Localization and depth determination of radionuclide contamination in concrete at a decommissioning nuclear power plant

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Localization and depth determination of radionuclide contamination in concrete at a decommissioning nuclear power plant

Performed at:

Medical Radiation Physics (LU)
Malmö Hospital

Barsebäck NPP

Localization and depth determination of radionuclide contamination in concrete at a decommissioning nuclear power plant

Background & Aim → Materials → Results: Method 1 → Results: Method 2 → Results: Method 3 → Conclusions

Background: Barsebäck NPP

- Boiling water reactors Barsebäck 1 (1975), Barsebäck 2 (1977)
- Shutdown: B1 in 1999, B2 in 2005
- Decommissioning and dismantling is in progress
- Presence of radionuclides
  - Contamination from *i.e.* process water
- Large concrete structures to be cleared

NPP = Nuclear power plant

Clearence process

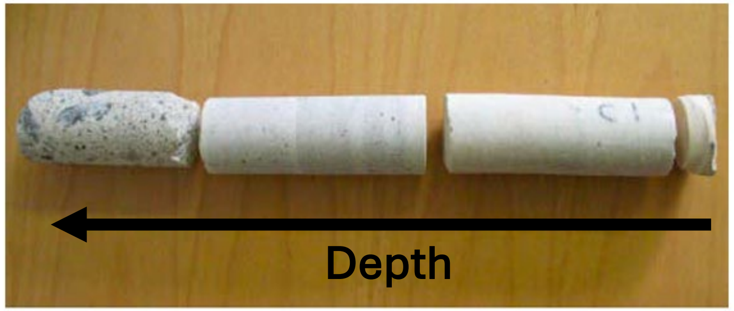
- Regulated by Swedish Radiation Safety Authority (SSMFS 2018:3)
- Clearence levels set by SSM for the ETM radionuclides are:
  - Cs-137: 100 kBq/m^2
  - Co-60: 10 kBq/m^2
- Contamination under the surface should be projected to the surface
- Clearence process: Assessment → Measurement → Decontamination → Approval

SSMFS 2018:3 = Strålsäkerhetsmyndighetens föreskrift om friklassning av material byggnadsstrukturer och områden

ETM = Easy-to-measure

Problem formulation

- Radionuclide contamination may extend below the surface
- Today's method: core sampling → time-consuming



Concrete core (SKB)

- Need for a more effective method for depth determination
- **Can gamma spectrometry directly on the surface be used to estimate the depth?**

Aim

- Evaluate gamma spectrometric methods for depth determination
- Focus on the ETM radionuclides Co-60 and Cs-137
- Test and evaluate three methods

1. Compton-to-peak ratio

    Based on *in situ* gamma spectrometry at Barsebäck NPP

2. Least squares spectrum fitting

    Based on laboratory measurements with point sources at LU

3. Core sampling (as reference)

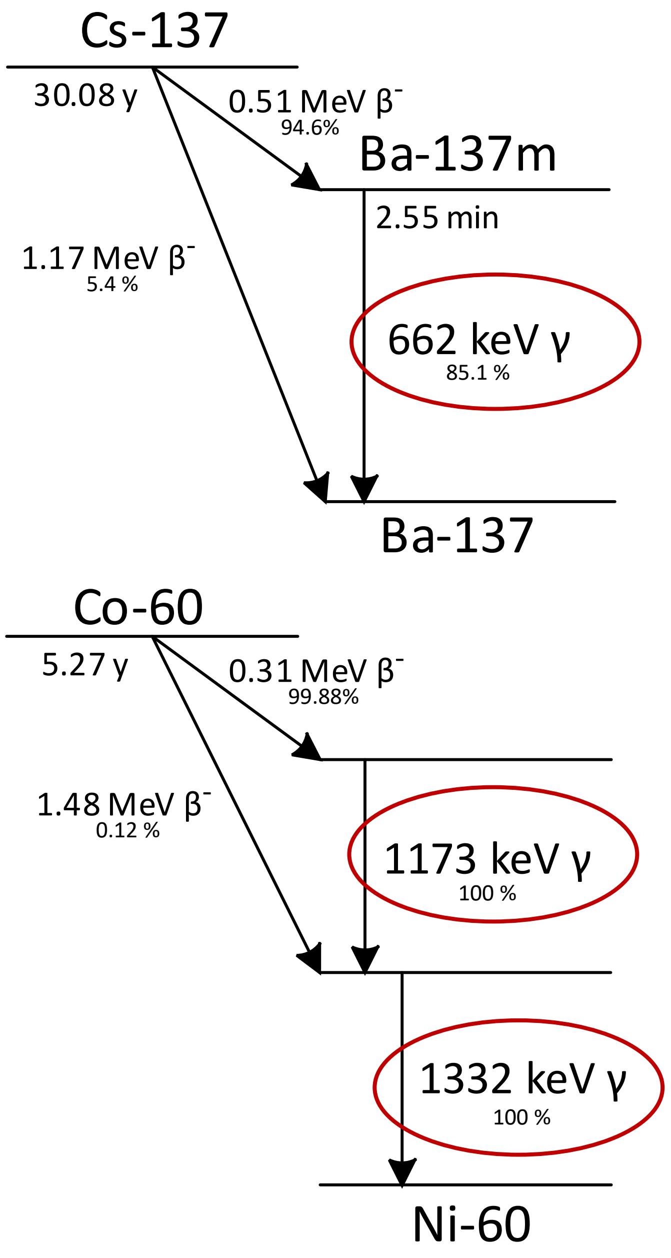
    Based on *ex situ* gamma spectrometry at Barsebäck NPP laboratory

In situ = On site

Ex situ = In laboratory

Radionuclides and detectors

- Cs-137: 662 keV gamma
- Co-60: 1173 and 1332 keV gammas
- Detectors used:
  - NaI(Tl): Low energy resolution
  - HPGe: High resolution

