Comparison between measurement methods for the characterisation of radioactively contaminated land

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Abstract
In the UK alone, it is estimated that there may be 20,000,000 cubic metres of contaminated land at Sellafield. Harwell and Dounreay are known also to have significant amounts of radioactive or non-radioactive contaminated land. It is therefore important to devise optimal methods for the characterisation of areas of land for radionuclide content, in order to enable cost-effective decommissioning.

With chemical contaminants, ex situ measurements are made on a larger volume of soil than are in situ measurements. However, the opposite is often true for the characterisation of radioactive contamination, when this involves the detection of penetrating radiation from γ-emitting radionuclides. This means that when investigating for hotspots of radioactive contamination at or near the ground surface, better coverage can be obtained using in situ methods. This leads to the question, what is the optimal strategy (e.g. percentage coverage, counting time) for in situ characterisation of radioactively contaminated land?

Surveys on light-moderate contaminated areas of ground were conducted at Dounreay in order to compare the relative effectiveness of in situ and ex situ methods, both for the detection of radioactive hotspots and also for estimating the average radionuclide content of such areas. These surveys suggest that continuous coverage by in situ devices is more effective at hotspot detection, with ex situ laboratory measurements being less effective. The surveys also highlighted that careful choice of an appropriate spatial model for the conversion of raw counts is critical to the estimation of activity concentrations over averaging areas.

Intuitively, continuous coverage may be considered optimal for hotspot identification, particularly for the detection of hot particles, where the particle is very small compared to the sampling target. However, desk studies, based on detector efficiency calculations made by Monte Carlo modelling , have suggested that in situ surveys exceeding 100% coverage may be optimal. This is due to the decrease in detection efficiency as the particle moves towards the edge of the detector's field-of-view. Work is ongoing to devise methods for the estimation of optimal in situ survey parameters, including counting times, measurement spacing and detector height, where the objective is to minimise overall survey time, or to minimise an expectation of financial loss based on measurement costs and the probabilistic costs of false results.

1.0 Introduction

1.1 Radioactive contamination and legislation in the United Kingdom

In England and Wales, there are a total of 32 licensed nuclear sites carrying out activities such as power production, nuclear fuel & waste processing, decommissioning, and site clean-up (EA, 2012). Associated events such as fuel processing activities, leaks from disposal facilities, dismantling of buildings, and historically authorized discharges, can lead to contamination of land areas, including the floors of buildings, with radioactive material. It has been estimated that there could be up to 20 million cubic meters of radioactively and non-radioactively contaminated land at the Sellafield site alone. Other sites such as Harwell, Dounreay and Springfields are also known to have significant amounts of contaminated land, some of which is radioactively contaminated. While this is not thought to pose a significant threat to the current workforce, further work is needed to determine what actions are necessary to avoid adverse effects on people and the environment in future (NDA, 2006).

Management of contaminated land in England and Wales is the responsibility of local authorities and the Environment Agency, under Part IIA of the Environmental Protection Act (National-Archives, 1995), and sets the context of this research. Modifications to this guidance for the special case of radioactively contaminated land came into force in 2006, with separate regulatory documents for England, Wales and Northern Ireland (National-Archives, 2006a, National-Archives, 2006c, National-Archives, 2006b). These require the benefits of

any remediatory intervention to be weighed up against the health detriment and cost of intervention in order to maximise those benefits. In Scotland, modifications were made to Part IIA of the Environmental Protection Act in 2007. Under these regulations, identification of radioactively contaminated land is now the responsibility of the Scottish Environment Protection Agency (SEPA). The local authority is required to notify SEPA of areas of land which they suspect may be radioactively contaminated (National-Archives, 2007). Statutory guidance accompanying the Scottish regulations requires possible risk from radioactive contamination to be considered both in terms of long-term exposure to distributed material, and acute exposure to concentrated material (Scottish-Government, 2007).

The keeping and use of radioactive substances in the United Kingdom is governed by a system of permitting. This includes the disposal of radioactive wastes. Legislation is provided in the Radioactive Substances Act 1993 (RSA93) and the Environmental Permitting Regulations 2010. Radioactively contaminated land is not specifically covered, because the radioactivity in contaminated land is not considered to be **kept** or **used**: however, once excavated, radioactively contaminated soil or concrete becomes waste, and this is covered by the permitting system. Threshold concentrations are defined for different natural and artificial radionuclides, based on estimates of radiation dose which could be received by members of the public, i.e. a maximum allowed dose of 10 µSv/year for artificial radionuclides. As an example, where the artificial radionuclide ^{137}Cs is used in an industrial process, activity concentrations of less than 1 Bq g^{-1} are considered out-of-scope of the legislation. Materials with activity concentrations above 1 Bq g^{-1} are considered to be radioactive materials, but may be exempt from permitting provided that other exemption criteria are met (DEFRA, 2011).

1.2 Characterisation of radioactively contaminated land

Depositions of artificial radionuclides, whether they be the results of accidental or authorised emissions, are often found to be highly heterogeneous. The presence of particulate activity in such depositions has frequently been demonstrated (Brown and Etherington, 2011, IAEA, 2011, Dennis et al., 2007, Poston et al., 2007, Salbu and Lind, 2005). Individual radioactive particles may present risks to human health. They may also lead to sample heterogeneity problems, and consequent false interpretation of measurements, if those measurements are used to characterise a bulk volume (IAEA, 2011, Dale et al., 2008). In addition to the requirement to characterise areas of land for average bulk content of radionuclide activity concentrations, there is therefore an additional requirement to identify particles which could pose a hazard to human health, especially when past historical practices on the site suggest that they might exist. Survey systems, such as the vehicular mounted Groundhog™ large area search system (Dennis et al., 2007), have been developed with this purpose in mind. Action threshold activities for radioactive particles are not specifically provided by Scottish law, however definitions have been provided by other bodies, for example the Dounreay Particles Advisory Group (DPAG), which categorises radioactive particles at the Dounreay site based on their implications to public health (DPAG, 2006).

