \sqrt{k_2\, T} \end{aligned} \tag{4}$$

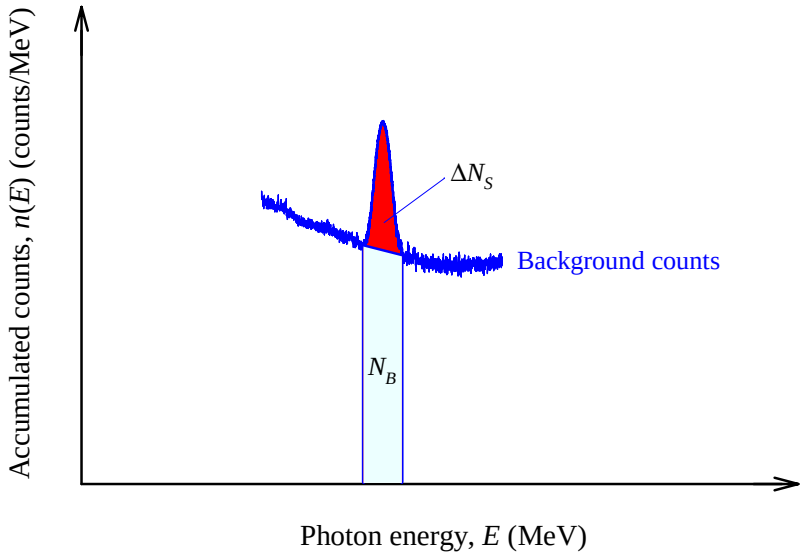

Figure 8. Time-accumulated counts, $n(E)$, as a function of photon energy in a gamma-spectrum. The energy-integrated source-related net counts, ΔN_S, in the full energy peak is given by the red-colored area and the energy-integrated background counts, N_B, is given by the light-blue-colored area.

As the significant activity content in the measured object, Q_{sign}, is related to the significant energy-integrated net counts in the full energy peak, $\Delta N_{S,\,\mathrm{sign}}$, by Eq. (2), it follows that the significant activity content is inverse proportional to the square root of the measuring time, T:

$$Q_{\mathrm{sign}} = \frac{k_3}{\sqrt{T}} \tag{5}$$

It follows from Eq. (5) that Q_{sign} for two different measuring times, T_1 and T_2, is:

$$Q_{\mathrm{sign}}(T_1) = \frac{k_3}{\sqrt{T_1}} \quad \wedge \quad Q_{\mathrm{sign}}(T_2) = \frac{k_3}{\sqrt{T_2}} \tag{6}$$

and, consequently, the significant activity content for different measuring times T_1 and T_2 and for the same source-detector geometry are related as:

$$Q_{\mathrm{sign}}(T_2) = Q_{\mathrm{sign}}(T_1) \cdot \sqrt{\frac{T_1}{T_2}} \tag{7}$$

Assuming that the minimum detectable activity, MDA, is equal to Q_{sign}, two different measuring times, *e.g.* $T_2 = 2 \cdot T_1$, will give minimum detectable activities related as $MDA(T_2) = MDA(T_1)/\sqrt{2}$.

For actual spectra collected in the Clearance Laboratory the *MDA* has been determined for a number of geometries and radionuclides using the software Genie 2000 [4] to analyse the smallest level of activity which can be detected with 95% confidence.

5.2 Measuring geometries

With the ISOCS software [1] it is possible to characterize the measured objects and materials in more than twenty different templates (geometries) for which the material composition and the assumed activity distribution has to be specified [2]. These geometries are sufficient to model the objects and materials originating from the decommissioning of the nuclear facilities at the Risø-site.

For a selected geometry, a response function is calculated based upon the characterisation of the detector. From the measured background-corrected γ-spectrum and the response function the nuclide-specific activity concentration is calculated.

The sensitivity of the germanium detector systems has been investigated for eight different geometries based on the minimum detectable activity, *MDA*, as specified above. This quantity defines the minimum detectable concentration as $MDC = MDA/M$, where M is the mass of the object. During measurements the detectors are surrounded by a 50 mm circular lead shield and the gamma-spectrum is corrected for background contribution measured over a long time period. The values of *MDC* for eight different radionuclides are shown in Table 2 for a measuring time of 15 minutes.

Table 2. Minimum detectable concentrations, MDC, for eight typical radionuclides distributed in eight different geometries. The volume, mass, height, diameter, length and width are given as V, M, H, D, L and W. The measuring time is 15 minutes.

Geometry	Geometry and activity distribution	MDC [Bq/g]
CYLINDER $V = 10\ \ell$ $M = 23.5$ kg $H = D$	The activity is homogeneously distributed in a concrete cylinder. During measurement the detector axis is at a right angle to the cylinder axis and points to the middle of the cylinder. The distance from the detector to the surface of the cylinder is equal to the cylinder height, H.	^{40}K: $1.5{\cdot}10^{-1}$ ^{51}Cr: $8.5{\cdot}10^{-2}$ ^{60}Co: $5.3{\cdot}10^{-3}$ ^{65}Zn: $1.7{\cdot}10^{-2}$ ^{110m}Ag: $6.9{\cdot}10^{-3}$ ^{133}Ba: $1.7{\cdot}10^{-2}$ ^{137}Cs: $8.3{\cdot}10^{-3}$ ^{152}Eu: $2.8{\cdot}10^{-2}$
CYLINDER $V = 100\ \ell$ $M = 235$ kg $H = D$	The activity is homogeneously distributed in a concrete cylinder. During measurement the detector axis is at a right angle to the cylinder axis and points to the middle of the cylinder. The distance from the detector to the surface of the cylinder is equal to the cylinder height, H.	^{40}K: $1.1{\cdot}10^{-1}$ ^{51}Cr: $7.3{\cdot}10^{-2}$ ^{60}Co: $4.0{\cdot}10^{-3}$ ^{65}Zn: $1.3{\cdot}10^{-2}$ ^{110m}Ag: $5.6{\cdot}10^{-3}$ ^{133}Ba: $1.4{\cdot}10^{-2}$ ^{137}Cs: $6.7{\cdot}10^{-3}$ ^{152}Eu: $2.1{\cdot}10^{-2}$
CYLINDER $V = 10\ \ell$ $M = 23.5$ kg $H = D$	The activity is placed as a point source at the top of a concrete cylinder at the cylinder axis. During measurement the detector axis is at a right angle to the cylinder axis and points to the middle of the cylinder. The distance from the detector to the cylinder surface is equal to the cylinder height, H.	^{40}K: $3.1{\cdot}10^{-1}$ ^{51}Cr: $4.7{\cdot}10^{-1}$ ^{60}Co: $1.2{\cdot}10^{-2}$ ^{65}Zn: $4.0{\cdot}10^{-2}$ ^{110m}Ag: $2.2{\cdot}10^{-2}$ ^{133}Ba: $8.5{\cdot}10^{-2}$ ^{137}Cs: $2.7{\cdot}10^{-2}$ ^{152}Eu: $5.9{\cdot}10^{-2}$