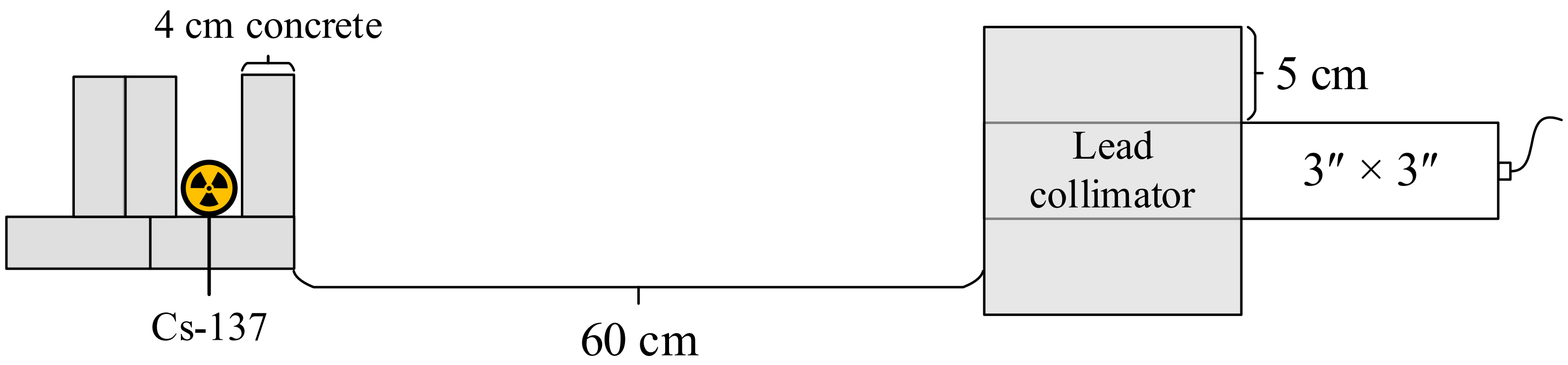


NaI(Tl) = Sodium Iodine, Thallium doped

HPGe = High purity Germanium

Laboratory measurements at LU

- Point sources (Cs-137, Co-60) placed behind concrete bricks
- Measurements with NaI(Tl) and HPGe, lead collimated
- Varied shielding thickness simulated different depths

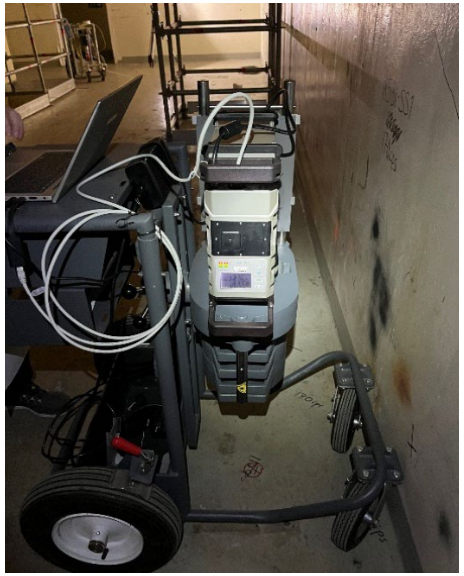


LU = Lund University (Medical Radiaiton Physics, Malmö)

In situ measurements at Barsebäck NPP

- Site: Room 1R5.31
- Contaminated spots were identified using pulse-rate scanning
- *In situ* gamma spectrometry with NaI(Tl) and HPGe, lead collimated
- Two lead collimators used: 30º and 90º opening angle

NaI(Tl)



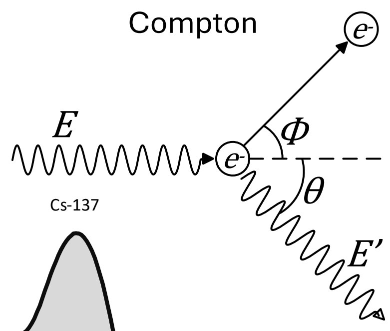
HPGe

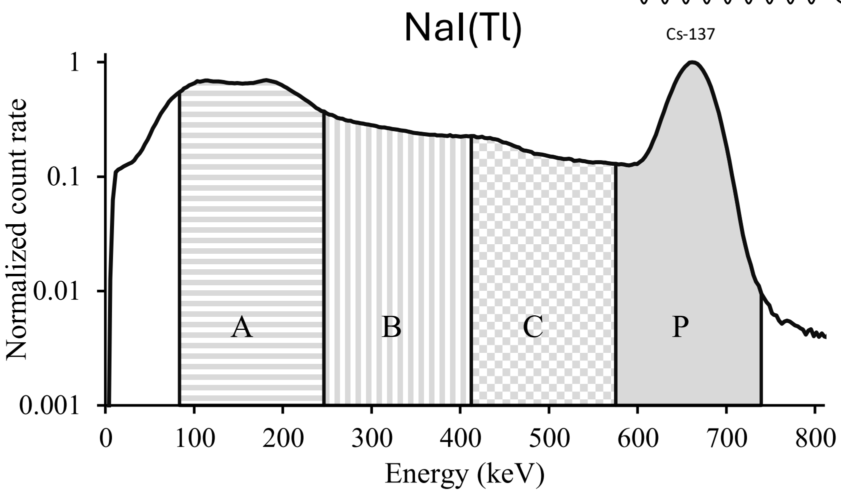
1. Compton-to-peak ratio (CPR)

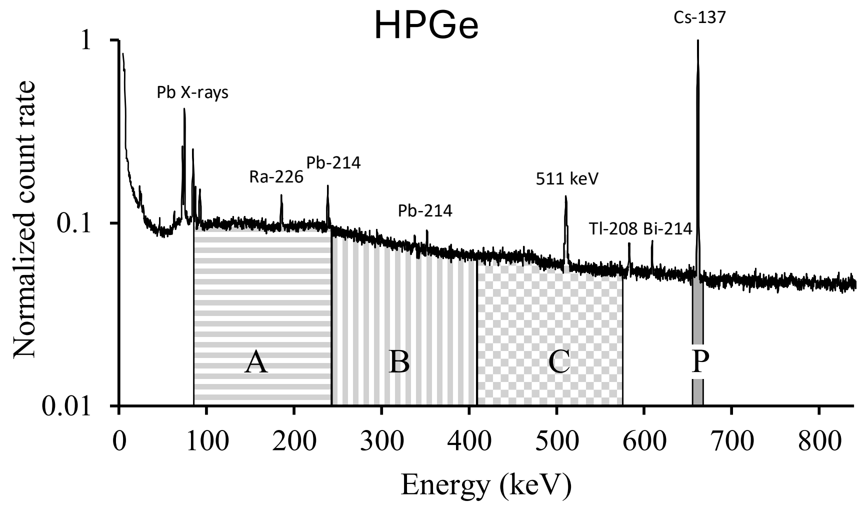
Method

- Compton can be used for shielding determination (depth)
- Gamma spectrum divided into regions of interest (ROI)
- For Cs-137: A, B, C and P

