Comparison between measurement methods for the characterisation of radioactively contaminated land

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PRESENTATION OUTLINE

1. **Introduction and objectives**

2. **Intro to case study site (Dounreay)**

3. **Survey of radioactively contaminated land**
   - **Estimation of measurement uncertainty**
   - **Fitness-for-purpose of measurements (averaging area)**
   - **Fitness-for-purpose of measurements (hotspot identification)**

4. **Further work - optimisation of *in situ* investigations**

5. **Conclusions**

Chemically contaminated land

***In situ* sampling mass < *Ex situ* sampling mass**

e.g. PXRF (~1g) compared with laboratory measurements (~1kg).

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Radioactively contaminated land:

***In situ* sampling mass > *Ex situ* sampling mass**
Due to remote detection of penetrating radiation

e.g. field gamma measurements (50 kg – >100 tonnes!) compared with laboratory measurements (~1kg).

Ex situ **laboratory analysis:**

- **very expensive, up to £1000 / sample for alpha radiation.**
  - → **low-resolution mapping.**
- **considered best practice.**

INTRODUCTION (Radioactively Contaminated Land)

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  → allows high resolution mapping.
- **currently poorly quantified.**

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Requirement to demonstrate the fitness-for-purpose of *in situ* measurement techniques.

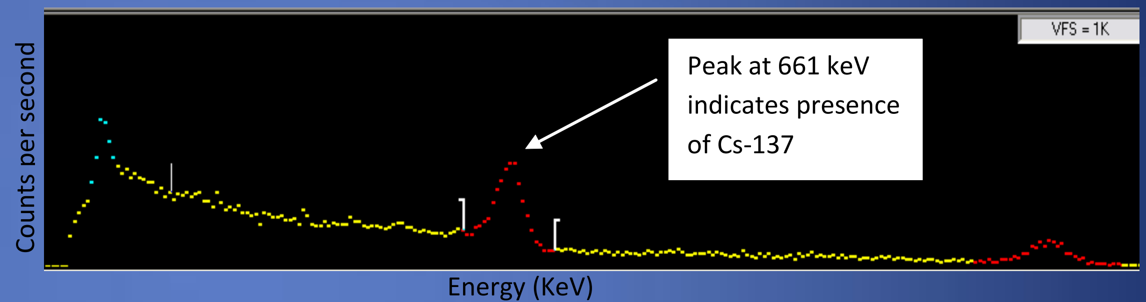
Fitness-for-purpose (FFP) *"the property of data produced by a measurement process that enables a user of the data to make technically correct decisions for a stated purpose." (Thompson & Ramsey, 1995).*

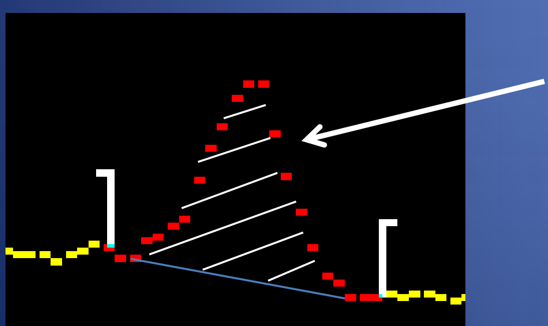
INTRODUCTION - MEASURING RADIOACTIVITY

Many radionuclides emit alpha or gamma radiation at one or more characteristic energy levels (e.g. ^{137}Cs daughter ^{137m}Ba at 661 keV).

Counts collected across a spectrum of energy levels.

Peak energy levels indicate existence of specific radionuclides.



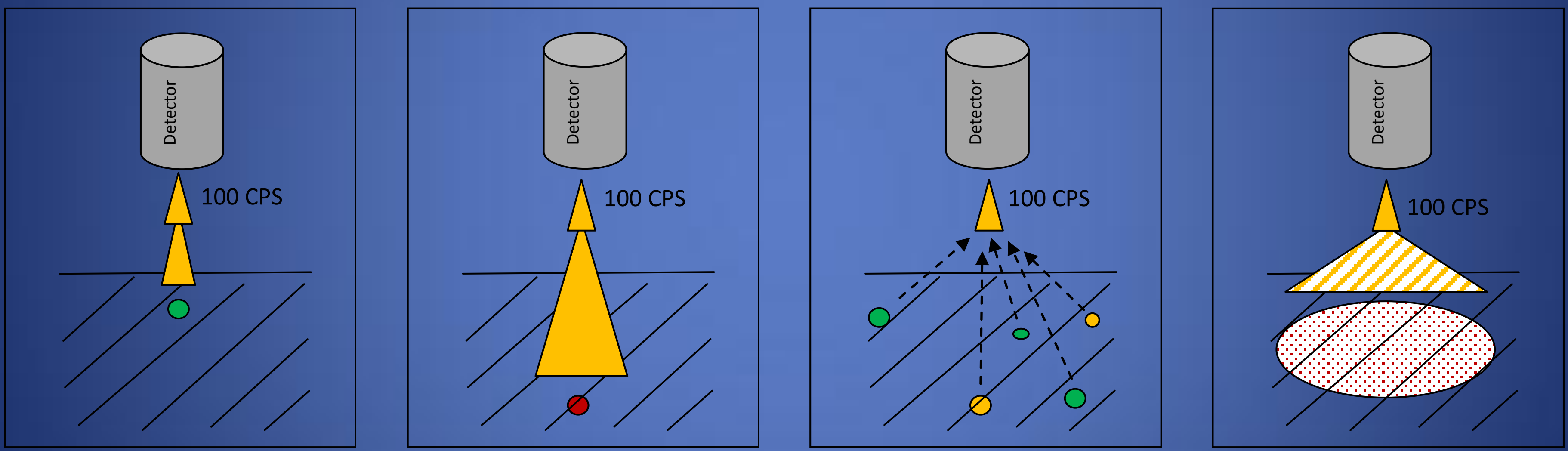


Peak area indicates intensity of radiation (counts per second, or CPS) received at detector.

No direct info re: amount of material sampled or its activity level. Interpretation required to convert CPS to activity concentration.

INTRODUCTION - *IN SITU* MEASUREMENT EXAMPLE

Example - Using a detector positioned above the ground surface, a gamma count of 100 CPS (detected decay photons) is recorded. This is used to estimate the activity of the source in Becquerels (Bq), based on the detector efficiency and emission probability) .



Low activity particle close to detector	**High activity particle distant from detector**	**Multiple low/medium activity particles**	**Volume of diffuse activity**

***In situ* methods may not be able to determine which of the above applies.**
Different for *ex situ,* where a soil sample is excavated and analysed in lab.

PRESENTATION OBJECTIVES

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  - **Estimating spatial distributions and mean activity concentration of contamination over the survey area;**
  - **Identification of hotspots of activity.**

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- **Introduce ongoing work being carried out by Sussex University in conjunction with Dounreay Site Restoration Ltd, with the general aim of optimising *in situ* investigations of radioactively contaminated land in order to achieve measurements which are Fit-for-Purpose (FFP).**

CASE STUDY SITE - DOUNREAY

Dounreay – the case study site

Dounreay - Britain's experimental fast breeder reactors.

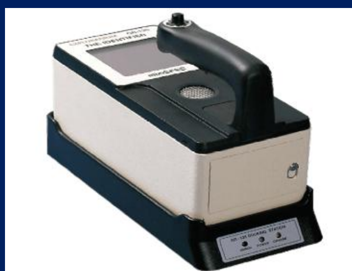
Reactors in operation from 1958-1994. Site now being decommissioned.

Characterisation of land areas and concrete floors of buildings for radionuclide content.

SURVEY METHODS

Canberra NaI 3”x3” scintillation detector with 90 degree collimation.