Table 2. Continued.

Geometry	Geometry and the activity distribution	MDC [Bq/g]
CYLINDER $V = 100\ \ell$ $M = 235$ kg $H = D$	The activity is placed as a point source at the top of a concrete cylinder at the cylinder axis. During measurement the detector axis is at a right angle to the cylinder axis and points to the middle of the cylinder. The distance from the detector to the cylinder surface is equal to the cylinder height, H.	^{40}K: $7.8\cdot10^{-1}$ ^{51}Cr: $7.2\cdot10^{0}$ ^{60}Co: $3.2\cdot10^{-2}$ ^{65}Zn: $1.3\cdot10^{-1}$ ^{110m}Ag: $1.1\cdot10^{-1}$ ^{133}Ba: $1.1\cdot10^{0}$ ^{137}Cs: $1.5\cdot10^{0}$ ^{152}Eu: $1.5\cdot10^{-1}$
FLAT BOX $V = 10\ \ell$ $M = 23.5$ kg $L = H = 10\cdot W$	The activity is homogeneously distributed in a flat concrete box. During measurement the detector axis is parallel to the surface normal of the major surface of the box and pointing to the center of the surface area. The distance from the detector to the major box surface is equal to the box height, H.	^{40}K: $1.7\cdot10^{-1}$ ^{51}Cr: $7.6\cdot10^{-2}$ ^{60}Co: $6.0\cdot10^{-3}$ ^{65}Zn: $1.8\cdot10^{-2}$ ^{110m}Ag: $6.8\cdot10^{-3}$ ^{133}Ba: $1.5\cdot10^{-2}$ ^{137}Cs: $8.2\cdot10^{-3}$ ^{152}Eu: $3.0\cdot10^{-2}$
FLAT BOX $V = 100\ \ell$ $M = 235$ kg $L = H = 10\cdot W$	The activity is homogeneously distributed in a flat concrete box. During measurement the detector axis is parallel to the surface normal of the major surface of the box and pointing to the center of the surface area. The distance from the detector to the major box surface is equal to the box height, H.	^{40}K: $1.0\cdot10^{-1}$ ^{51}Cr: $5.5\cdot10^{-2}$ ^{60}Co: $3.6\cdot10^{-3}$ ^{65}Zn: $1.1\cdot10^{-2}$ ^{110m}Ag: $4.5\cdot10^{-3}$ ^{133}Ba: $1.1\cdot10^{-2}$ ^{137}Cs: $5.4\cdot10^{-3}$ ^{152}Eu: $1.9\cdot10^{-2}$
FLAT BOX $V = 10\ \ell$ $M = 23.5$ kg $L = H = 10\cdot W$	The activity is distributed as a slab in the middle of a flat concrete box. During measurement the detector axis is parallel to the surface normal of the major surface of the box and pointing to the center of the surface area. The distance from the detector to the major box surface is equal to the box height, H.	^{40}K: $1.8\cdot10^{-1}$ ^{51}Cr: $8.2\cdot10^{-2}$ ^{60}Co: $6.2\cdot10^{-3}$ ^{65}Zn: $1.9\cdot10^{-2}$ ^{110m}Ag: $7.1\cdot10^{-3}$ ^{133}Ba: $1.6\cdot10^{-2}$ ^{137}Cs: $8.6\cdot10^{-3}$ ^{152}Eu: $3.3\cdot10^{-2}$
FLAT BOX $V = 100\ \ell$ $M = 235$ kg $L = H = 10\cdot W$	The activity is distributed as a slab in the middle of a flat concrete box. During measurement the detector axis is parallel to the surface normal of the major surface of the box and pointing to the center of the surface area. The distance from the detector to the major box surface is equal to the box height, H.	^{40}K: $1.1\cdot10^{-1}$ ^{51}Cr: $7.4\cdot10^{-2}$ ^{60}Co: $3.9\cdot10^{-3}$ ^{65}Zn: $1.2\cdot10^{-2}$ ^{110m}Ag: $5.3\cdot10^{-3}$ ^{133}Ba: $1.4\cdot10^{-2}$ ^{137}Cs: $6.4\cdot10^{-3}$ ^{152}Eu: $2.0\cdot10^{-2}$

For the eight geometries the ratio of the radionuclide-specific minimum detectable concentration, *MDC*, to the clearance level, *CL* [5], is calculated as:

$$\frac{MDC}{CL} = \frac{MDA}{M \cdot CL} \tag{8}$$

where *MDA* is the minimum detectable activity for the given object and M is the mass of the object. The values of *MDC/CL* are shown on Fig. 9 - 12 as a function of the measuring time, T, for the eight radionuclides given in Table 2.