$$C/P = \frac{\sum Count\ rate\ in\ ROI\ C}{\sum Count\ rate\ in\ ROI\ P}$$





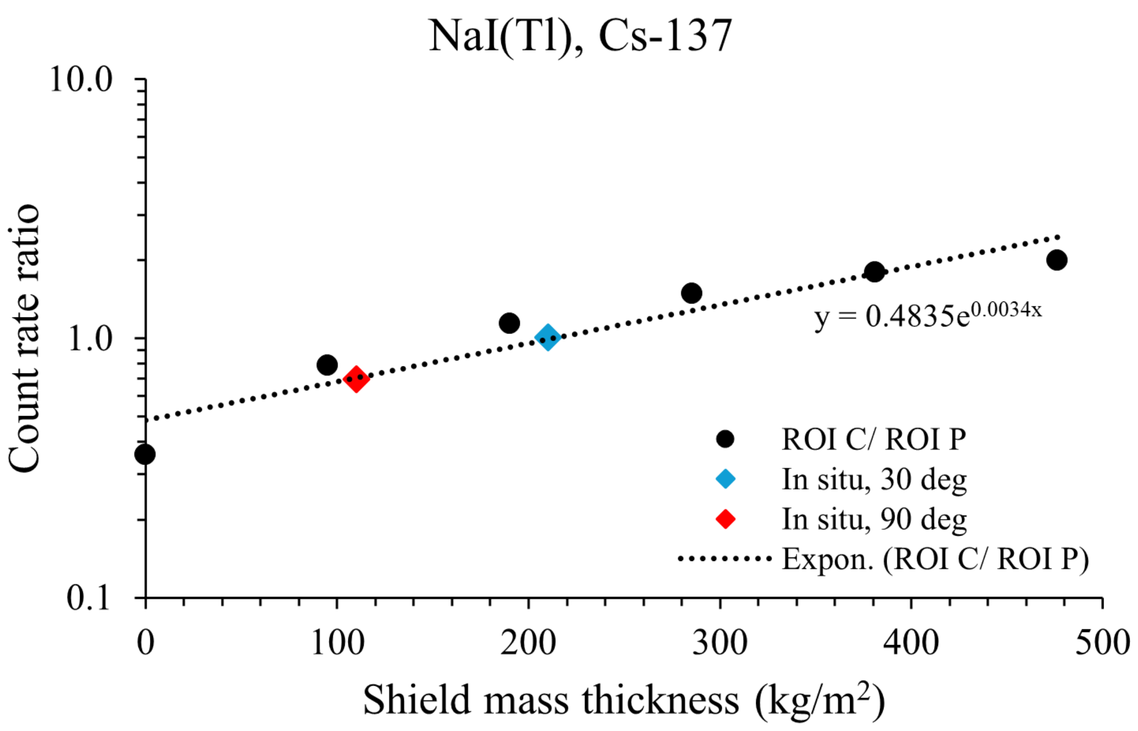


1. Compton-to-peak ratio (CPR)

Results

- CPR follows an exponential increase with shielding (black dots)

- Calculated CPR values for NPP *in situ* measurements with 30º (blue) and 90º (red)

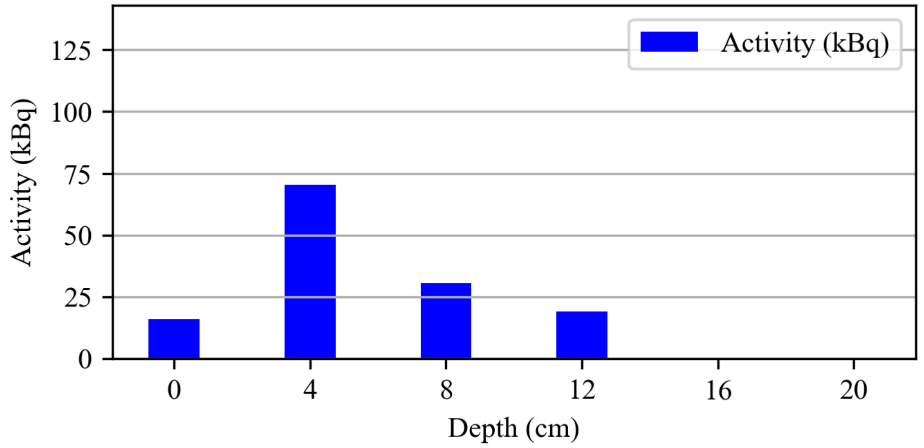


2. Least squares fitting
Method and results

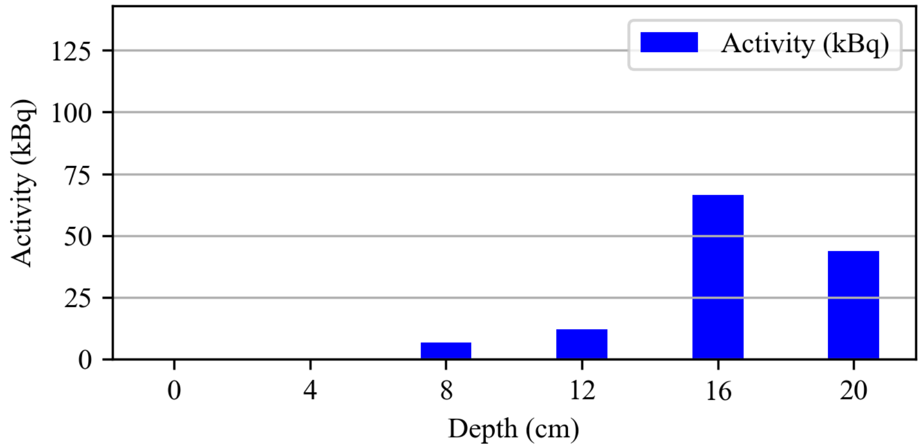
- The second method tested
- Spectrum fitting

  using weighted sum of reference spectra from laboratory measurements
- Estimates depth distribution of activity