A considerable amount of work has been done to devise efficient methods for the characterisation of **chemically** contaminated land. This is typically achieved by *ex situ* methods, that is by taking soil samples, transporting them to a laboratory, processing them and performing targeted analyses for suspected contaminants. One potential issue with the alternative of making *in situ* measurements of chemically contaminated land, is that the techniques used, such as portable x-ray fluorescence (PXRF), only analyse a very small mass of soil (~1 g). This is a much smaller mass than would be collected when a single or composite soil sample is excavated, which would likely be of the order of 0.25kg - 1 kg (more in the case of composite sampling). These latter samples are usually dried and homogenised prior to laboratory analysis, and thus can be considered to be more representative than the small sample mass analysed by *in situ* PXRF. Patchy chemical contamination of soil typically involves fairly large areas (10s or 100s of square metres) and search protocols to find such areas are well understood and involve practicable numbers of measurements, e.g. Department of the Environment (1994).

The situation is quite different in the case of **radioactively** contaminated land. Again, *ex situ* soil samples or concrete cores are frequently excavated and analysed by laboratory methods, and the mass of these samples may typically be of the order of 0.25-1.0 kg. However, *in situ* methods are based on the remote detection of penetrating radiation. In the case where gamma radiation detection followed by spectroscopy is used, useful information about the presence, intensity and source of radiation may be achievable over sample spacings of up to a few tens of metres. Thus in this case the **sampling target,** defined as "the portion of material, at a particular time, that the sample (therefore the measurement result) is intended to represent"(Eurachem/CITAC, 2007) is typically much larger for *in situ* measurements than it is for *ex situ* measurements, and may be of the order of some tens of tonnes.

A potential advantage, therefore, of *in situ* gamma-ray measurements is that these may give a more representative picture of mean activity concentrations in a soil bulk, than would the analysis of excavated soil samples. This assumes that there is a high probability of gamma-emitting radionuclides at or near the ground surface. Other advantages of *in situ* measurements are that as they are taken in real time, they can be analysed immediately, and may represent a substantial cost saving when compared with often expensive laboratory analyses. In addition, where there is high heterogeneity of radioactive contaminants, possibly due to the presence of small (<<1mm) high-activity particles, high coverage *in situ* surveys would be more likely to locate radiation "hotspots" that would likely be missed in a soil sampling programme, provided the detection method is adequate and that the hotspots are near enough to the ground surface that attenuation of radiation by overlying material is not a significant factor. For such a small area of contamination, the required sampling and analysis spacing using conventional *ex situ* methodologies would result in complete excavation. However, *ex situ* analysis of soil samples may be more amenable to characterisation of the depth distributions of radionuclides, the identification of alpha/beta emitters (since this radiation is non-penetrating), and may also be prescribed by regulatory authorities (IAEA, 1998).

1.3 Uncertainty in measurements / Fitness for Purpose

Investigations of areas of potentially contaminated land will always be subject to uncertainties in the measurements. These uncertainties can contain both random and systematic errors, and include contributions from not only the analytical process, but also from the sampling and sample preparation. The random component of the uncertainty is a measure of how repeatable the measurements are, and can be expressed as the **precision** of a method. The systematic component of the uncertainty is expressed as the **bias** of a method. The ISO definition of bias is "the closeness of agreement between a test result and the accepted reference value" (Ramsey, 1998) . Estimates of bias can be made using certified reference materials (CRMs), when they are available. One part of the bias can also be expressed as the difference between two measurement methods, e.g. between an *in situ* method and an *ex situ* method, especially when the latter has its own bias estimated by the use of CRMs. Where soil samples are taken from radioactively contaminated land that is highly heterogeneous, there will frequently be a large variation between samples. This variation can often be reduced by increasing the size of the sample representing the sampling target. This might suggest a potential advantage to the use of *in situ* over *ex situ* methods when measuring mean values in a bulk volume of soil or concrete. It is therefore important to obtain estimates of the magnitudes of uncertainty in radioactively contaminated land investigations, first and foremost to understand the potential impact that measurement uncertainty can have in decision making, and secondly in order to be able to evaluate whether the measurement techniques are fit for their intended purposes (Ramsey and Argyraki, 1997, IAEA, 1998, Ramsey et al., 2002).

Methods for evaluating the Fitness-for-Purpose (FFP) of measurements made on chemically contaminated land have been proposed previously. An early method was based on the proportion of variance contributed by the measurement uncertainty, in relation to the total variance across the site (Ramsey et al., 1992). A more comprehensive approach has subsequently been suggested which aims to minimise the expected financial losses incurred by the cost of the measurement process, and the probabilistic losses that could result from misclassification. For example, a misclassification that results from falsely identifying an area as exceeding some action level (such as a threshold activity concentration) , when in fact it does not (Ramsey et al., 2002, Thompson and Fearn, 1996). Adaptation of this method to radioactively contaminated land is intended to be the subject of subsequent future work.

This paper draws on information extracted from Rostron *et al* (2012), which provides details of surveys carried out at Dounreay, Caithness, Scotland and reports on the potential advantages of using *in situ* measurements for the characterisation of radioactively contaminated land.