The conditions for materials and objects to be cleared is that the mass of the objects, M, should be less than one tonne and that the sum of the ratios of measured nuclide-specific concentrations, C, in the object to the corresponding

clearance levels, CL, should be less than or equal to 1:

$$\sum_{i=1}^{N}\left(\frac{C_i}{CL_i}\right) \leq 1 \tag{9}$$

Due to the uncertainty of the measurement of activity concentration it is necessary to include this uncertainty in the clearance criterion in Eq. 9 so there is a 95% probability that the measured objects have activity concentrations below the clearance level. Assuming that each of the measured activity concentrations, C_i, has its own normal distribution, and that the standard deviation for each distribution is σ_i, being equal to the uncertainty of the measured concentration, the extended clearance criterion can be expressed as [2]:

$$\sum_{i}\left(\frac{C_i}{CL_i}\right) + 1.65 \cdot \sqrt{\sum_{i}\left(\frac{\sigma_i}{CL_i}\right)^2} \leq 1 \tag{10}$$

The measurement time necessary to comply with the clearance criterion in Eq. (10) will depend on how close the concentrations are to the clearance levels. For small values of C_i/CL_i, rather large uncertainties, σ_i, would be acceptable without violating the clearance criterion in Eq. (10). On the other hand, for values of C_i/CL_i close to one, long measurement times might be necessary to respect a 95% probability that the concentrations would comply with the clearance criterion.

Radionuclide-specific mass-specific clearance levels, CL, are given in [5]. For the radionuclides given in Table 2 the mass-specific clearance levels are as follows:

^{60}Co, ^{137}Cs, ^{152}Eu:	0.1	Bq/g
^{133}Ba:	1	Bq/g
^{40}K, ^{65}Zn, ^{110m}Ag:	10	Bq/g
^{51}Cr:	100	Bq/g

Radionuclide-specific surface-specific clearance levels, CL, are given in [6]. For the radionuclides given in Table 2 the surface-specific clearance levels are as follows:

^{60}Co, ^{137}Cs, ^{152}Eu, ^{65}Zn, ^{110m}Ag:	1 Bq/cm^2
^{40}K:	10 Bq/cm^2

For the pure β-emitters ^{3}H, ^{14}C and ^{90}Sr the surface-specific clearance levels are 2 - 3 orders of magnitude higher than those for ^{60}Co and ^{137}Cs.

A. Cylindrical geometry

It appears from Fig. 9 that the minimum detectable concentration, *MDC*, for the selected radionuclides at a measuring time, T, of *e.g.* 20 minutes ranges from about 0.1% - 20% of the corresponding clearance level, *CL*, depending on the radionuclide.

The values of *MDC/CL* (and the values of *MDC*) are less for a 100 ℓ concrete cylinder compared to a 10 ℓ concrete cylinder due to the fact that the total activity content is 10 times less in the smaller of the two cylinders.

The dominating nuclides in neutron-activated concrete are ^{60}Co, ^{152}Eu and ^{133}Ba. If the measuring time for activated concrete that potentially can be cleared is set so the concentration of ^{152}Eu can be determined with a high confidence, concentrations of ^{133}Ba and ^{60}Co being 2 - 5 times lower than that of ^{152}Eu will be detected with a similar high confidence (see values of *MDC* in Table 2).

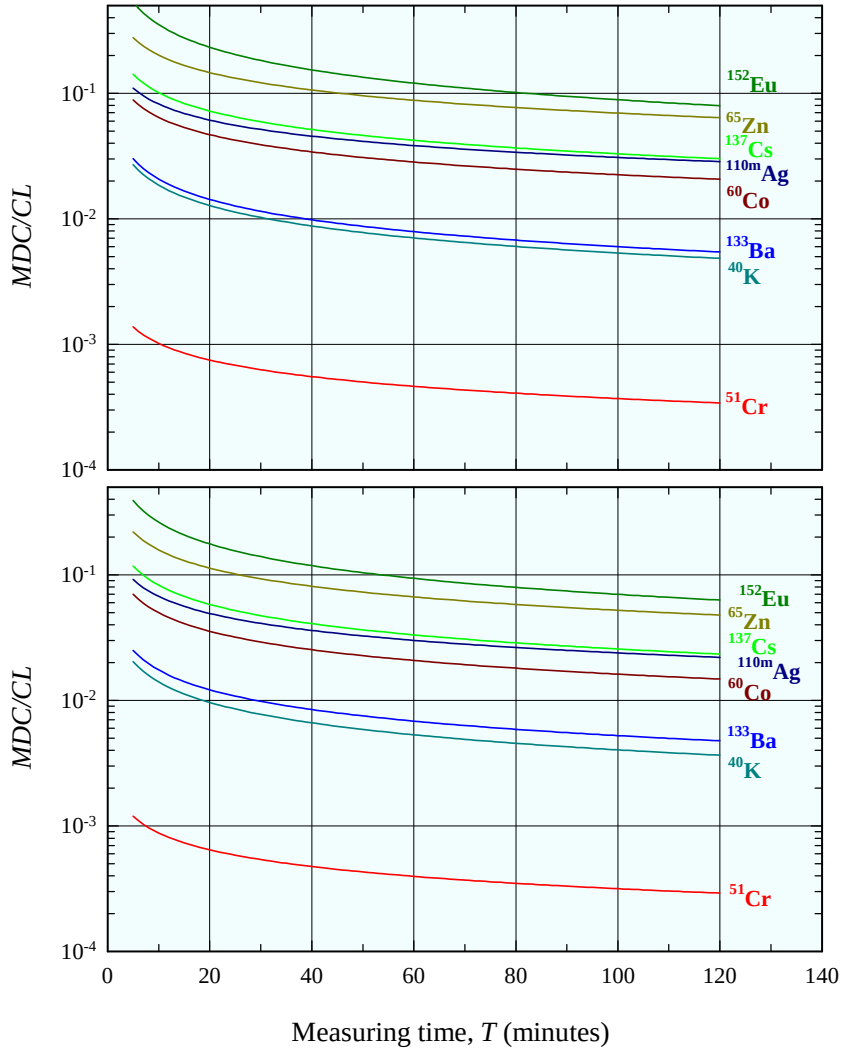

Figure 9. Ratio of the minimum detectable concentration, MDC, to the clearance level, CL, as a function of the measuring time with a germanium detector for different radionuclides homogeneously distributed in a 10 ℓ concrete cylinder (upper graph) and in a 100 ℓ concrete cylinder (lower graph).