NaI(Tl), 130 kBq Cs-137, 4 cm



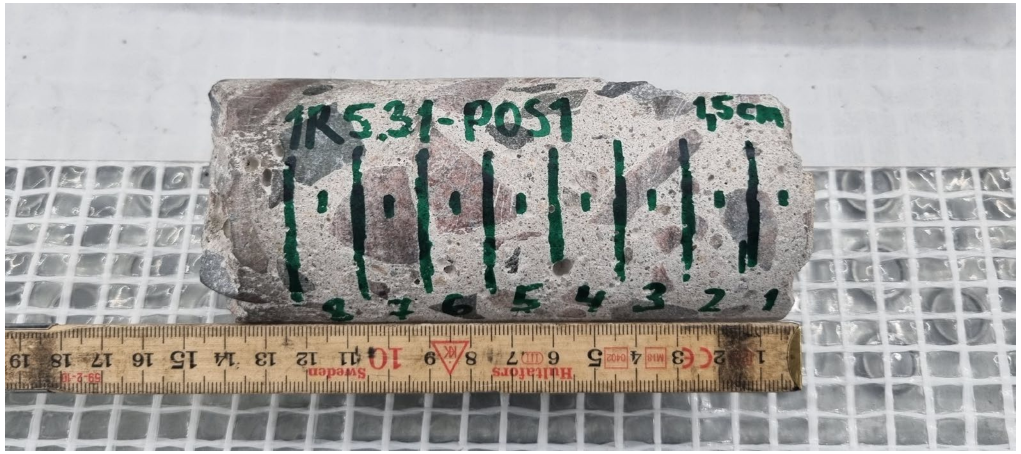
NaI(Tl), 130 kBq Cs-137, 16 cm



3. Concrete core sampling
Method

- Two samples were successfully taken, in room 1R5.31
- Cut into ~1.5 cm slices
- Measured *ex situ* with HPGe in lead-cave
- Results in a reference activity-depth profile

Ex situ = In laboratory



Concrete core from contaminated site

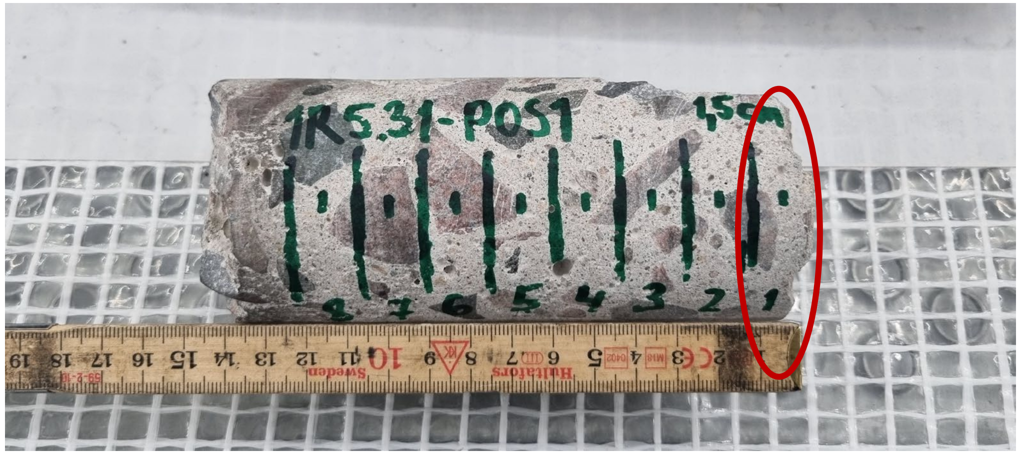
3. Concrete core sampling

Results

- More than 95% of the activity was in the first slice,

  With a sharp decrease with depth

- Further investigation → the activity is in the first millimeters of the first slice



Concrete core from contaminated site

Discussion and conclusions

- Three methods were compared
- Each method have different advantages
- In situ gamma spectrometry could be used for estimating contamination depth

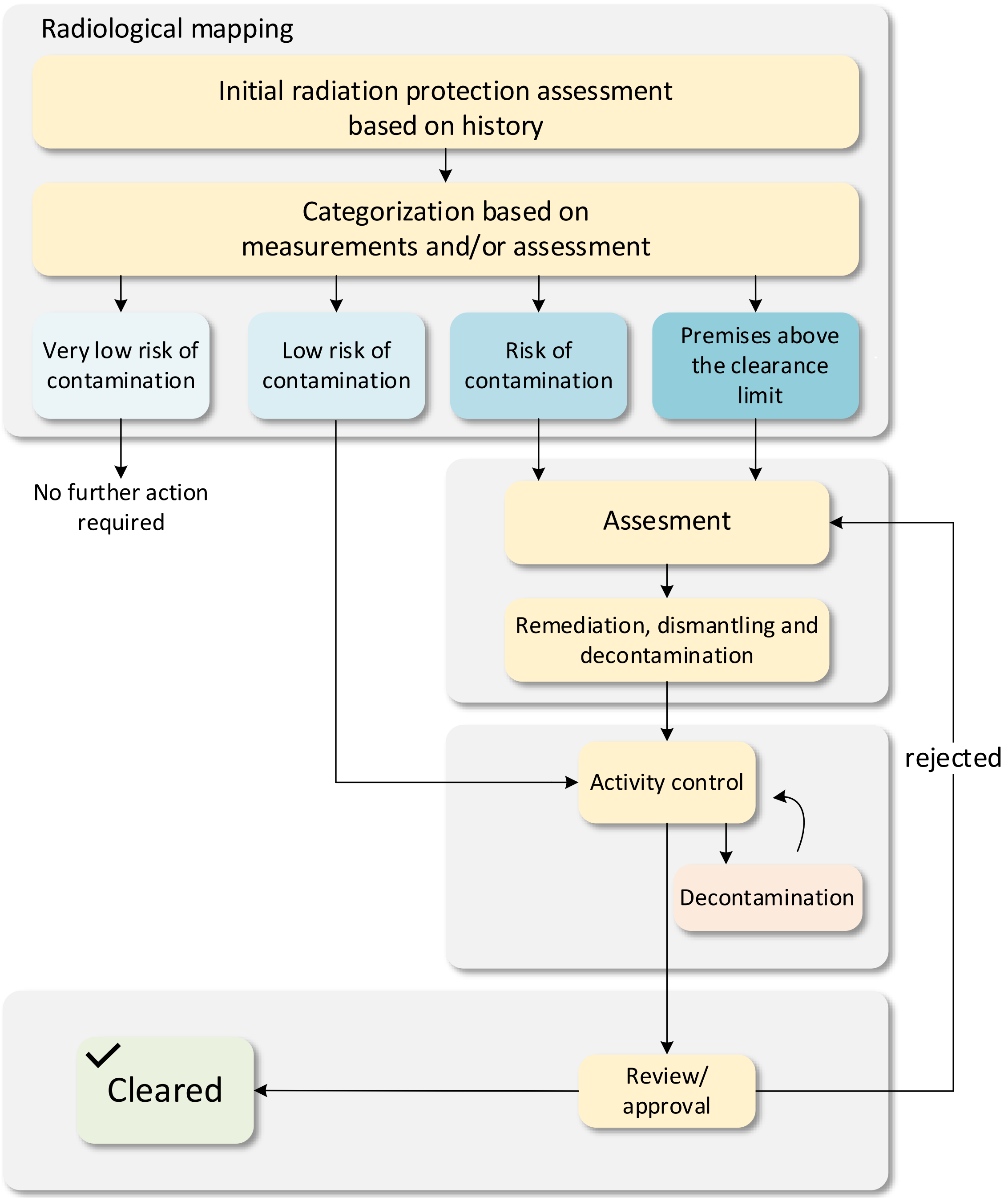
  Typical or abnormal distributions
- However, further measurements are needed, to validate the method
  - Using the same detector for calibration measurements and *in situ* measurements