1.4 Objectives of this paper

The objectives of this paper are as follows:

1. Describe five measurement methods for measuring radioactive contamination that were carried out at the Dounreay site, three of which were carried out by the authors during this research, and two of which were conducted by others during previous surveys;
2. Provide estimates of the uncertainty in the measurements from each of these five techniques;
3. Comment on the relative effectiveness of these investigations for the purposes of:
    a. Estimating the spatial distributions and mean activity concentration of contamination over the survey area;

b. Location of hotspots of activity;

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

2.0 Methods

Two areas in the Dounreay site were selected for experiments intended to compare *in situ* and *ex situ* measurement methods.

2.1 Survey 1 - Zone 12 area

The first survey area (Zone 12) is an unused area of rough ground with uncut grass near the north-east corner of the site. This had not previously been built on or used to store radioactive materials, however it had previously been used as a temporary dumping ground for rubble from demolished buildings. This had been removed at the time of the survey. It was expected to be relatively uncontaminated, with the possibility of a few areas of elevated activity. Data from two previous surveys was available:

a) High-coverage survey with a Groundhog vehicle in 2009. Groundhog records a number of counts per second (CPS) that occur in a spectrum window which is set to include the characteristic energy level commonly used to identify the presence of ^{137}CS (661 keV, emitted by its short-lived daughter ^{137m}Ba. Recordings are taken every second as the vehicle moves slowly (~1m/s) along overlapping parallel swathes of land.

b) SEC survey. This was a site-wide survey conducted by a contractor in 2003-2004, using back-pack mounted gamma detection with a recording interval of 1 second. Recorded activities were converted to activity concentrations for five target radionuclides, including ^{137}Cs.

Both the Groundhog and SEC survey measurements were recorded with GPS positional data. In addition, data was available from previous health physics surveys which showed some areas of activity that were elevated above the local background activity.

Part of the Zone 12 area was surveyed by the authors in 2010. The area surveyed measured 20m x14m, and a total of 88 measurement locations were designated covering an area of 280m^2, laid out as a regular grid with 2m spacing between measurement locations. Prior to survey commencement, the grid was marked out using a Trimble differential GPS unit for the four corners of the area (with a spatial accuracy of a few cm) and tape measures used to position each location. Three measurement methods were used:

a) *In situ* measurements at 88 measurement locations were recorded using a Canberra 3x3" NaI detector fitted with 90 degree, 20mm lead collimation, mounted on a wheeled trolley at a height of 280mm above the ground surface. A Canberra "Inspector" unit was used to record spectra at each measurement location. These were subsequently analysed using Canberra Genie software with ISOCS modelling, in order to convert measurements from CPS into estimates of activity concentration. The counting time used was 600 seconds at each location.

b) *In situ* measurements at 88 measurement locations using un-collimated Exploranium GR135 portable detector, incorporating a 2x2" NaI scintillation detector. After clearing the grass, this instrument was placed directly on the ground surface with the detector volume over each measurement location. The counting time used was 600 seconds.

c) *Ex situ* measurements: Soil samples were excavated at 20 randomly selected measurement locations by extracting the top 10cm of soil using an 8cm diameter bulb planter. These were placed into plastic pots and transferred to the on-site laboratory for gamma analysis. The average soil mass excavated was 330g.

The collimated Canberra unit was selected as it has been characterised to enable conversion of recorded activity (in CPS) into activity concentrations using ISOCS modelling. The Exploranium used did not have this capability, but is a small, portable unit which is much easier to set up and use. The sampling and analytical components of uncertainty were estimated using the "balanced design" methodology as described in the Eurachem guide (Eurachem/CITAC, 2007). The method relies on assigning a number (a minimum of eight) of the measurement locations as **duplicate** measurement locations. At each of these, it is then necessary to take **two** measurements at the primary measurement location, and an additional **two** measurements at a duplicate measurement location. Subsequent analysis of variance (ANOVA) enables the component variances that are due to the analytical method, and due to the sampling protocol (i.e., the spatial position of primary measurement

locations), to be estimated. The question of where to take the duplicate measurements in relation to the primary measurements had to be addressed at the survey planning stage. A common approach is to place the duplicate location at the extremity of the expected error in positioning of the measurement locations. In this case, however, the expected absolute positional error of the Trimble differential GPS unit was of the order of a few cm (less than the diameter of the bulb planter), so it was decided to take the duplicate measurements 20cm (10% of the inter-measurement spacing) from the original measurement location. The intention of this approach was to assess the small-scale heterogeneity of the site and its contribution to the measurement uncertainty. Each duplicate measurement location was thus assigned to be 20cm from the original measurement location in a randomly selected direction N,S,E or W. For *in situ* measurements, **nine** duplicate measurement locations were randomly selected. At each of these, two counts were recorded at the primary measurement location, and a further two counts recorded at the duplicate measurement location. For *ex situ* measurements, **eight** of these locations were designated as duplicate *ex situ* measurement locations. Two soil samples were extracted, one at the primary measurement location, and one at the secondary measurement location. In addition, soil from a depth 10-20cm was excavated and stored separately from the 0-10cm samples, for these locations only. The soil samples so obtained were each analysed twice (second count on a different detector) by the on-site laboratory.

Survey parameters for the Zone 12 survey are summarised in Table 1.

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: 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

Table 1 Parameters used in the Zone 12 survey

2.2 Survey 2 – Barrier 31 area

The second survey conducted by the authors was carried out in 2011 within a barriered area of the site, where it was expected to find low-moderate levels of soil contamination. The area itself was a patch of rough land under cut grass, adjacent to an underground radioactive waste storage silo. The survey was carried out between two previously used active drains which run along the east and west edges of the site. No Groundhog or SEC survey data were available for this area, however some previous survey data was available at the design stage, from a database of health physics surveys and laboratory analysis of extracted soil samples.