B. Cylindrical geometry with point source

If concrete is contaminated in spots only at the surface, the value of *MDC* would be rather low if the object is rotated during the measurement. If, however, the spot is "hidden" for the detector, a substantial self-shielding by the object will increase the value of *MDC* considerably.

This is shown in Fig. 10 for a geometry where the activity is placed as a point source at the top of a 10 ℓ and a 100 ℓ concrete cylinder at the cylinder axis. It appears from the figure that the minimum detectable concentration, *MDC*, at small measuring times will exceed the clearance level for some of the selected radionuclides placed on top of a 100 ℓ cylinder and that several hours of measuring therefore would be necessary. Only for ^{51}Cr and ^{60}Co is the value of *MDC* lower than the clearance level also for low measuring times. Due to a lower self-shielding in a 10 ℓ concrete cylinder the value of *MDC* will be less than the *MDC* for a 100 ℓ cylinder, but for some nuclides, a 30 - 60 minutes measuring time might be necessary.

The geometry is not realistic and is included here only for illustration of the capability of the measuring system. If the activity being placed at the top of a 100 ℓ

concrete were ^{137}Cs, the value of *MDC* at a measuring time of 20 minutes would correspond to a point source activity of about 35 kBq. This activity would easily have been detected by the contamination/radiation screening before the material would have been transported to the Clearance Laboratory.

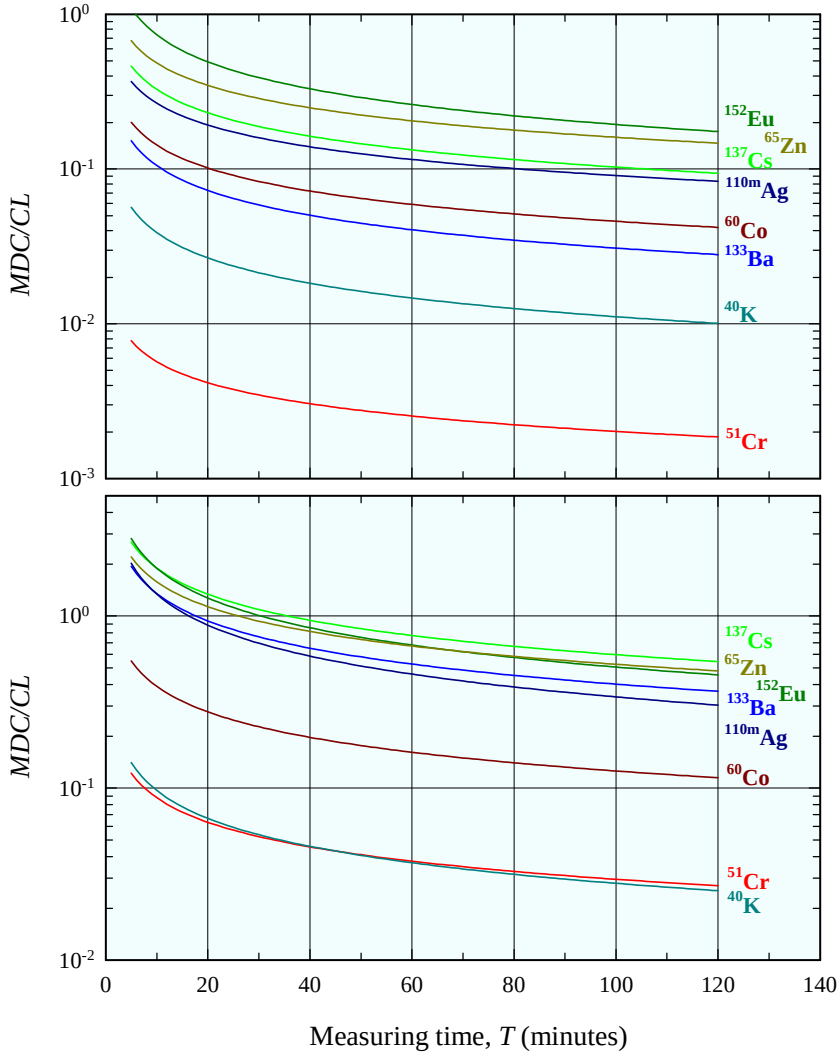

Figure 10. Ratio of the minimum detectable concentration, MDC, to the clearance level, CL, as a function of the measuring time with a germanium detector for different radionuclides placed as a point source at the top of a 10 ℓ concrete cylinder (upper graph) and at the top of a 100 ℓ concrete cylinder (lower graph) in the centerline of the cylinder.

C. Flat box geometry

Comparing the results in Fig. 9 and Fig. 11 reveals that the *MDC* for a cylindrical and a flat box geometry is rather equal when the detector is pointing towards the major surface of the box. In addition, a flat concrete box with a 100 ℓ volume will have a lower value of *MDC* compared to a 10 ℓ concrete box due to a higher activity content in the larger box.