Thank you for listening!

Special thanks to Medical Radiation Physics Malmö, and the people at Barsebäck NPP

Extra slides

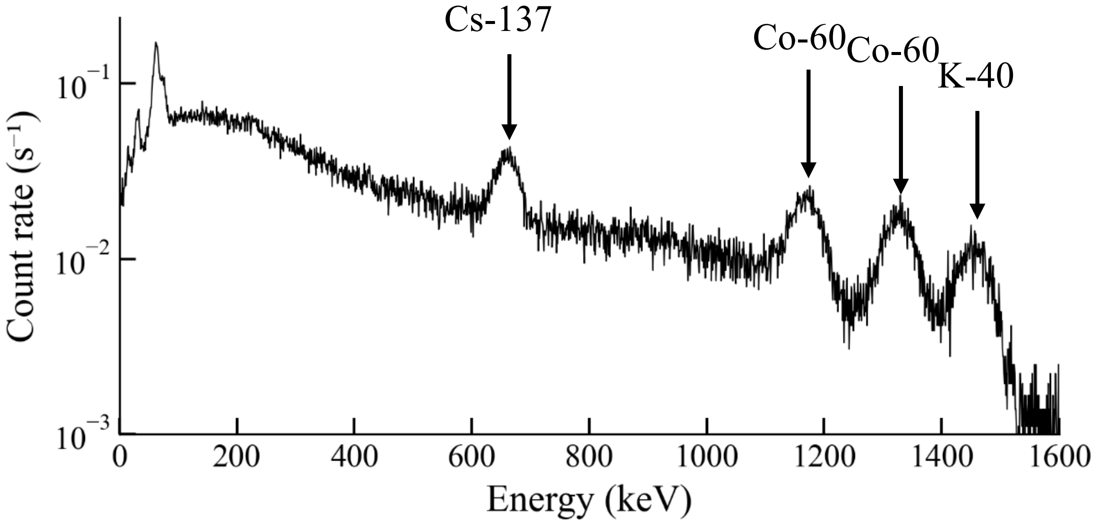
Clearence process



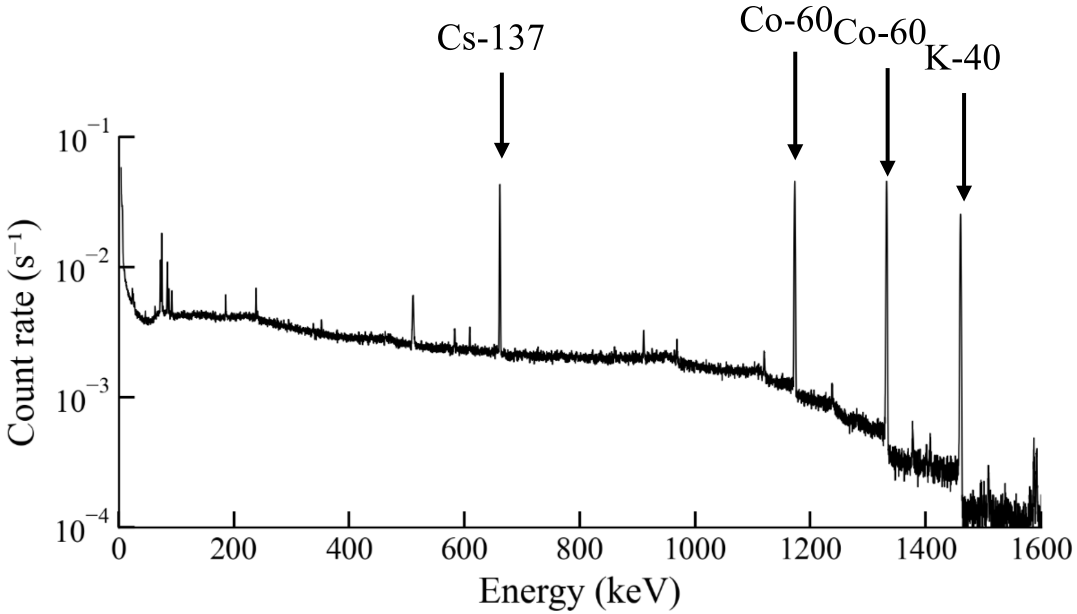
In situ spectra

- Barsebäck NPP in situ spectra

NaI(Tl), pos 1, LT 243223 s (~67.7 h)