Type: *In situ*
Height: ~250mm
Count time: 10mins
Sample mass: Up to 30 tonnes!
Output: Activity concentration (Bq g^{-1})



Hand-held **Exploranium** 2”x2” NaI scintillation detector (un-collimated), placed on ground surface.

Type *In situ*
Height: ~25mm
Count time: 10mins
Sample mass: ~ 3 tonnes.
Output: Activity (CPS)

Groundhog vehicle. Wide array of gamma detectors. Permits 100% coverage.

Type: *In situ*
Height: ~250mm
Count time: ~ 1 second
Sample mass: Greater than Canberra ?
Output: Activity (CPS)

Ex situ top-soil samples using bulb planter to 10cm depth.

Type: *Ex situ* / Laboratory measurements
Height: -10mm
Count time: 12 hours in laboratory
Sample mass: ~ 0.5kg
Output: Activity concentration (Bq g^{-1})

Survey Objectives

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1. Estimate uncertainty in the measurements of Cs-137 contamination in soil, using the Canberra and Exploranium *in situ* detectors and one *ex situ* method.

2. Evaluate fitness-for-purpose of the 3 measurement sets for

- **a: The characterisation of the intensity of radioactive contamination over an averaging area.**
- **b: The identification of hotspots of radioactive activity.**

- *'An estimate attached to a test result which characterises the range of values within which the true value is asserted to lie'* (ISO, 1993)

  - Partially generated by measurement error from the sampling and sampling prep as well as the analytical process.

  - Sampling uncertainty arises due to the heterogeneous nature of the contaminant distribution in soil, and ambiguity in the sampling protocol.

SURVEYS PERFORMED BY UNIVERSITY OF SUSSEX

Zone 12

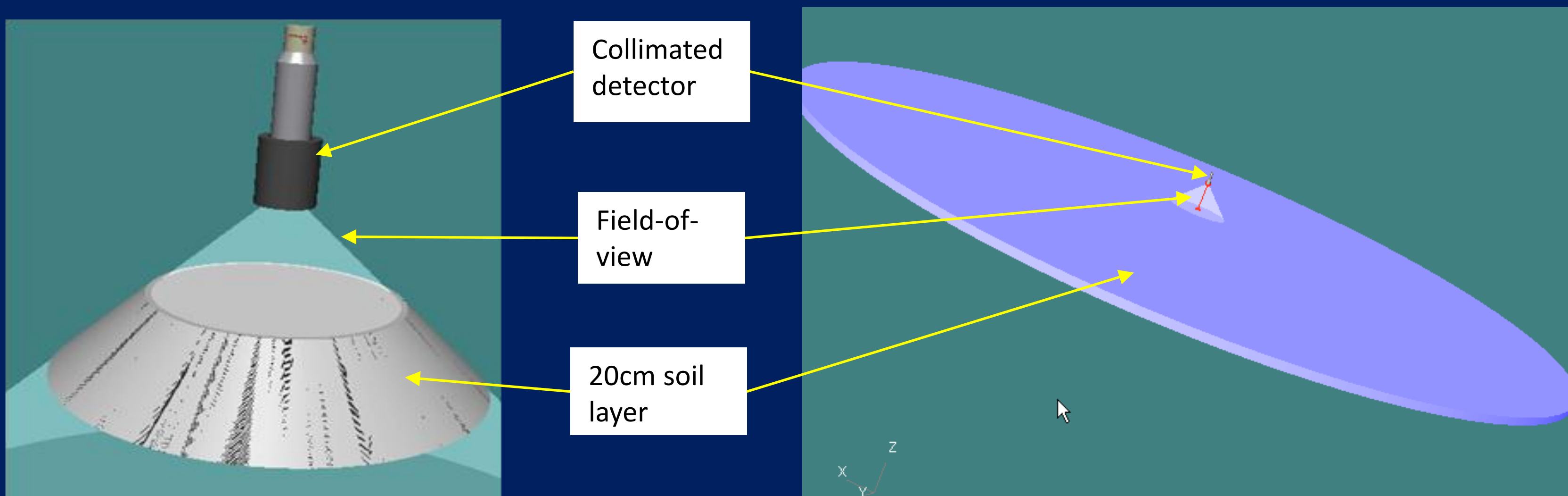
Area: Rectangular 20m x 14m = 280m^2
Measurement spacing: 2m
***In situ* detector 1: Canberra 3X3” NaI, 90^o 20mm lead collimator**
In situ detector 1 height: nominally 280mm
Coverage: 6.2%
***In situ* detector 2: Exploranium GR135**
In situ detector 2 height: 0mm
In situ counting time: 600 seconds both detectors
No. *in situ* measurement locations: 88
No. *in situ* duplicate measurement locations: 9
No. *ex situ* soil samples: 20 x 0-10cm, 8 x 10-20cm
No. *ex situ* duplicate locations: 8
Sample duplicate spacing: 20cm

Barrier 31

Area: Irregular 206m^2
Measurement spacing: 1.3m
***In situ* detector: Canberra 3X3” NaI, 90^o 20mm lead collimator**
In situ detector height: nominally 920mm
Coverage: 100% of ground covered
In situ counting time: 600 seconds
No. *in situ* measurement locations: 122
No. *in situ* duplicate measurement locations: 12
No. *ex situ* soil samples: 20 x 0-10cm, 20 x 10-20cm
No. *ex situ* duplicate locations: 8
Sample duplicate spacing: 13cm

IN SITU CONVERSION OF CPS TO Bq g^{-1}

Canberra *in situ* spectra interpreted using modelling software (ISOCS).



Conversion from Bq to Bq g^{-1} using a cone-shaped modelled soil volume of 10cm depth, defined by the theoretical field-of-view (FOV) of the 90° collimator. Large (150%) bias compared to *ex situ* measurements.

Changing the modelled soil volume to a disk shape larger than the FOV substantially reduced activity concentrations, suggesting some of the observed bias was caused by the model initially used.