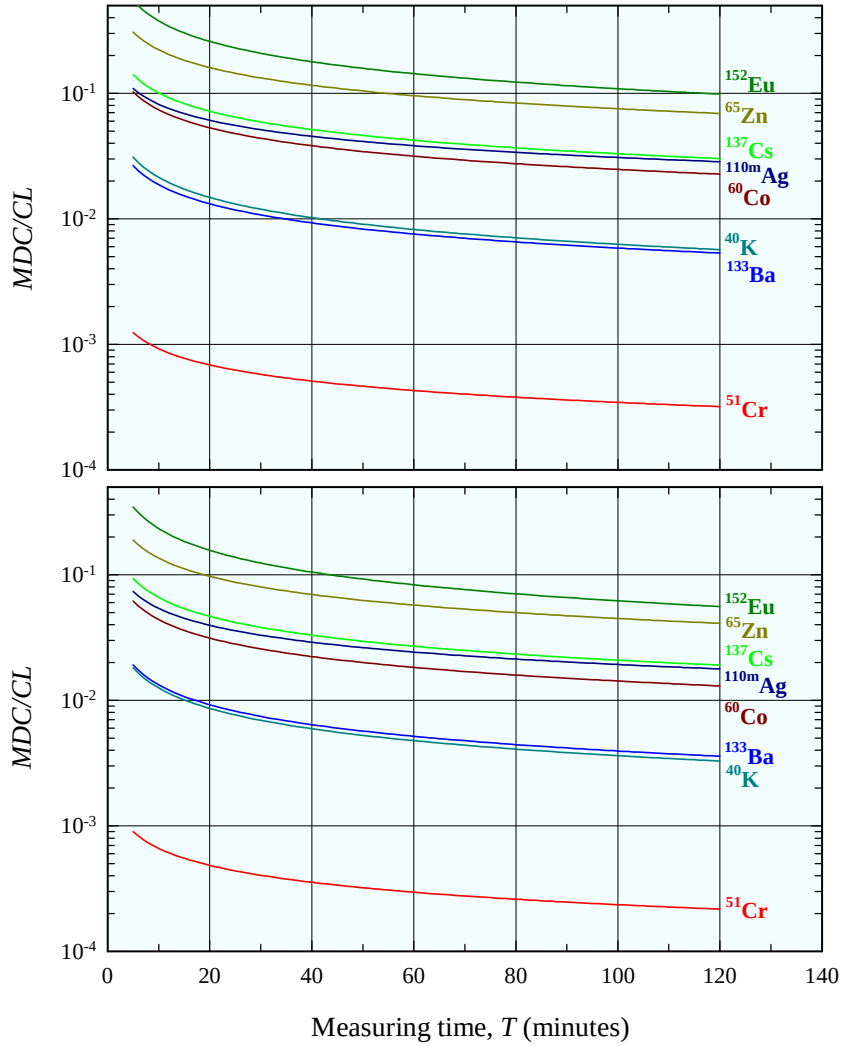

Figure 11. Ratio of the minimum detectable concentration, MDC, to the clearance level, CL, as a function of the measuring time with a germanium detector for different radionuclides homogeneously distributed in a 10 ℓ flat concrete box (upper graph) and in a 100 ℓ flat concrete box (lower graph).

Even if the activity content was embedded as a slab in the middle of the flat concrete box, the value of the *MDC* would only increase marginally which appears by comparing the results in Fig. 11 and 12. The reason is that a flat box ($L = H = 10 \cdot W$) has a lower self-shielding than a more cubic shaped box.

D. Flat box geometry with embedded slab source

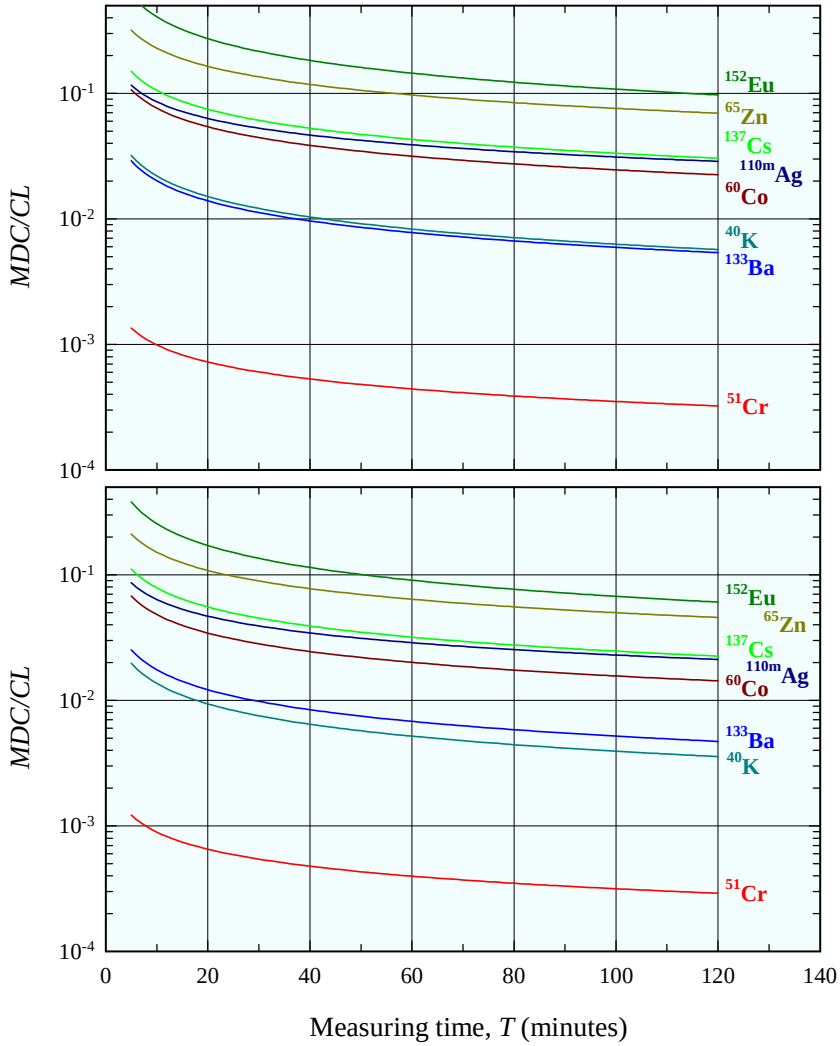

Figure 12. Ratio of the minimum detectable concentration, MDC, to the clearance level, CL, as a function of the measuring time with a germanium detector for different radionuclides distributed as a thin layer in the middle of a 10 ℓ flat concrete box (upper graph) and in the middle of a 100 ℓ flat concrete box (lower graph).