Two measurement methods were used in the current research:

- **a)** *In situ* measurements at 122 measurement locations were recorded using a Canberra 3x3" NaI detector fitted with 90 degree, 20mm lead collimation, mounted on a wheeled trolley at a height of 920mm above the ground surface. Spectra were recorded on a laptop computer at each measurement location and subsequently analysed using Canberra Genie software with ISOCS modelling, in order to convert measurements into estimates of activity concentration. The counting time used was 600 seconds.
- **b)** *Ex situ* measurements: Two soil samples were acquired at 20 randomly selected measurement locations by extracting the top 0-10cm of soil, and the lower 10-20cm layer, using a 8mm diameter bulb planter. These were placed into plastic pots and transferred to the on-site laboratory for gamma analysis. The average soil mass excavated was 264g.

Sampling and analytical measurement uncertainty was estimated using the same method as for Zone 12. In the case of Barrier 31, a total of twelve measurements were designated duplicate measurement locations for the Canberra *in situ* measurements. A randomly assigned selection of eight of these *in situ* duplicate measurement locations were designated as duplicate measurement locations for *ex situ* measurements. Note that in both the Zone 12 and the Barrier 31 surveys, the *ex situ* duplicate measurement locations were designated at positions which had previously been designated as duplicate measurement locations for *in situ* methods. This enabled comparison of uncertainty estimates for both methods.

Survey parameters for the Barrier 31 survey are summarised in Table 2.

Area: Irregular 206m^2
Measurement spacing: 1.3m
In situ detector: Canberra 3X3" NaI, 90° 20mm lead collimator
In situ detector height: 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

Table 2 Parameters used in the Barrier 31 survey

2.3 ISOCS modelling

It is worth noting that in order to convert detector activity counts (in CPS) to estimates of activity concentrations, necessitates either the use of a calibration target of known concentration (which is hard to achieve when surveying a land area) or the use of calibration software. In this study, activities recorded by the Canberra *in situ* detector were converted to activity concentrations using Canberra software (ISOCS). When the Zone 12 measurements were initially converted in this way, a high positive bias (>200%) was observed between the mean of the Canberra *in situ* measurements and the mean of the *ex situ* measurements. This was later found be due to the use of an inadequate ISOCS model, which was initially designed such that the mass of soil analysed was just contained within the field-of-view of the 20mm lead collimator. Further ISOCS experiments showed that the as the radius of the modelled soil volume was increased, the value of the bias dropped significantly, and stabilised when the model diameter reached approximately 25m (Figs 1 & 2). This suggests that, even with 20mm lead collimation, a significant proportion of the radiation received within the detector body may have its source in the much larger area of ground surrounding the field-of-view.

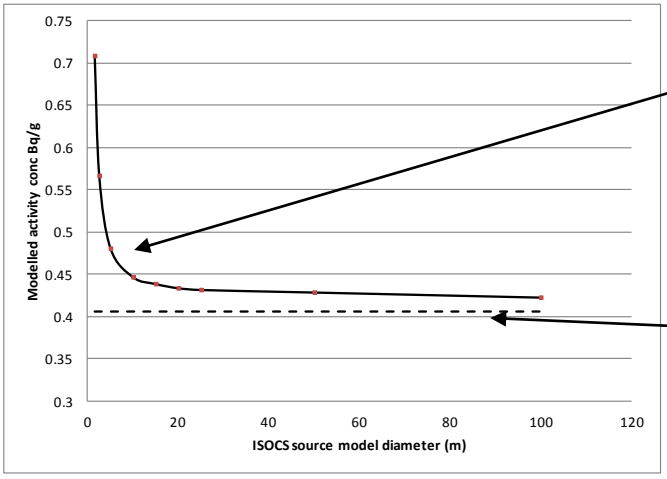

Fig 1 Enlarging the diameter of the source volume in the ISOCS model outside of the field-of-view of the collimator reduces the estimated activity concentration when measurements are interpreted. This reduction appears to stabilise at a source diameter of about 25m. The curve shown is for ^{40}K at Zone 12 measurement point E7. The reason for choosing ^{40}K was is that the characteristic energy level (1461keV) is on the high end of the energy spectrum, and is therefore more likely to penetrate the collimator walls. It is intended to represent a "worst case" situation.

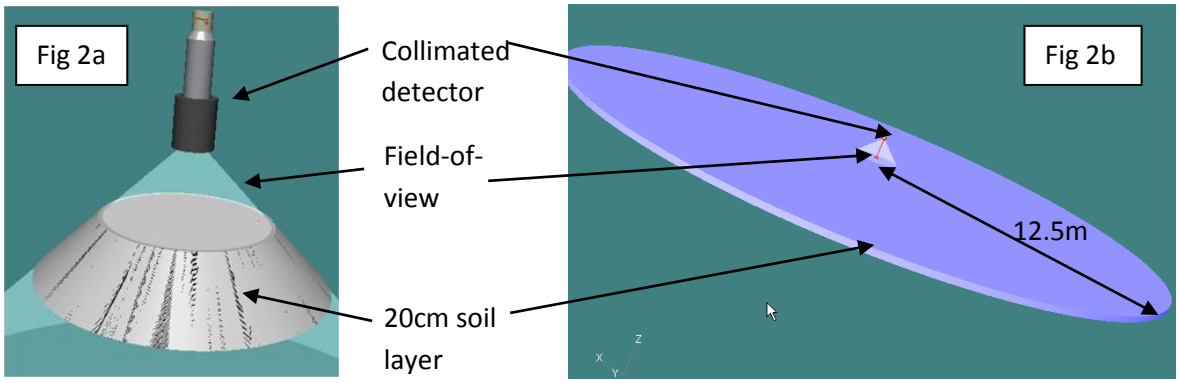

Fig 2 Showing two ISOCS source / detector geometry models used in the Zone 12 survey. Initial conversion to activity concentration based on the model in Fig 2a resulted in a positive bias when compared to *ex situ* measurements. The model in Fig 2b reduced this bias significantly. This was the model subsequently used to convert the Canberra *in situ* measurements in both surveys.