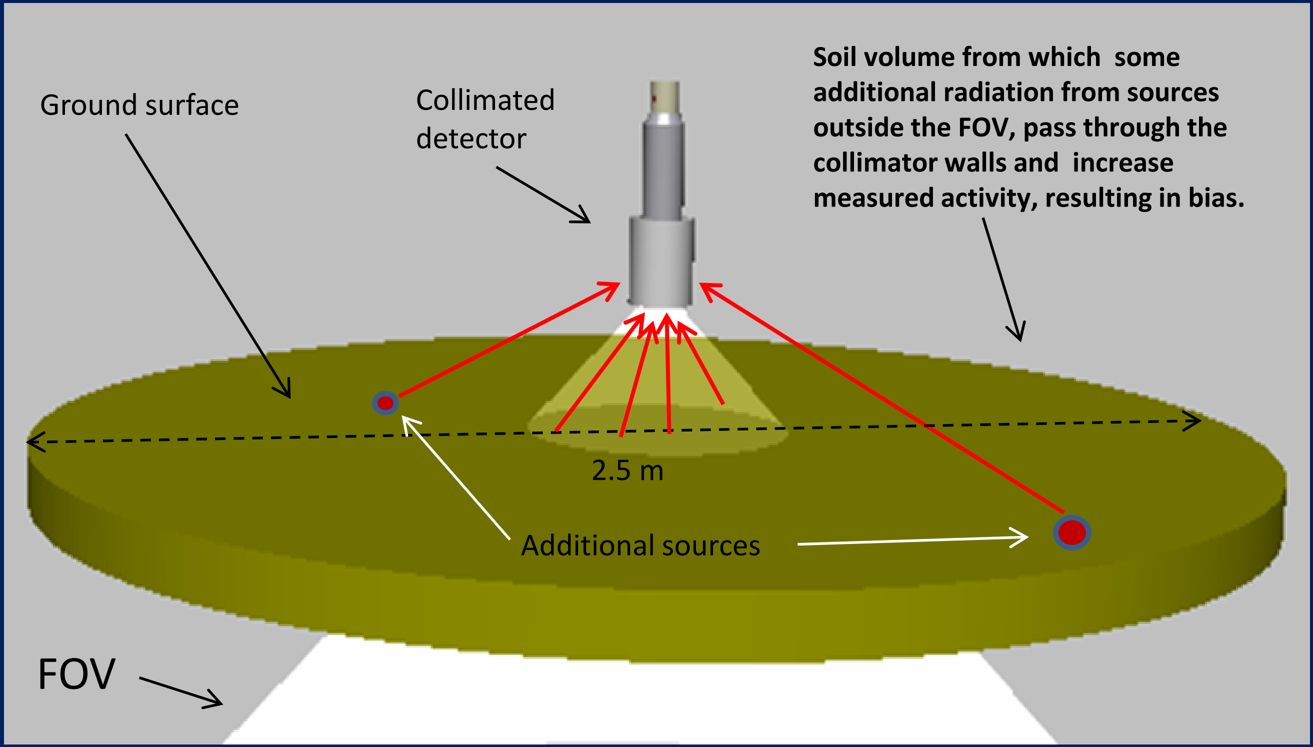
BIAS DUE TO INITIAL COMPUTER MODEL

A **90 degree 20mm thickness** lead collimator was used in the survey.

Some additional gamma radiation from the soil volume around the detector passes through the collimator side walls.

This effect could be substantially reduced by using a 20mm tungsten collimator, or (best) a 50mm lead collimator.

A 50mm lead collimator would weigh >70kg!

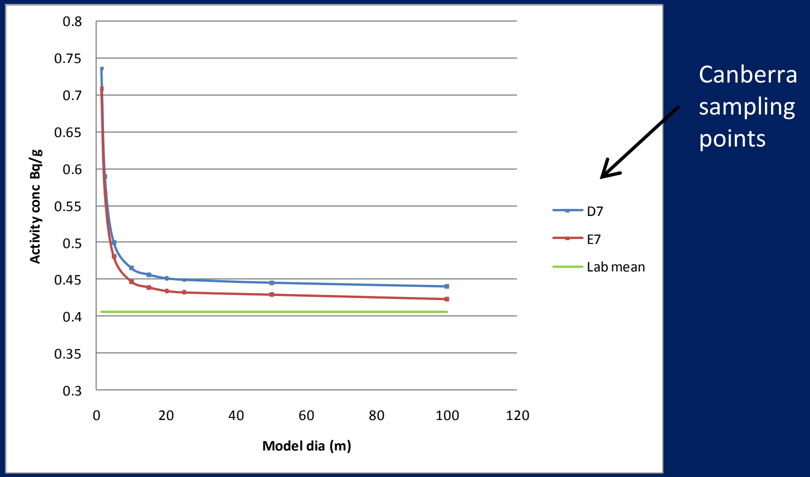


REDUCED BIAS USING IMPROVED MODEL

Increasing size of modelled soil volume **reduces** calculated activity concentration.

Desk experiment to establish minimum model dimensions for soil volume.

Based on ^{40}K gamma emissions at 1461 KeV (high penetration).



Modelled soil volume dimensions were increased until calculated activity concentration stabilized - at approximately 1m thickness and 25m diameter.

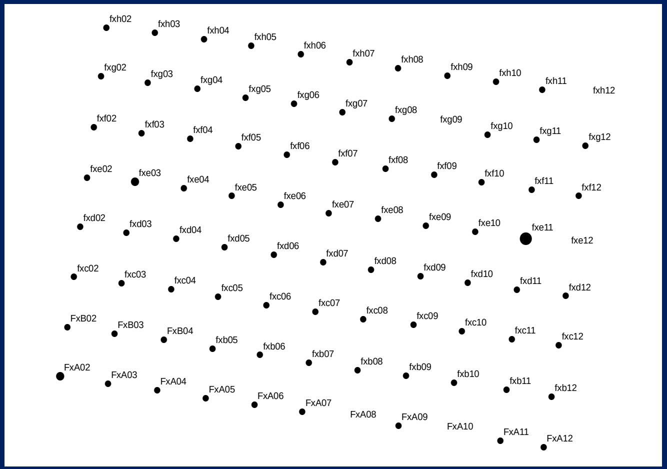
In situ relative bias reduced from **+215% (using cone model)** to **+48% (25m dia disk model)** for **single highest ^{137}Cs** measurement based on 10cm thick soil layer.

RESULTS

Zone 12 Results – Random MU (^{137}Cs)

	Mean Activity	Expanded relative uncertainty (%)		
		U_{samp}	U_{anal}	U_{meas}
Canberra *in situ*	0.043 Bq g^{-1}	0	43.9	43.9
Exploranium *in situ*	365 CPS	34.5	31.8	46.9
Ex situ 0-20cm	0.066 Bq g^{-1}	43.6	18.7	47.4
GROUNDHOG	137 CPS	N/A	12.4-18.7	12.4-18.7
SEC	0.076 Bq g^{-1}	N/A	53-303	53-303

High uncertainty for all methods. Due to proximity to Detection Limit (MDA=0.026 Bq g^{-1})?



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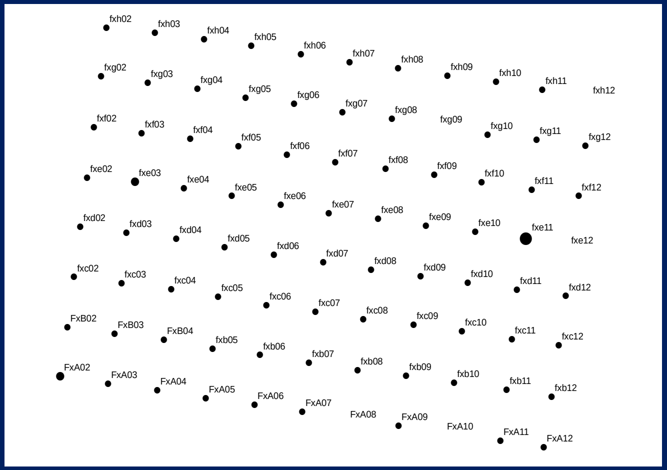