5.3 Experimental verification of MDA-calculations

The calculation of MDA has been experimentally "verified" by measuring a 25 ℓ water-filled polyethylene container in which a ^{137}Cs-activity of 9.2 kBq ± 10% is homogeneously distributed. The container was placed at a distance of 194 cm from the detector as shown in Fig. 13. The measurement was stopped after a counting time of 198 seconds when the 662 keV photo peak from ^{137}Cs just became visible on the computer screen. At this counting time, the activity in the container was taken to be the MDA for this counting time and geometry.

The container was afterwards removed and a spectrum was collected over 198 seconds. The MDA was calculated to be 8.3 kBq for the collected spectrum and the detector efficiency at 662 keV for a 25 ℓ container at a distance of 194 cm from the detector.

This calculated efficiency was used to determine the ^{137}Cs-activity in the container from the first collected spectrum. The activity was found to be 9.4 kBq ± 2.5 kBq, which indicates that the visual determined MDA is slightly overestimated.

A more “careful” visual determination would give an even better agreement with the calculated value.

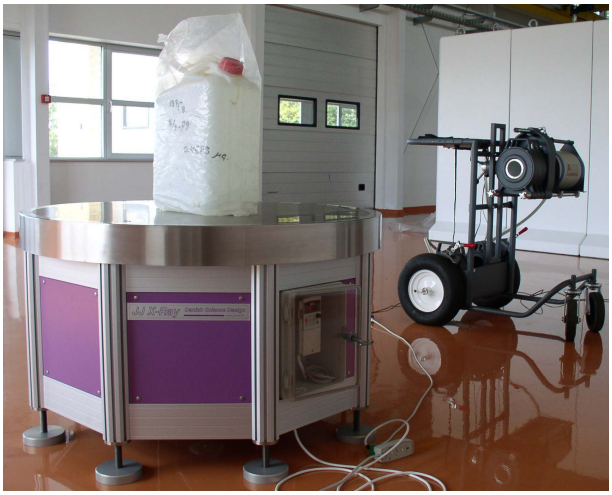

Figure 13. Measurement of MDA for a water-filled 25 ℓ polyethylene container.

6 Capacity of the laboratory

All materials that potentially can be cleared during the decommissioning of the nuclear facilities are subdivided into two categories [2, 7, 8]. One category of materials, which could not have been in contact with radioactive materials and a second category of materials, which could have been. The second category is subdivided into two classes, class I and class II. Objects and materials from class I will be measured with a high ‘measuring density’ whereas objects and materials from class II will be measured with a somewhat lower ‘measuring density’ [2].

Analyses for content of pure β-emitters, *e.g.* ^{55}Fe and ^{63}Ni, cannot be performed with the germanium detectors. The content of β-emitters will be determined from assessed activity ratios of β-emitting radionuclides to γ-emitters like ^{60}Co and ^{152}Eu and from laboratory analyses of samples taken from the objects. Methods for determining the content of pure β-emitters in concrete and steel have recently been developed [9].

6.1 Total measuring time

The necessary total time, T_{total}, for measurements on objects that potentially can be cleared is calculated from the following parameters:

- M is the estimated total amount of materials that can be cleared during decommissioning of all the nuclear facilities [10]
- f is the fraction of the materials to be measured in the laboratory (different for class I and II materials)
- m_{sub} is the mass of the objects being measured one at a time at each of the measuring setups in the clearance laboratory
- T_{sub} is the measuring time per object for each of the measuring setups
- T_{day} is the daily measuring time for both of the measuring setups
- T_{lab} is the annual number of days for which the measuring setups are in use

Intervals in which the parameters were varied are given in Table 3. These parameter ranges were used to calculate the probability distribution of the necessary number of years for clearance measurements (Eq. (14)) and the probability distribution of the annual amount of materials that can be measured in the laboratory (Eq. (15)).

Table 3. Parameter value ranges for estimating the total measuring time and the annual amount of materials that can be measured in the laboratory.

Parameter	Variation range
M	15 000 - 18 000 tonnes [10]
f	20% - 60%
m_{sub}	50 - 250 kg/object
T_{sub}	0.3 - 0.8 h/object
T_{day}	8 - 12 h/day (two setups)
T_{lab}	190 - 220 days/year

The measurement time per unit mass of materials is given as:

$$\left(\frac{T_{\mathrm{sub}}}{m_{\mathrm{sub}}}\right) \qquad [\mathrm{h/kg}] \tag{11}$$

The annual number of measurement hours in the laboratory is given as:

$$T_{\mathrm{day}} \cdot T_{\mathrm{lab}} \qquad [\mathrm{h/year}] \tag{12}$$

The total amount of materials to be measured in the laboratory is:

$$f \cdot M \qquad [\mathrm{kg}] \tag{13}$$

Consequently, the total measuring time, T_{total}, for all the materials to be measured in the laboratory can be calculated as:

$$T_{\mathrm{total}} = f \cdot M \cdot \frac{\left(\frac{T_{\mathrm{sub}}}{m_{\mathrm{sub}}}\right)}{T_{\mathrm{day}} \cdot T_{\mathrm{lab}}} \qquad [\mathrm{years}] \tag{14}$$

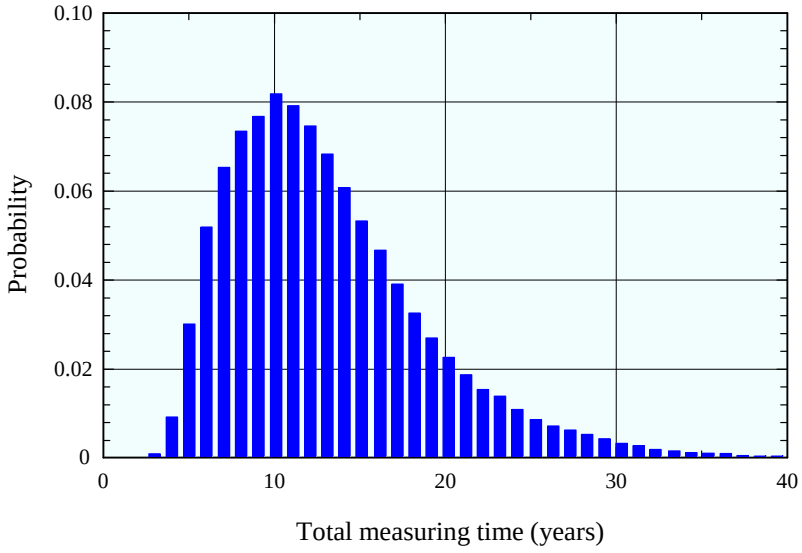

Figure 14. Calculated probability distribution of the total measuring time, T_{total}, in the Clearance Laboratory with two measuring setups.