2.4 Estimates of random measurement uncertainty

2.4.1 Random uncertainty in the Zone 12 and Barrier 31 surveys

As previously described, uncertainties for the *in* situ and *ex situ* measurements obtained by the authors were estimated using the balanced design method (Eurachem/CITAC, 2007). To estimate the random components of measurement uncertainty, the measurements obtained from the sampling and analytical duplicates were analysed using robust ANOVA (AMC, 1989). This method splits the total variance in the measurements between the sampling and analytical processes. Expanded relative components of the uncertainty (sampling and analytical) are then calculated from the standard deviations as follows:

$$U_{samp}\ (\%) = 200 * s_{samp} / (\text{Robust mean})$$

$$U_{anal}\ (\%) = 200 * s_{anal} / (\text{Robust mean})$$

Where s_{samp} and s_{anal} are the standard deviations corresponding to the contributions of the sampling and analytical processes to the overall variance.

The expanded relative measurement uncertainty (U_{meas}) is then calculated as:

$$U_{meas} = \sqrt{(U_{samp}^2 + U_{anal}^2)}$$

2.4.2 Random uncertainty in the Groundhog and SEC survey data

It was not possible to use this method for the data provided by the previous Groundhog and SEC surveys, because intentional duplicate sampling and analysis had not been carried out. However, both databases contained a number of spatially coincident measurement pairs. In the case of Groundhog, this would occur when the vehicle stopped moving. In the case of the SEC survey, measurements were considered to be spatially coincident if the recorded positions were within 1m, as this was the reported accuracy of the GPS unit used when tested against a reference point at the beginning of each day.

A method to estimate precision (used here as the random component of uncertainty) from duplicated measurements has previously been suggested (Thompson and Howarth, 1976). This method has the advantage that uncertainty is estimated as a function of concentration, thus taking into account any variation in the uncertainty across the range of measured values. It assumes that samples are drawn from a normally distributed

population, and uses the fact that the median of the absolute differences between pairs of values drawn at random from a normal distribution tend towards 0.9539σ of the parent distribution (in this case, the parent distribution is the distribution of measurements). In brief, the method calculates the mean and absolute difference between each duplicate measurement pair, arranges these in ascending order of the mean, and assigns them to groups of equal numbers of pairs. The median absolute difference of each group is then regressed against the group mean. The regression coefficients are multiplied by 1.0483 (i.e. 1/0.9539) to adjust them to the standard deviation of the measurements distribution in order to obtain estimates of the standard deviation over the full range of measurements (Thompson and Howarth, 1976).

In both cases, these measurement pairs might be considered to be analogous to the analytical duplicates described previously. This assumption needs to be treated with caution as the reasons for these duplicates are unclear, e.g. the Groundhog vehicle may have stopped moving due to the identification of an area of raised activity. Thus using this method could have introduced a bias to the estimation of random uncertainty.

The method described above was applied using a total of 796 spatially coincident measurement pairs that were extracted from the Groundhog database for the Zone 12 area. As there were insufficient numbers of such pairs in Zone 12 to be able to use the method for the SEC survey, pairs were extracted from the SEC database for the entire Dounreay site.

3.0 Results and Discussion

3.1 Estimates of activity and activity concentrations

All measurement data and uncertainty estimates are based on CPS or activity concentrations of ^{137}Cs, which was the only artificial radionuclide which consistently showed activity levels above the Minimum Detectable Amount (MDA) in both the Zone 12 and the Barrier 31surveys. A summary of the measurements obtained in surveys and by all the methods defined is presented in Table 3. Dot maps of the Canberra *in situ* measurements are shown for both survey areas in Figs 3 and 4.

Table 3 Summaries of the measurements obtained in the Zone 12 and Barrier 31 surveys for ^{137}Cs. Some components have been extracted from (Rostron et al., 2012).