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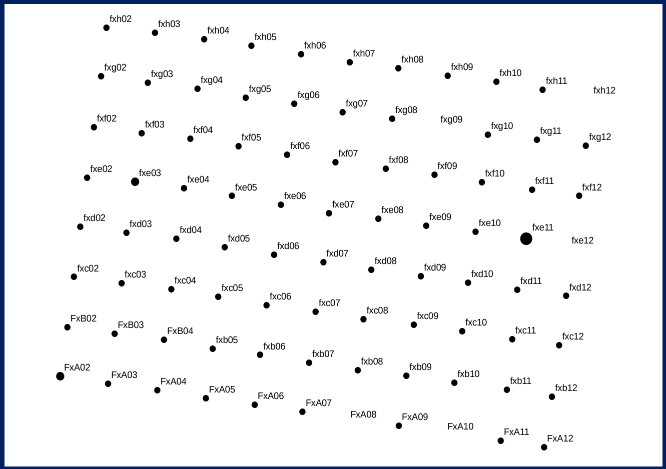
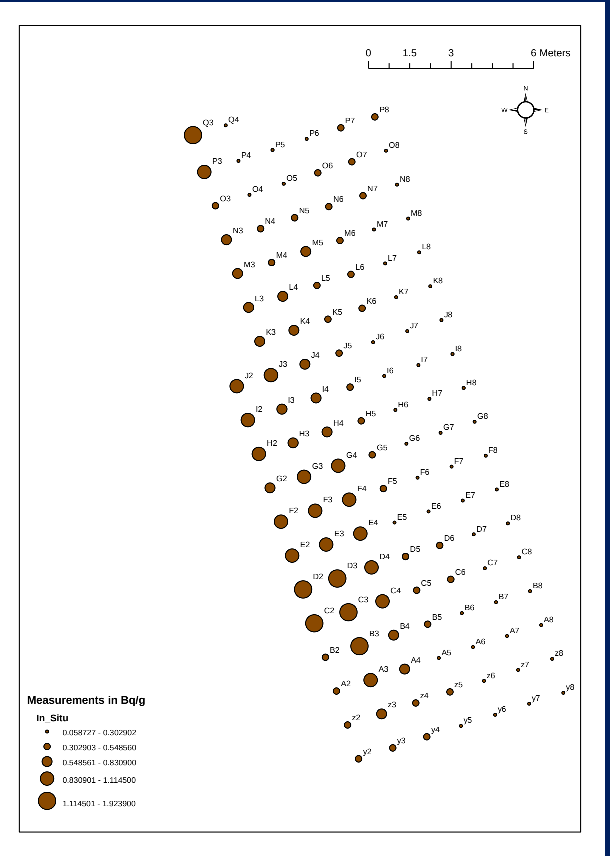
Dominated by analytical U for Canberra. Due to averaging over large soil mass?



Barrier 31 – Random MU (^{137}Cs)

		Expanded relative uncertainty (%)		
	Mean Activity	***U*samp**	***U*anal**	***U*meas**
Canberra *in situ*	**0.51 Bq g^{-1}**	**10.2**	**7.5**	**12.6**
Ex situ 0-20cm	**0.60 Bq g^{-1}**	**72.5**	**5.1**	**72.6**

Mean measurements one order magnitude > Zone 12, consequent reduction in analytical U for Canberra.



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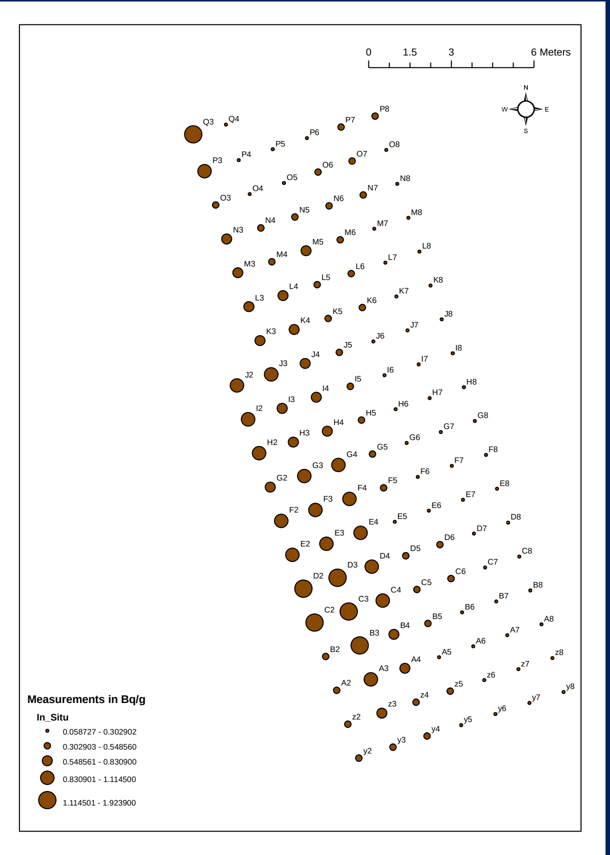

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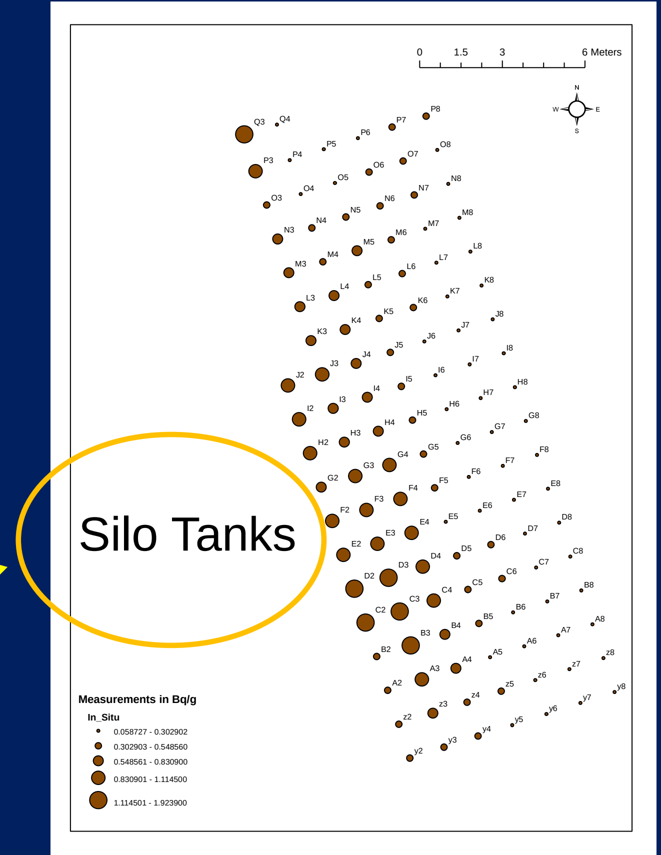
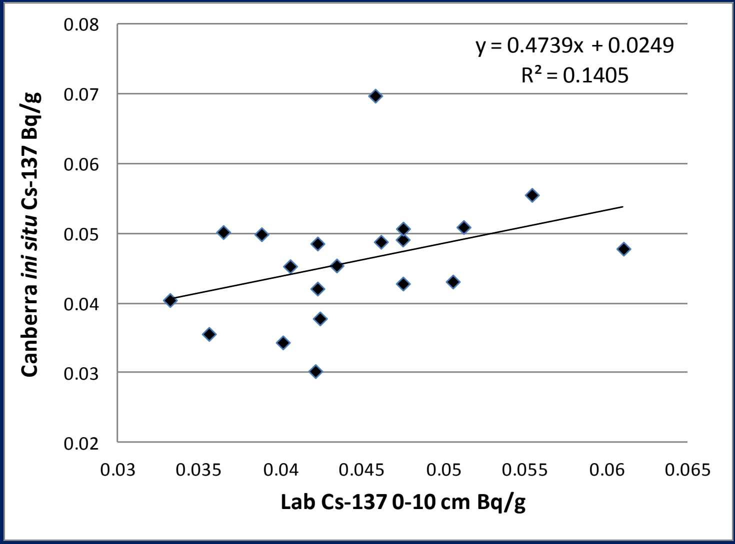
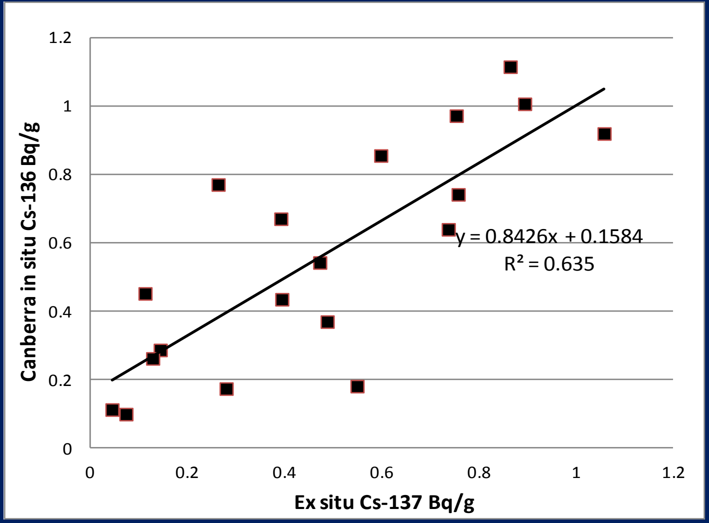
***In situ* may *be* affected by shine from the nearby silo.**



BOTH SURVEYS – SYSTEMATIC MU (BIAS)

ZONE 12 - Non-significant correlation or bias between *in situ* Canberra measurements and *ex situ* measurements (p<0.05) when single outlier excluded (high MU?)