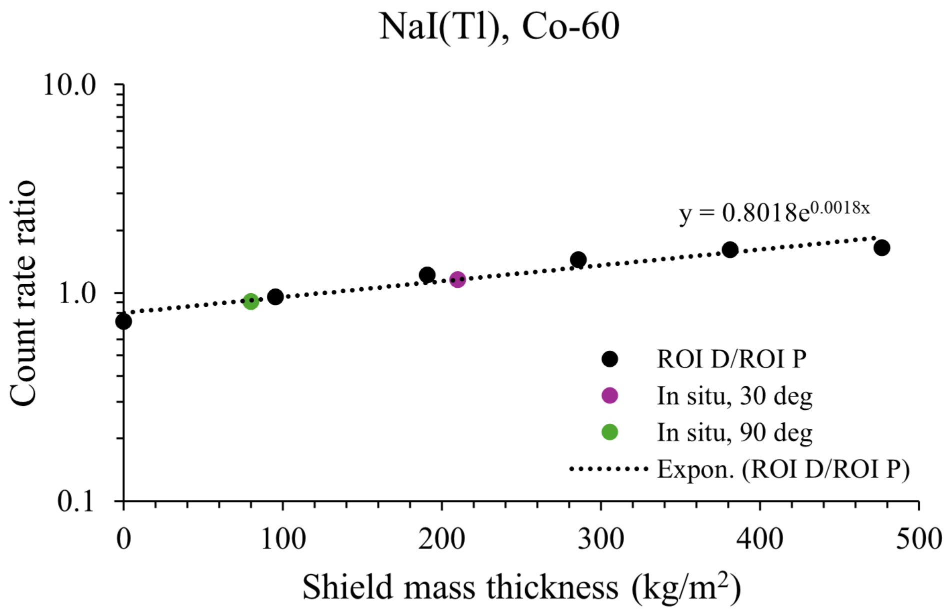


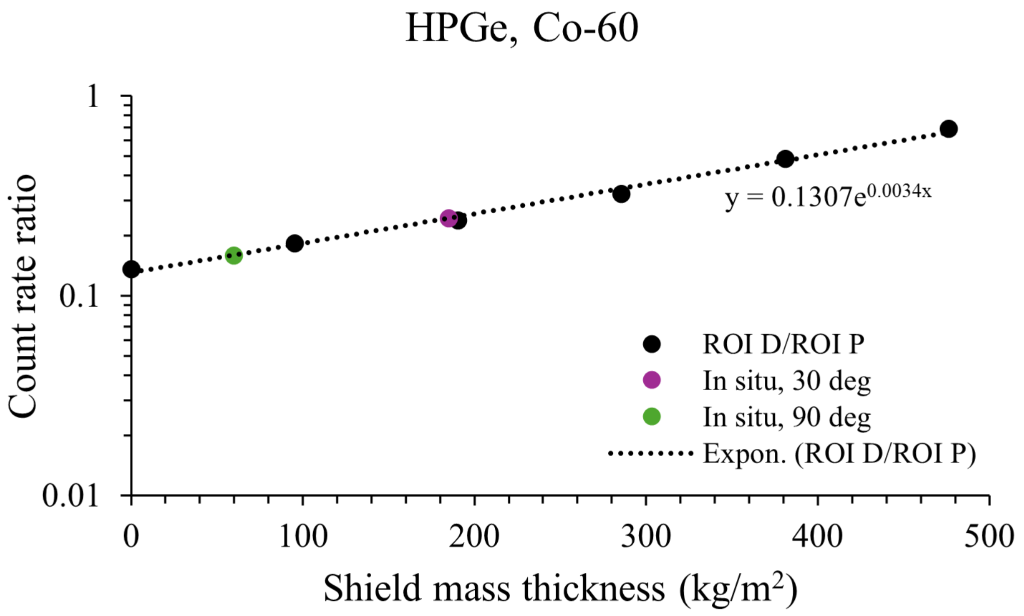
HPGe, pos 1, LT 3600 s



PCR Co-60 results

- Co-results





CPR In situ Barsebäck NPP

Table 7. Compton-to-peak count rate ratio for in situ measurements at Barsebäck NPP.

Detector	Collimator opening	Radionuclide	Count rate ratio	Uncertainty (%)
NaI(Tl)	30º	Cs-137	1.01	1.1
		Co-60	1.16	1.7
NaI(Tl)	90º	Cs-137	0.695	0.8
		Co-60	0.912	0.8
HPGe	30º	Cs-137	0.068	0.5
		Co-60	0.245	0.3
HPGe	90º	Cs-137	0.049	2.7
		Co-60	0.160	1.3

Least squares fitting method

- From thesis

The observed measurement, $S_{obs}(E)$ represent a gamma spectrum over various energies E originating from a radioactive contamination from Cs-137 distributed over various depths i within concrete and can be described as in Bu *et al.* (2021).

$$S_{obs}(E) = \sum_{i=1}^{N} A_i \cdot S_{Cs,i}(E) \qquad (23)$$

Where A_i is the weighting factor for the calibration measurements $S_{Cs,i}$ at different depths i. For all energy channels M in the desired energy interval the matrix is

$$X = \begin{bmatrix} S_{Cs,1}(E_1) & S_{Cs,2}(E_1) & \cdots & S_{Cs,N}(E_1) \\ S_{Cs,i}(E_2) & S_{Cs,2}(E_2) & \cdots & S_{Cs,N}(E_2) \\ \vdots & \vdots & \ddots & \vdots \\ S_{Cs,i}(E_M) & S_{Cs,2}(E_M) & \cdots & S_{Cs,N}(E_M) \end{bmatrix} \qquad (24)$$