		No. measurements	Mean Activity	Median Activity	Standard deviation	Range
Zone 12 *In situ*	Canberra	88	0.043 Bq g^{-1}	0.043 Bq g^{-1}	0.015 Bq g^{-1}	0.01-0.148 Bq g^{-1}
	On *ex situ* locs[1]	8	0.056 Bq g^{-1}	0.045 Bq g^{-1}	0.035 Bq g^{-1}	0.034-0.148 Bq g^{-1}
	Exploranium	88	365 **CPS**	333 **CPS**	251 **CPS**	215-2525 **CPS**
Ex situ	*Ex situ* 0-10cm	20	0.047 Bq g^{-1}	0.043 Bq g^{-1}	0.013 Bq g^{-1}	0.033-0.098 Bq g^{-1}
	Ex situ 10-20cm	8	0.081 Bq g^{-1}	0.047 Bq g^{-1}	0.090 Bq g^{-1}	0.033-0.318 Bq g^{-1}
	Ex situ 0-20cm[2]	8	0.066 Bq g^{-1}	0.047 Bq g^{-1}	0.048 Bq g^{-1}	0.033-0.189 Bq g^{-1}
Previous *in situ*	GROUNDHOG	~12000[3]	137 **CPS**	135 **CPS**	20.2 **CPS**	77-306 **CPS**
	SEC[4]	~2000[3]	0.076 Bq g^{-1}	0.072 Bq g^{-1}	0.042 Bq g^{-1}	0.0-1.92 Bq g^{-1}
Barrier 31 *In situ*	Canberra	122	0.51 Bq g^{-1}	0.37 Bq g^{-1}	0.41 Bq g^{-1}	0.06-2.79 Bq g^{-1}
	On *ex situ* locs[1]	20	0.63 Bq g^{-1}	0.59 Bq g^{-1}	0.44 Bq g^{-1}	0.10-1.92 Bq g^{-1}
Barrier 31 *Ex situ*	*Ex situ* 0-10cm	20	0.67 Bq g^{-1}	0.53 Bq g^{-1}	0.83 Bq g^{-1}	0.03-3.94 Bq g^{-1}
	Ex situ 10-20cm	20	0.53 Bq g^{-1}	0.39 Bq g^{-1}	0.46 Bq g^{-1}	0.06-2.04 Bq g^{-1}
	Ex situ 0-20cm[2]	20	0.60 Bq g^{-1}	0.48 Bq g^{-1}	0.64 Bq g^{-1}	0.04-2.99 Bq g^{-1}

[1]Canberra on *ex situ* locations – Measurements that were obtained at *ex situ* measurement locations.
[2]Ex situ 0-20cm measurements calculated by taking averages of the two depths 0-10cm and 10-20cm.
[3]Groundhog and SEC survey data for Zone 12 was extracted from a larger area than defined in Table 1.
[4]SEC - Negative measurements of activity concentration and high value outliers have been excluded.

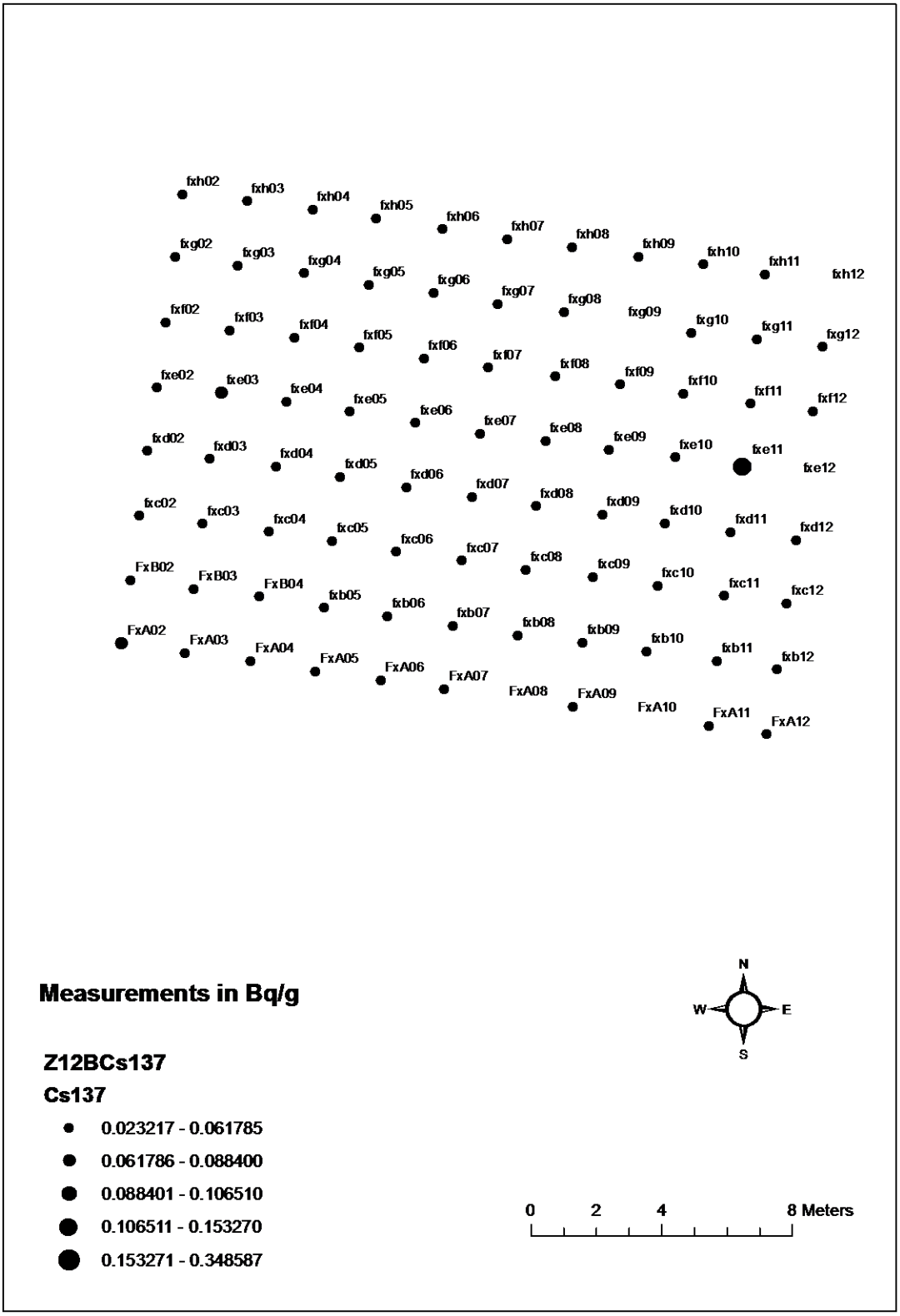

Fig 3 Dot map of the Canberra *in situ* measurements of ^{137}Cs activity concentrations in the Zone 12 survey. Dot size is proportional to recorded activity concentration. Reproduced from (Rostron et al., 2012)