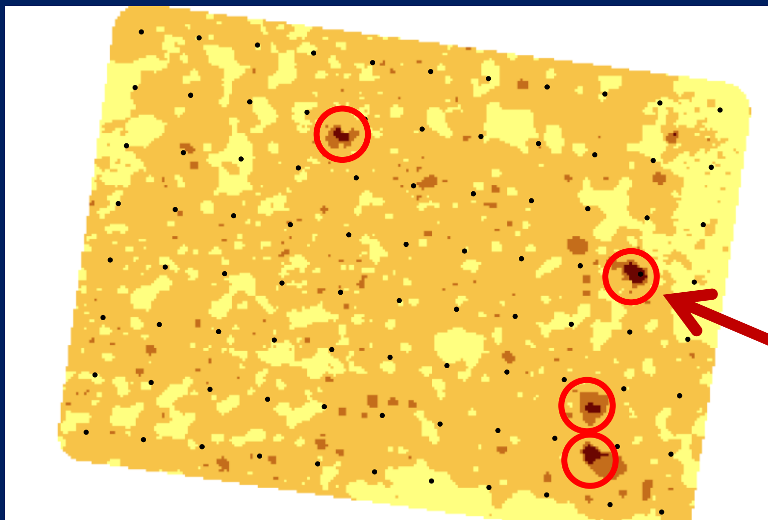


Barrier 31 – Significant correlation but **non-significant** bias when single outlier excluded.

(Bias: Slope = 1, intercept = 0)

Mann-Whitney - No significant differences Zone 12 or Barrier 31 (p<0.05)

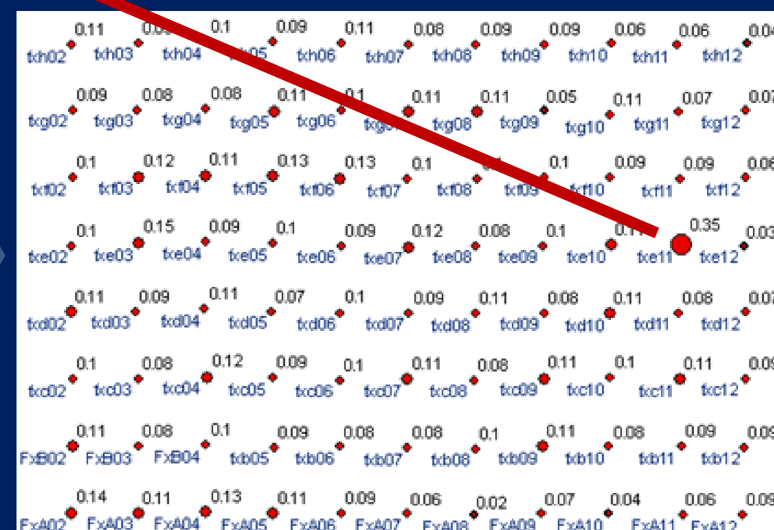
LOCATING HOTSPOTS OF ACTIVITY



Interpolated **Groundhog CPS** measurements of the survey area, showing **4 distinct “hotspots”.**

Canberra detector - Only **one** hotspot found with measurements significantly higher than the mean.

Also the case for the Exploranium and *ex situ* measurements. Even with 2m grid spacing.



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FITNESS FOR PURPOSE

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Contribution to total variance (%)		Measurement U (%)
Zone 12	Canberra *in situ*	**92.5**
	Exploranium *in situ*	**82.7**
	Ex situ 0-20cm	**51.8**
Barrier 31	Canberra *in situ*	**0.8**
	Ex situ 0-20cm	**33.4**

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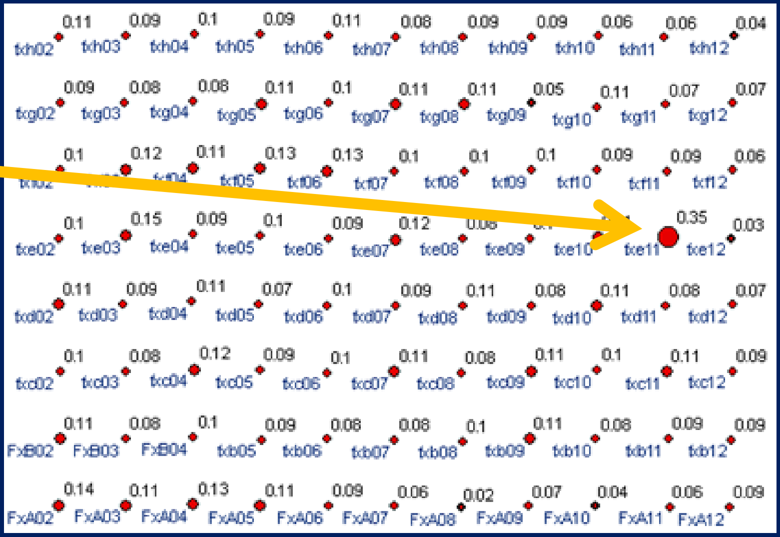
Only Canberra *in situ* FFP and only in Barrier 31

- *Low levels of analytes (Zone 12).*
- *Insufficient counting times (Zone 12).*
- *High levels small-scale heterogeneity (Barrier 31).*

FITNESS FOR PURPOSE

Hotspot identification: Only 1 located out of 4 previously found by Groundhog survey.

Neither *in situ* nor *ex situ* measurements are FFP in Zone 12.



-Limited coverage by in situ *methods.*

-Small sample size ex situ (~0.5kg).

-Low activity concentrations ^{137}*Cs.*

IN SITU SURVEY OPTIMISATION

- Basis of a decision support tool for optimising *in* situ survey methods.

- Based on probability of identifying a small particle using *in situ* measurements.

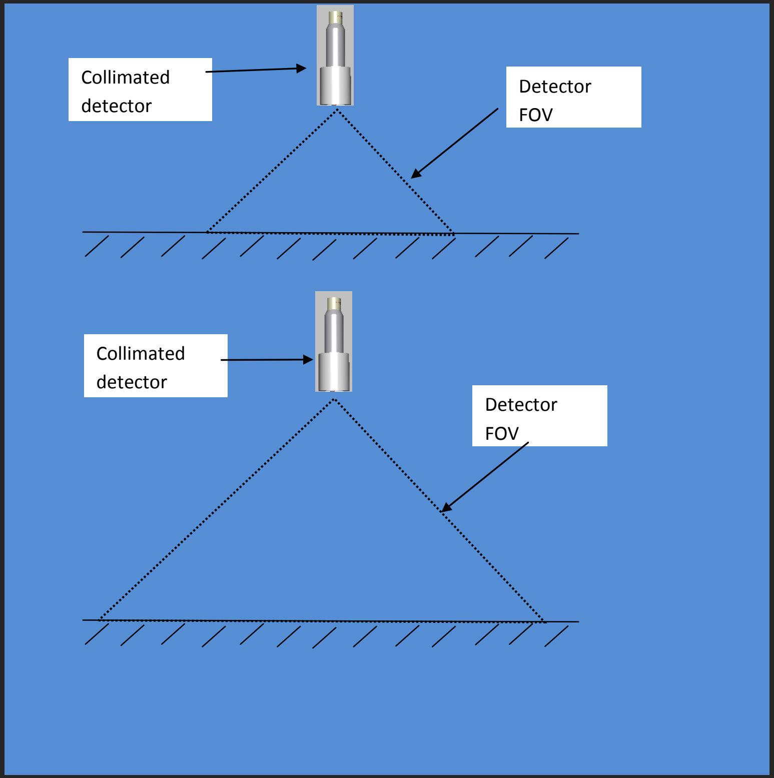
- Optimisation parameters:
  - Detector height
  - Counting time
  - Coverage% (assumed to be minimum of 100%).