The goal is to minimize the difference between the calculated spectrum and the observed measurement. In other words, to minimize

$$\|X \cdot A - S_{obs}\|^2 \qquad (25)$$

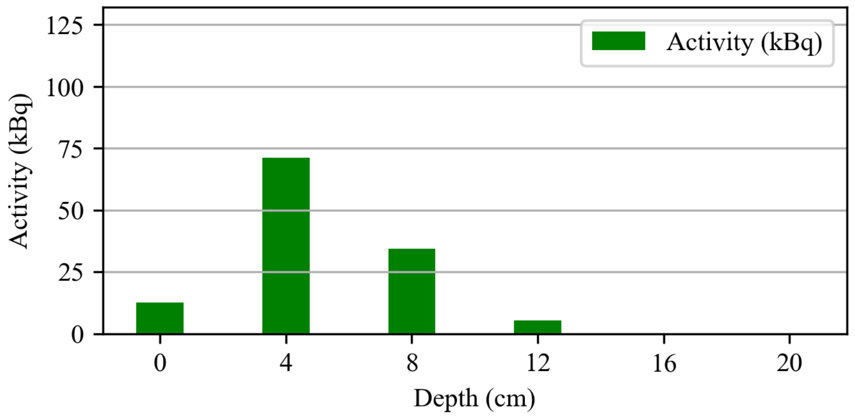
To solve Eq. 14, non-negative least squares are used, *i.e.* to not have negative activities $A_i > 0$. Also, the residuals R are analyzed, and calculated as

$$R = S_{obs} - X \cdot A \qquad (26)$$

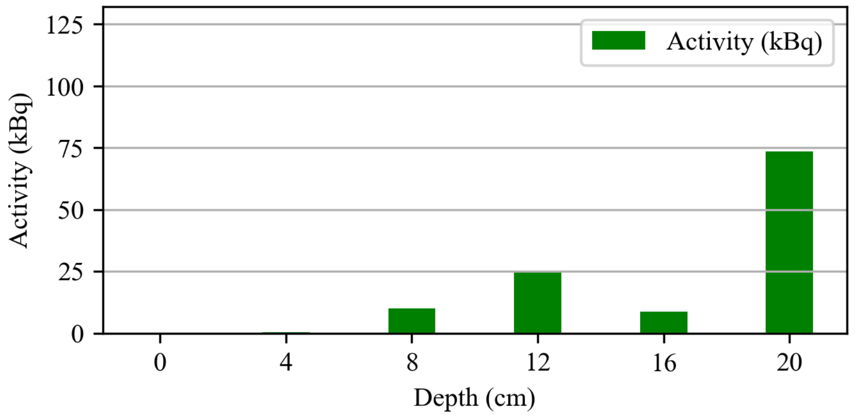
Least squares Co-60 results

- Co-results

NaI(Tl), Co-60, 4 cm



NaI(Tl), Co-60, 16 cm



Pulse rate measurement at Barsebäck NPP

- The

Table 8. Pulse rates (β) at three positions at Barsebäck NPP Unit 1 (1R5.31).

Position no.	**Pulse rate** (CPS) $\pm$ σ
1	60.8 $\pm$ 3.4
2	77.2 $\pm$ 3.3
3	51.0 $\pm$ 4.9

Concrete core sampling results

Table 9. Gamma spectrometric measurements and characteristics of the concrete core samples from positions 1 and 2. Depth is the top depth of each slice; the cutting blade was approximately 0.3 cm wide which created a gap of approximately 0.3 cm between each slice, slice L is the width of each slice

Slice no.	Pos. no.	LT (s)	Mass (g)	Cs-137 (Bq)	Co-60 (Bq)	Intact surface (%)	Depth (cm)	Slice L (cm)	Cs-137 (kBq/m^2)	Co-60 (kBq/m^2)
1	1	1200	50.4	185±2.9	122±2.2	60 %	0	1.0	99.0	65.0
2	1	3600	76.2	1.48	3.12	85 %	1.5	1.25	0.56	1.18
3	1	10800	77.5	0.26	0.32	100 %	3	1.35	0.08	0.10
4	1	10800	87.8	0.20	0.32	100 %	4.5	1.3	0.07	0.10
5	1	10800	77.5	0.19	0.26	100 %	6	1.1	0.06	0.08
1	2	1800	112	187±2.4	64.9±1.6	100 %	0	1.5	60.0	20.8
2	2	3600	75.8	0.30	0.60	100 %	1.5	1.0	0.10	0.19
3	2	7200	62.0	0.20	0.33	100 %	3	0.8	0.06	0.11
4	2	7200	123	0.21	0.36	100 %	4.5	2.0	0.07	0.11

Concrete core measurements

- The first slice from position 1 and 2 was measured with floor-side towards the detector and away from the detector

Table 10. Activity ratios for samples measured in orientation: Down - the sample being faced with floor-side down towards the detector; Up - the sample being faced with floor-side up, away from the detector. Measurements were made for two different distances between the detector and the sample.

Slice no.	Pos. no.	Det. Dist. (cm)	Orient.	Cs-137 (Bq)	Up/Down Ratio	Co-60 (Bq)	Up/Down Ratio
1	2	0	Up	67	36%	34	53%
1	2	0	Down	187		65	
1	2	10	Up	76	71%	37	61%
1	2	10	Down	107		60	

ISOCS/LabSOCS results

Table 11. Surface activity estimates based on ISOCS/LabSOCS simulation model. Assuming that the activity is evenly distributed over the first 5 cm, based *in situ* measurements on position 1.

Detector	**Collimator opening**	**Radionuclide**	**MDA** (kBq/m2)	**Activity** (kBq/m2)	**Uncertainty** (kBq/m2)
NaI(Tl)	30º	Cs-137	95.2	820	55.6
		Co-60	38.3	489	17.2
NaI(Tl)	90º	Cs-137	2.80	99.6	6.06
		Co-60	1.66	75.2	2.20
HPGe	30º	Cs-137	2.12	274	16.5
		Co-60	1.03	279	7.93
HPGe	90º	Cs-137	0.89	84.2	5.11
		Co-60	0.67	62.3	1.83