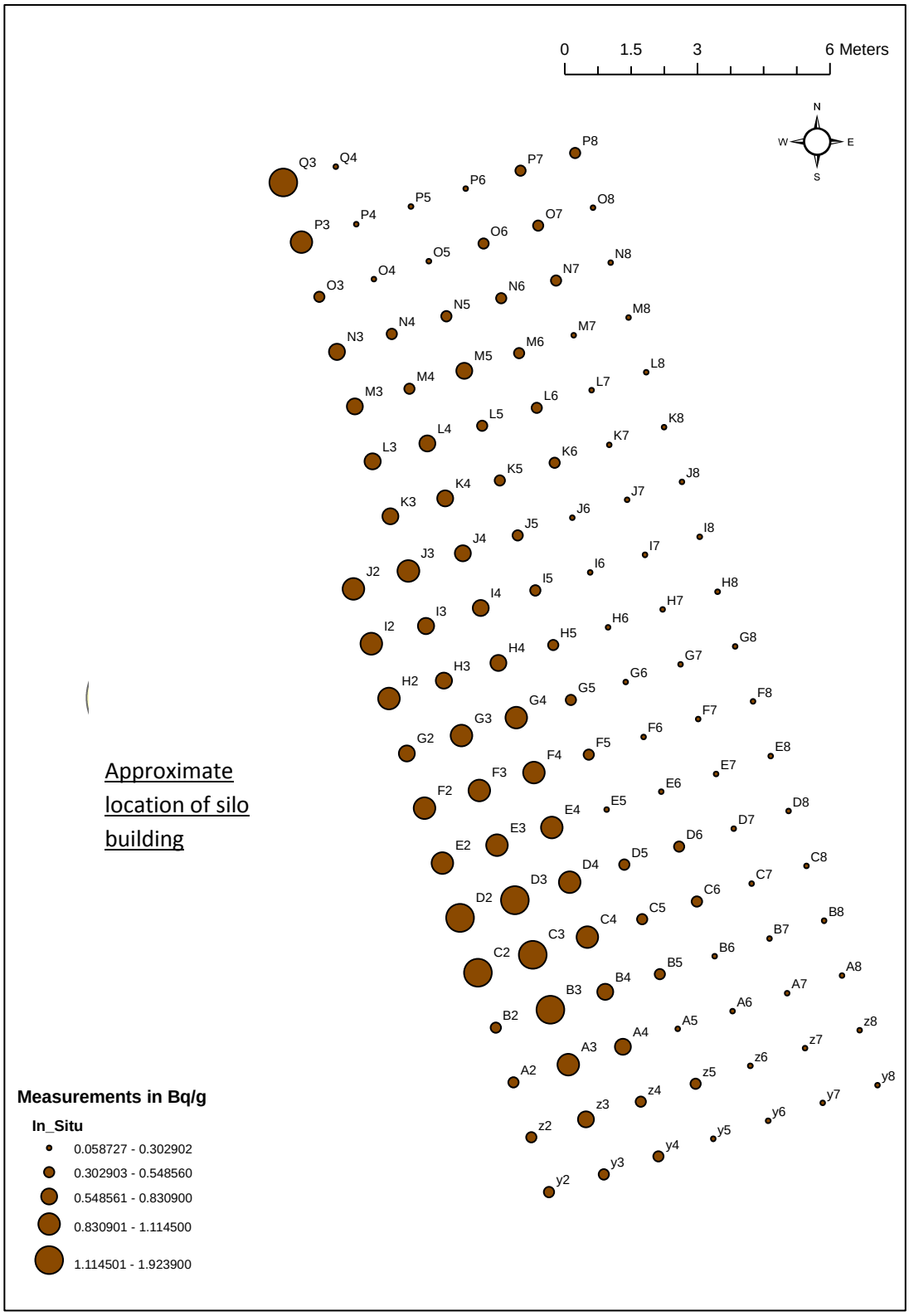

Fig 4 Dot map of the Canberra *in situ* measurements of activity concentrations of ^{137}Cs in the Barrier 31 survey. Dot size is proportional to recorded activity concentration. Reproduced from (Rostron et al., 2012)

3.2 Random component of measurement uncertainty

The estimated uncertainties for the Zone 12 and Barrier 31 surveys are summarised in Table 4.

Table 4 Summary of the random components of measurement uncertainties estimated for the Zone 12 and Barrier 31 surveys for ^{137}Cs measurements. Some components have been extracted from (Rostron et al., 2012).