- Introduces financial considerations (Ramsey *et al,* 2002).

- Does require estimation of background activity levels (mean and standard deviation) for target radionuclide.

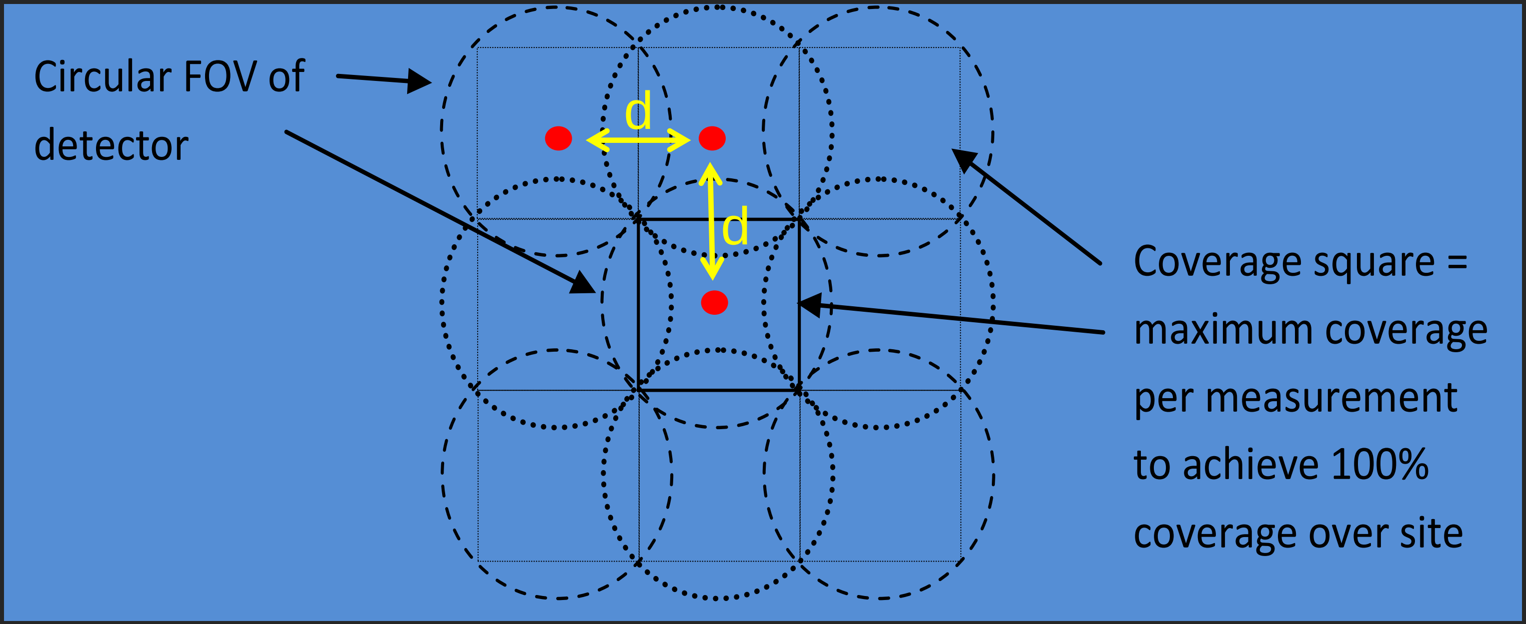
IN SITU SURVEY OPTIMISATION – HEIGHT & TIME

Increasing detector height increases the amount of ground covered by the collimator's field-of-view.

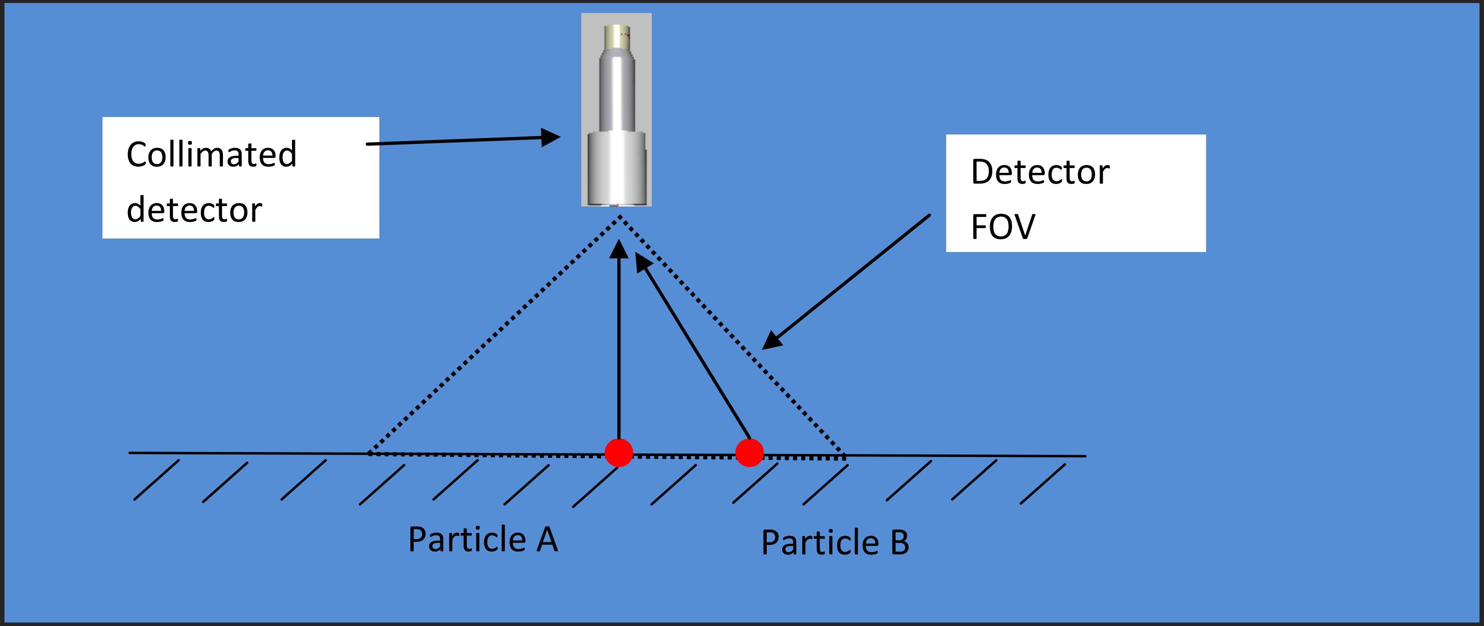


Increasing the counting time increases the probability of particle detection.

In situ measurement spacing (d) required to give 100% coverage of the ground surface by the collimator's field-of-view (FOV), when using a regular square grid design.



IN SITU SURVEY OPTIMISATION – COVERAGE %



Particle A results in more counts at the detector than Particle B.