ISOCS/LabSOCS results

30 deg 90 deg

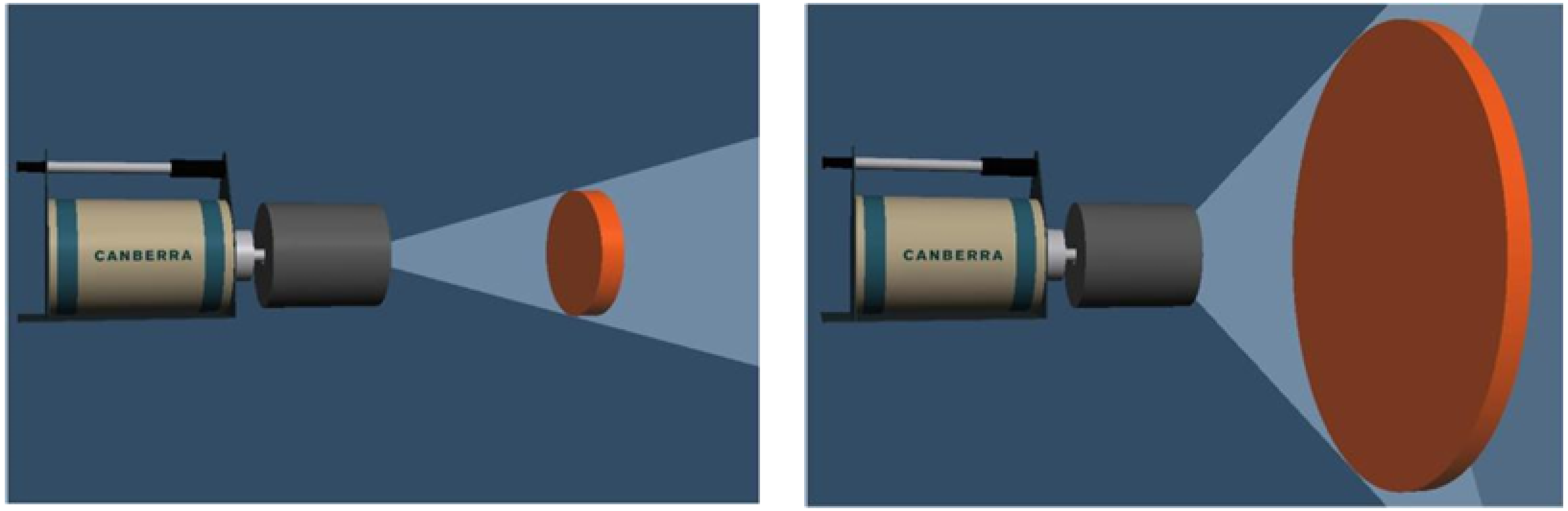
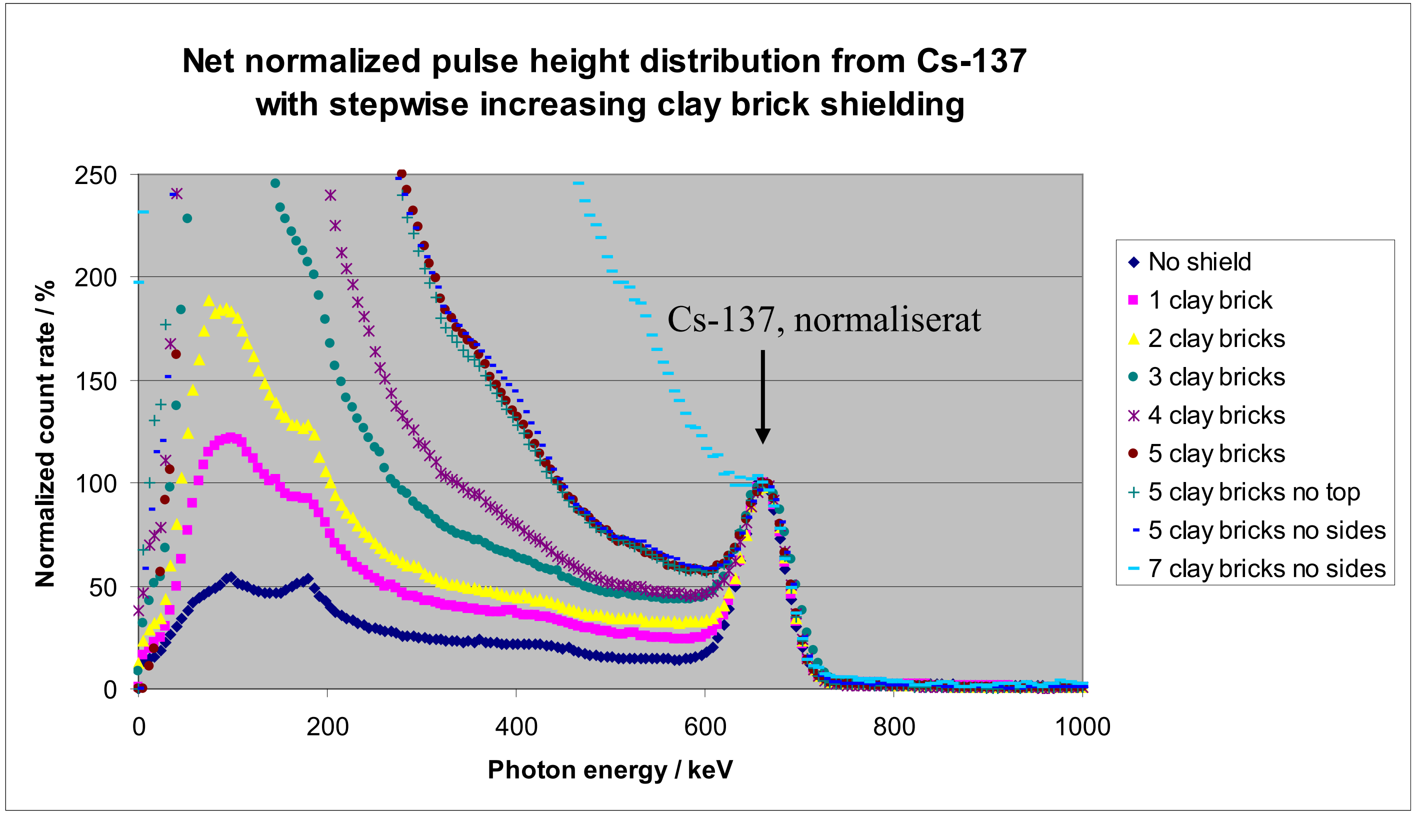


Figure 11. ISOCS/LabSOCS calculation for in situ measurements at Barsebäck NPP with 30º collimator opening (left) and 90º collimator opening (right). For this calculation, the activity was assumed to be evenly distributed in the first 5 cm of concrete.

Effekten av skärmning på gammaspektrum

- Comptonbidraget ökar jämfört med primärtoppen



Mätningar med olika mycket skärmning framför en strålkälla, Cs-137 (R Finck)