From the value ranges given in Table 3, the probability distribution of T_{total} has been calculated for two measuring setups in the Clearance Laboratory. Latin Hypercube sampling has been used [11]. An equal probability of having a value in the value ranges has been assumed. The result is shown in Fig. 14.

The calculation shows that with two measuring setups in the laboratory there is a 5% probability that the total measuring time would be less than 6 years and a 95% probability of being less than 25 years. The mean value of the total measuring time is 13 years.

6.2 Annual amount of materials

The annual amount of materials that can be measured at the Clearance Laboratory can be calculated from:

$$m_{\text{annual}} = \frac{T_{\text{day}} \cdot T_{\text{lab}}}{\left(\frac{T_{\text{sub}}}{m_{\text{sub}}}\right)} \qquad [\text{kg/year}] \tag{15}$$

From the value ranges given in Table 3 the probability distribution of m_{annual} has been calculated in a similar way with two measuring setups in the Clearance Laboratory. The result is shown in Fig. 15.

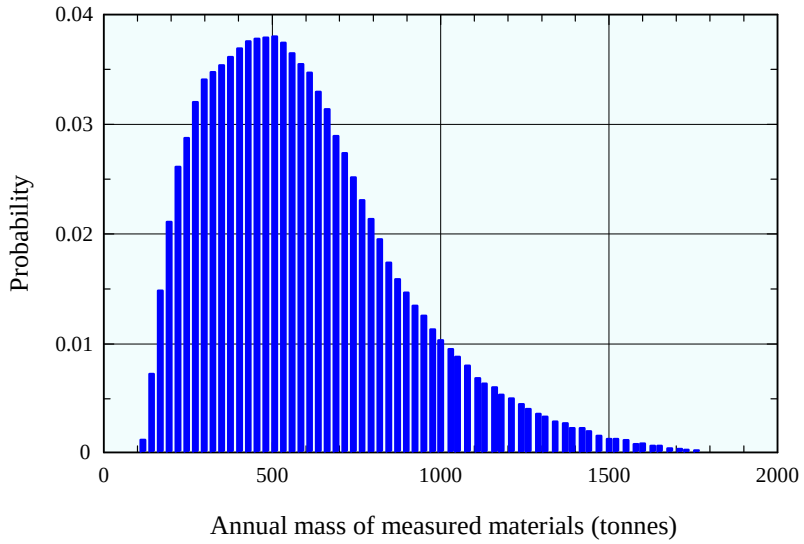

Figure 15. Calculated probability distribution of the annual amount of materials, m_{annual}, that can be measured in the laboratory with two measuring setups.

The calculation shows that there is a 5% probability that the annual amount of materials that can be measured in the laboratory would be less than 220 tonnes and a 95% probability of being less than 1,200 tonnes. The mean value of the annual amount of materials that can be measured is 600 tonnes.

If the incoming rate of materials that potentially can be cleared is too low compared with the availability of the detectors, the total measuring time, T_{total}, will not be changed but the completion of the measurements will be delayed accordingly. With an incoming rate higher than the availability of the detectors, materials and objects would have to be stored, awaiting to be measured at a later time. The total measuring time, T_{total}, will not be changed and the time for completion of all measurements will be as expected.

If the rate of clearance measurements with two measurements setups is found to be too low, the number of setups could be increased. Therefore, the laboratory has

been designed so one or two additional measuring setups can be installed without an extension of the measurement hall.

7 Conclusions

The research reactors and other nuclear facilities at the Risø-site will be decommissioned during the next 10 - 15 years. A large part of the materials and waste originating from the decommissioning can be cleared as non-radioactive waste. This has to be verified from measurements as required by the authorities. A clearance laboratory, where materials and waste can be controlled for content of radioactive materials, has therefore been built at the Risø-site.

Building materials with a low content of naturally occurring radionuclides have been used for the major structures of the laboratory and for moveable concrete shielding walls. A membrane to block radon transport from the soil into the laboratory has been used to reduce the general background radiation in the laboratory.

The laboratory has been equipped with two high-efficiency germanium detectors. Background measurements in the laboratory with unshielded and shielded detectors reveal low background levels of naturally occurring radionuclides. The only anthropogenic radionuclide detected in the background spectra is ^{137}Cs being dispersed in the environment from the Chernobyl accident in 1986 and fallout from the nuclear weapons testing in the 1950's and 1960's.

Sensitivity measurements and calculations show that the detectors can determine a small fraction of the clearance levels in larger objects within a measuring time less than 20 - 50 minutes in most cases when measurement uncertainties and the presence of several radionuclides are taken into consideration. Very large objects may need a somewhat longer measuring time.

The software system to analyse and calculate nuclide-specific values of Minimum Detectable Activity (MDA) in measured γ-spectra for specific source-detector geometries has been verified experimentally in one case and good agreement was found between automatic and visual determination of the MDA.

The capacity of the laboratory has been determined from probability calculations. The mean value of the necessary total clearance measuring time on materials and waste from decommissioning of all the nuclear facilities has been determined to be 13 years. Similarly, the mean value of the annual amount of materials that can be measured in the laboratory has been determined to be 600 tonnes.