Consequently a shorter counting time is required to detect particle A with specified probability.

May be advantage to increasing coverage to > 100%, to optimise the balance between number of measurements and counting time.

IN SITU SURVEY OPTIMISATION TOOL (INPUTS/OUTPUTS)

INPUTS (based on Barrier 31 survey)

Site area (m²)	206
Particle threshold (Bq)	10000

Detector Height Range	
Single or Min mm	300
Max mm (if range)	1200
Step mm (if range)	100

Cost Parameters		
Cost Type	Per	£
Measurement	Site (e.g. MOB)	423.54
	Measurement	4.68
	Minute	0.41
False +ve	Square metre	50
	Site	

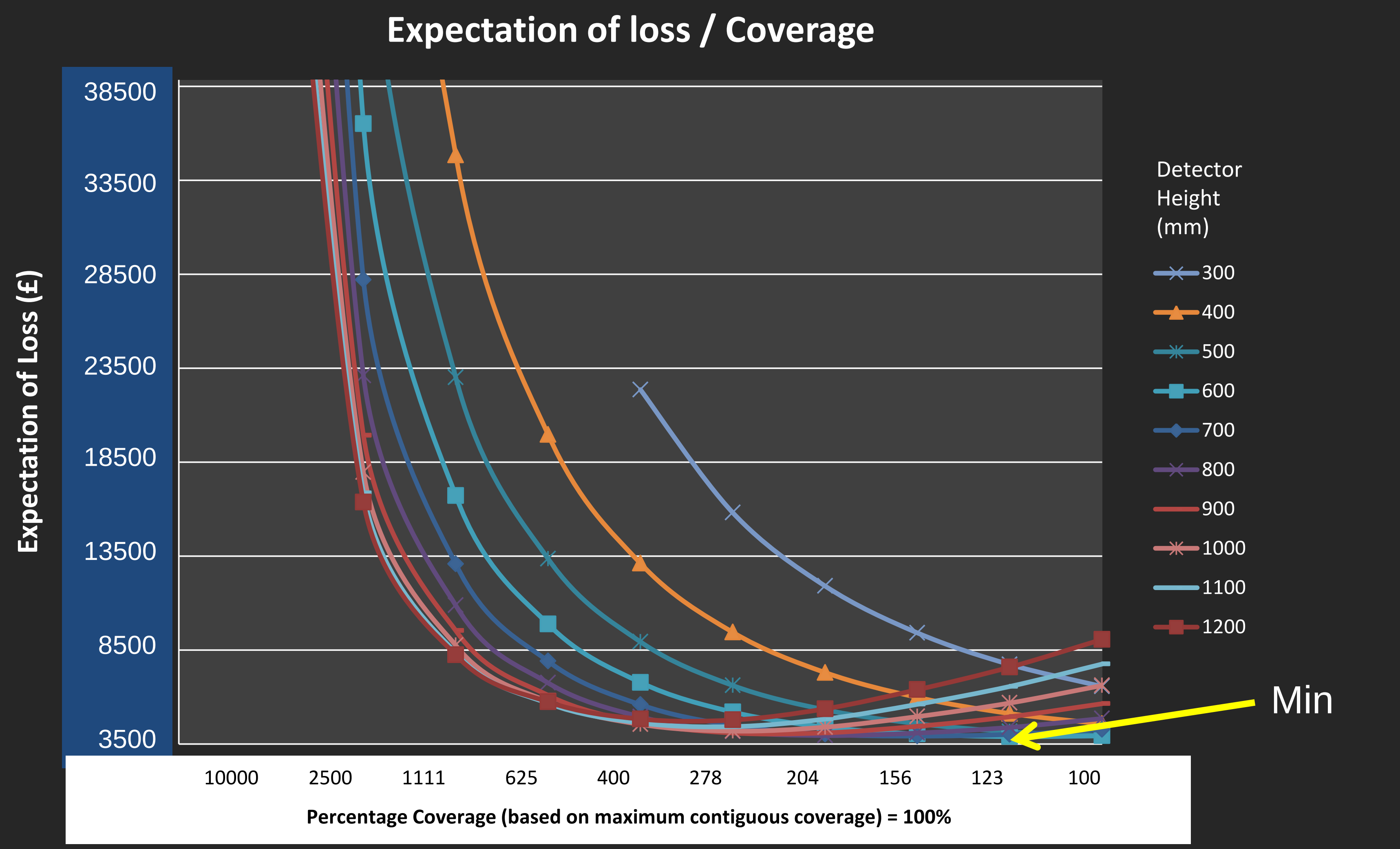
Probability ranges / measurement			
-	Single or Start	Max (if range)	Step (if range)
False +ve	0.01	0.1	0.02
False -ve	1E-06		

OUTPUTS

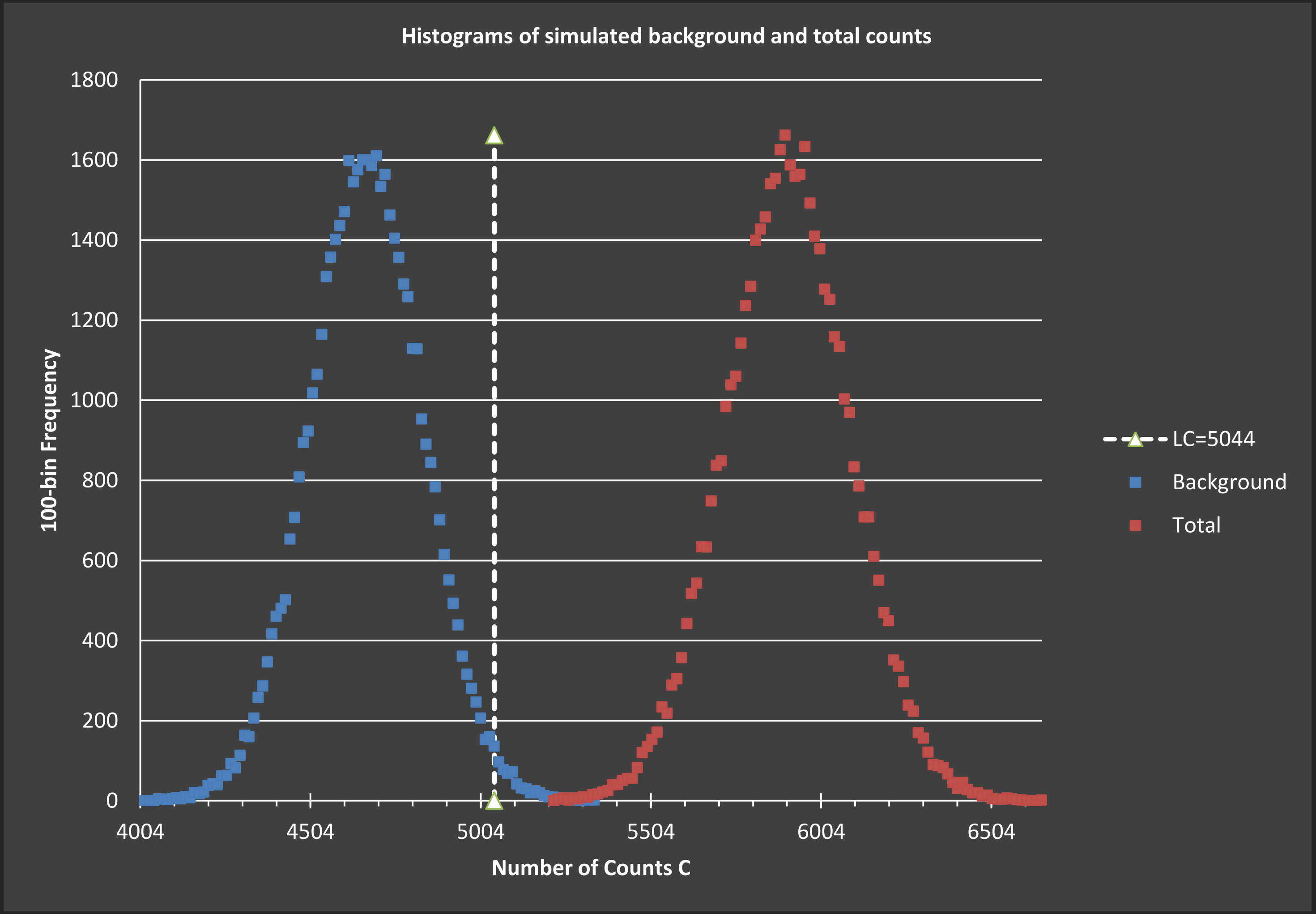
Optimised survey parameters		
Detector height	600	mm
Measurement spacing	764	mm
Counting time	707	Seconds
Critical level	5044	Counts