The assumptions behind these figures include considerations on the use of the detectors for clearance measurements on buildings and land, frequent control measurements of background levels in the laboratory, check of detector calibration and an efficient infrastructure regarding transport of materials in and out of the laboratory.

It is concluded that the Clearance Laboratory can measure the foreseen amount of materials originating from decommissioning of the nuclear facilities at the Risø-site within the expected time span of 10 - 15 years. If needed, the capacity of the laboratory can be increased without any extension of the building.

References

[1] Canberra Industries, Inc. *Model S573/S574 ISOCS/LabSOCS - Validation & Verification Manual.* V4.0 (2002).

[2] Hedemann Jensen, P., Lauridsen, B., Søgaard-Hansen, J., Warming, L., *Clearance of materials, buildings and land with low content of radioactive materials. Methodology and documentation.* Risø-R-1303(DA), Risø National Laboratory (2003) (In Danish).

[3] Currie, L.A., *Limits for Qualitative Detection and Quantitative Determination.* Analytical Chemistry **40**, 586 - 593 (1968).

[4] Canberra Industries, Inc. *Genie 2000 Spectroscopy System - Operations.* V2.1 (2002).

[5] International Atomic Energy Agency. *Application of the Concepts of Exclusion, Exemption and Clearance.* Safety Guide No. RS-G-1.7, IAEA, Vienna (2004).

[6] European Commission. *Recommended radiological protection criteria for the clearance of buildings and building rubble from the dismantling of nuclear installations.* Radiation protection 113, European Communities, Luxembourg (2000).

[7] *Multiagency Radiation Survey and Site Investigation Manual (MARSSIM).* DRAFT for Public Comment. U.S. Nuclear Regulatory Commission, NUREG-1575, EPA 402-R-96-018, NTIS-PB97-117659, December 1996.

[8] *Radiological Surveys for Controlling Release of Solid Materials.* Draft Report for Comment. U.S. Nuclear Regulatory Commission, NUREG-1761, July 2002.

[9] Hou, X., Frøsig Østergaard, L., Nielsen, S.P., *Determination of ^{63}Ni and ^{55}Fe in nuclear waste sampling using radiochemical separation and liquid scintillation counting.* Analytica Chimica Acta **535**, 297 - 307 (2005).

[10] Bagger Hansen, M., Larsen, K., *Decommissioning of the nuclear facilities at Risø National Laboratory in Denmark.* In: Proceedings of an International Conference on Safe Decommissioning for Nuclear Activities: Assuring the Safe Termination of Practices Involving Radioactive Materials. Berlin, Germany, 14 - 18 October 2002, 269 - 288, IAEA, Vienna (2003).

[11] *CRYSTAL BALL - Forecasting & Risk Analysis for Spreadsheet Users.* Crystal Ball 2000 User Manual, Decisioneering, Inc., Denver, Colorado, www.decisioneering.com.

Bibliographic Data Sheet **DD-R-15(EN)**

Title and author(s)

Clearance Laboratory - Capability and measurement sensitivity

Per Hedemann Jensen, Bente Lauridsen, João Silva, Jens Søgaard-Hansen, Lisbeth Warming

ISBN

87-7666-020-6; 87-7666-021-4 (Internet)

ISSN

Dept. or group

Section of Applied Health Physics
Danish Decommissioning

Date

August 2005

Journal No.

DD-2005-470-1

Project/contract No.

Pages	Tables	Illustrations	References
19	3	15	11

Abstract (Max. 2000 char.)

A new low-level Clearance Laboratory has been built at the Risø-site. Building materials with a low content of naturally occurring radionuclides have been used. To minimize transport of radon gas from soil into the laboratory the foundation has been supplied with a membrane.

The laboratory has been equipped with two high-efficiency germanium detectors. These detectors will be used for clearance measurements on the predicted amount of 15,000 - 18,000 tonnes of non-active or nearly non-active materials, which will originate from the decommissioning of all the nuclear facilities at the Risø-site. They will be used also for clearance measurements on buildings and land.

Objects and materials to be measured for clearance are placed on a rotation table that can carry up to one tonne and can rotate once a minute to simulate some averaging of inhomogeneously distributed activity. Sensitivity and background measurements reveal that measuring times of 20 - 50 minutes would normally be sufficient to detect radionuclide concentrations of only a small fraction of the nuclide-specific clearance levels with a sufficiently low uncertainty.

Probability calculations of the measurement capacity of the Clearance Laboratory indicate that the mean value of the total measuring time for all materials that potentially can be cleared would be 13 years with a 95% probability of being less than 25 years. The mean value of the annual amount of materials that can be measured in the laboratory is 600 tonnes with a 95% probability of being less than 1,200 tonnes. If needed, there is room for additional measuring systems to increase the capacity of the laboratory.

Descriptors INIS/EDB

BACKGROUND RADIATION; CAPACITY; CLEARANCE; GAMMA SPECTROSCOPY; GE SEMICONDUCTOR DETECTORS; DECOMMISSIONING; NUCLEAR FACILITIES; RADIATION DETECTION; RISOE NATIONAL LABORATORY; SENSITIVITY; SOLID WASTES; WASTE MANAGEMENT

DANSK DEKOMMISSIONERING

Mission

Danish Decommissioning will dismantle the nuclear facilities at the Risø site and release buildings and areas to “green field” (unrestricted use) within a time frame of 11-20 years.

Vision

Danish Decommissioning will dismantle the nuclear facilities at the Risø-site at a high safety level so that employees, public and the environment are protected.

The decommissioning will be carried out in an economically effective manner within the budget and in accordance with international recommendations.

The decommissioning will be carried out in an open dialogue with the local population as well as with the society in general.

ISBN 87-7666-020-6
ISBN 87-7666-021-4 (Internet)

Danish Decommissioning
Post Box 320
4000 Roskilde
Telephone 4677 4300
dd@dekom.dk
Fax 4677 4302
Website www.dekom.dk