Optimised survey information		
Total survey time	98.76	Hours
Coverage	123	%
Number of measurements	353	(N)
Measurement cost	3782	£
Expectation of Loss	3885	£
P(false +ve), adjusted	0.0100	
P(false -ve), adjusted	0.000001	
16.5	Six hour days	

IN SITU SURVEY OPTIMISATION TOOL – OUTPUT GRAPHIC



IN SITU SURVEY OPTIMISATION TOOL - SIMULATION



	Program	Sim
CPS B	4668.874	4669.534
SD B	165.1408	164.8433
CPS T	5906.825	5906.48
SD T	185.7485	183.4508
LC	5044.206<<<<<<<<	
P(Fp)	**0.0115**	**0.0115**

B = Background (blue curve)

T = Background + Particle (red curve)

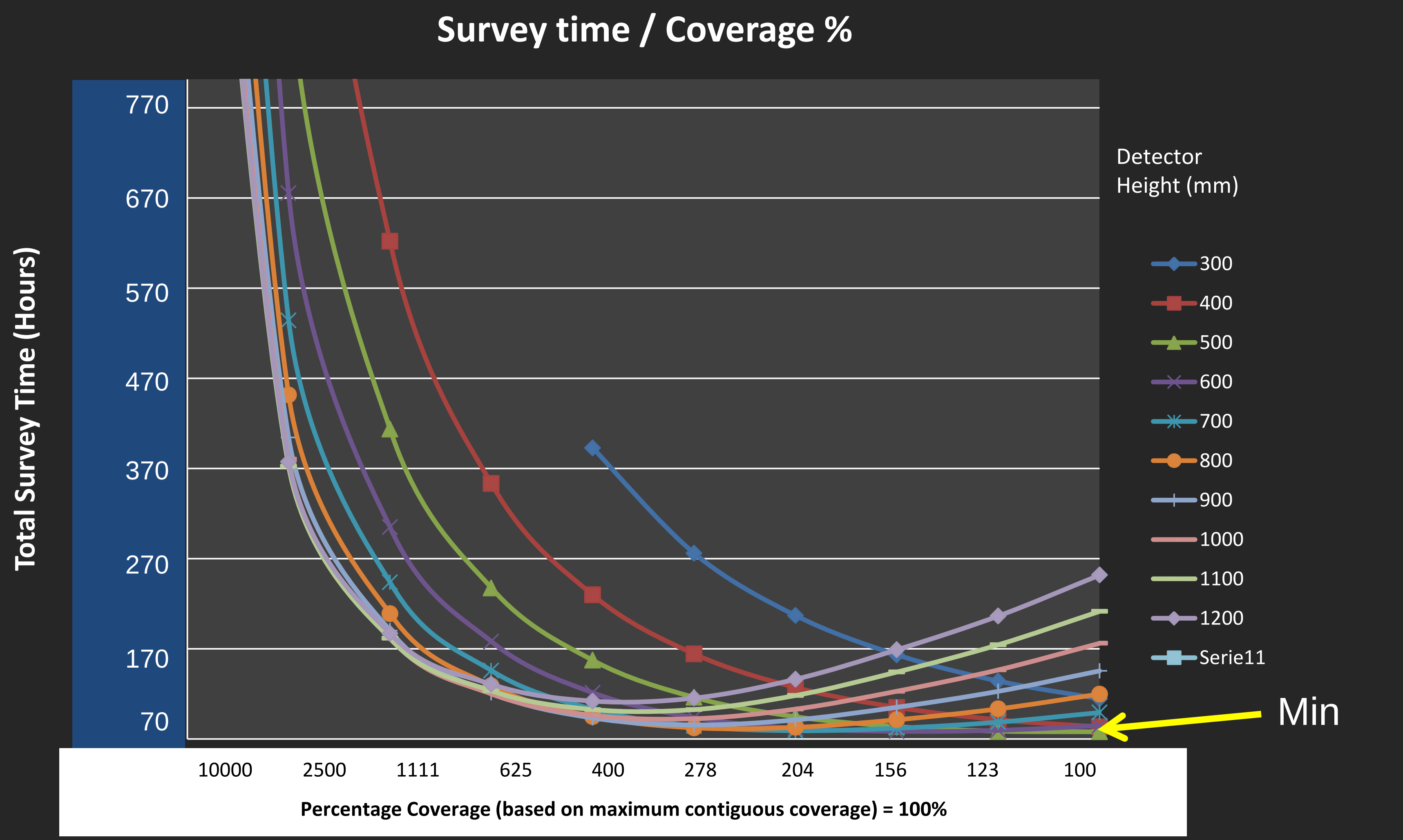
IN SITU SURVEY OPTIMISATION TOOL – TIME BASED

Optimisation on *time taken to complete survey*: 3 days shorter, but increased estimated cost (by ~£500)

Optimised survey parameters		
Detector height	**500**	**mm**
Measurement spacing	**707**	**mm**
Counting time	**403**	**Seconds**
Critical level	**2803**	**Counts**

Optimised survey information		
Total survey time	**80.41**	Hours
Coverage	**100**	%
Number of measurements	**412**	(N)
Measurement cost	**3485**	£
Expectation of Loss	**4412**	£
P(false +ve), adjusted	**0.0900**	
P(false -ve), adjusted	**0.000001**	
13.4	Six hour days	

IN SITU SURVEY OPTIMISATION TOOL



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   Probably due to a smaller effective sample mass (~1kg / several tonnes).

2. **No significant bias was found between *in situ* and *ex situ* measurements for either survey (after ISOCS model adjusted). However Zone 12 measurements were subject to high random uncertainty and would not be considered FFP according to criteria of Ramsey *et al* (1992). Probably due to proximity of measurements to MDA (these activities were well below anything of regulatory concern).**

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5. **At least 100% coverage by FOV of collimated *in situ* detector may be required to identify small hotspots of activity. Greater than 100% coverage may be optimal where it is required to find small particles with defined probability.**

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Optimised investigation of radioactively contaminated land.

Funded by EPSRC and industrial CASE partner Nuclear Decommissioning Authority via the National Nuclear Laboratory.

SURVEYS – FURTHER INFORMATION

Units = Bq (counts per second) or Bq/g (activity concentration)

Exploranium detector Cs-137 counts (Bq) estimated from recorded spectra. Not converted to activity concentration.

Canberra detector Cs-137 activity concentration (Bq/g) subsequently estimated by spectral analysis using Genie 2000 modelling software.

Soil samples analysed by on-site laboratory. Activity concentration (Bq/g) estimates provided.

Duplicate samples/analysis collected for all methods at eight or more locations for uncertainty estimation, using the balanced design method recommended by Eurachem/CITAC .