		Expanded relative uncertainty (%)		
		U_{samp}	U_{anal}	U_{meas}
Zone 12	Canberra *in situ*	0	43.9	**43.9**
	Exploranium *in situ*	34.5	31.8	**46.9**
	Ex situ 0-10cm	31.5	20.3	**37.5**
	Ex situ 10-20cm	56.8	17.2	**59.4**
	Ex situ 0-20cm	43.6	18.7	**47.4**
	GROUNDHOG	N/A[1]	12.4-18.7	**12.4-18.7**
	SEC	N/A[1]	53-303	**53-303**
Barrier 31	Canberra *in situ*	10.2	7.5	**12.6**
	Ex situ 0-10cm	40.1	5.1	**40.4**
	Ex situ 10-20cm	96.1	4.9	**96.2**
	Ex situ 0-20cm	72.5	5.1	**72.6**

[1]U_{samp} not calculated for Groundhog or SEC measurements

3.2.1 Random measurement uncertainty in the Canberra *in situ* measurements.

The high uncertainty in Zone 12 for the Canberra *in situ* measurements is likely to be a result of the proximity of the mean activity concentrations (Table 3) to the MDA. The MDA was calculated using Genie spectrum analysis software. The average MDAs for Zone 12 and Barrier 31 are 0.026 Bq g^{-1} and 0.029 Bq g^{-1} respectively. For Zone 12, all the uncertainty is analytical uncertainty (Table 4), suggesting that measurements in this survey were strongly affected by analytical noise, due to proximity of these measurements to the MDA. The duplicate method estimated 0% sampling uncertainty in Zone 12, and 10.2% for Barrier 31. In both cases the ISOCS calibration model used for conversion of detector activity to activity concentration was based on a large volume of soil of 25m diameter and an estimated 160 tonnes in mass. It may be that the there was sufficient variation in measured activity concentration in Barrier 31 to create sampling uncertainty, and it may also be that radiation shine from the silo building to the West of the survey area (discussed below) had an effect on this.

3.2.1 Random measurement uncertainty in the Exploranium *in situ* measurements.

Measurements made by the Exploranium Gr135 were recorded as counts per second, and no attempt was made to convert these to activity concentrations. It is interesting to note that, although the unit was not collimated, in estimations of measurement uncertainty the sampling uncertainty dominates at 34.5% (Table 4). It might be expected that the sampling uncertainty would be no greater than that of the Canberra (0% in Zone 12) because the measurements would be "smoothed out" across a larger soil volume than that measured by the collimated Canberra unit. However, as the Exploranium was placed directly on the ground surface at each measurement location, whereas the Canberra detector was positioned at a height of 280mm, it is likely that the Exploranium measurements were much more seriously affected by attenuation of radiation within the soil itself. This might be expected to have a collimating effect on the Exploranium measurements, resulting in them "seeing" radiation from a smaller soil volume than the Canberra. If this is the case, then it would imply that there is significant heterogeneity of ^{137}Cs activity across the site, but that this was smoothed out in the Canberra measurements due to the size of the sampling volume.

3.2.3 Random measurement uncertainty in the *ex situ* measurements.

Random uncertainty for the *ex situ* measurements at 0-20cm was estimated at 47.4% (Zone 12) and 72.6% (Barrier 31). Sampling uncertainty dominates in both cases, contributing 84% and 99% to the measurement uncertainty respectively. This suggests that the small scale heterogeneity of ^{137}Cs activity concentrations causes significant uncertainty when analysing comparatively small soil samples. It might then be suggested that

estimations of activity concentration by *in situ* methods (e.g. the Canberra) are more representative of activity in the site area due to the use of a much larger sampling volume. The higher analytical component of this uncertainty in Zone 12 is to be expected, because the mean activity concentrations are an order of magnitude lower than those in Barrier 31.

3.2.4 Random measurement uncertainty in the previously conducted Groundhog survey

An estimate of analytical uncertainty of 12.38-18.68% was made for Groundhog, using spatially coincident measurement pairs. In common with the Exploranium measurements, no attempts were made to convert the recorded CPS into activity concentrations, and it is not clear how large the sampling volume would be. The Groundhog vehicle uses an array of five large un-collimated NaI detectors, at a height of approximately 250mm above the ground surface. It would therefore be expected that the sampling volume comprises a larger mass of soil than that for the Canberra. It is interesting that the estimate of analytical uncertainty is less than for any of the other methods in Zone 12, especially given the short counting time of 1 second, and is probably a result of the large detector volume. The figure of $U_{meas} < 20\%$ could be considered to be reasonable for this system, the primary function of which is to identify where activity is raised compared to the immediate surroundings.

3.2.5 Random measurement uncertainty in the previously conducted SEC survey.

Analytical uncertainty was estimated at 53-303% for the SEC survey, using spatially coincident measurement pairs. These very high uncertainty levels could be partly due to the short counting time of 1 second with a single 3" x 3" NaI detector. However, this estimate of uncertainty may also not be reliable, due to the method of selecting coincident measurement pairs (i.e. points within 1 metre). The primary objectives of the SEC survey was to provide 5-component analysis of gamma radiation, and to report any Cs-137 activity > 10^5 Bq. Total counts per second (TCPS) were also recorded. Using an alternative method to estimate uncertainty from measurement pairs, the Mean Absolute Difference method (Gill, 1997), yields estimates of 112% when based on Cs-137 activity concentrations, but just 8% when applied to TCPS. This might suggest that a large component of the uncertainty arose during spectrum analysis, and that the counting time was simply too low to be able to reliably quantify ^{137}Cs activity concentrations at the levels recorded.

3.2.6 Assessment of fitness for purpose for random measurement uncertainties.

A method to assess the Fitness For Purpose (FFP) of measurements, suggested by Ramsey *et al* (1992), was used to assess whether the Canberra, Exploranium and *ex situ* measurements might be considered FFP. The assessment criterion used is that the variance contributed by the measurement uncertainty should be less than 20% of the overall variance. These proportions are given in Table 5, showing that none of the measurements could be considered FFP for Zone 12. However, in Barrier 31, the Canberra *in situ* and the *ex situ* measurements for the 0-10cm layer can be considered FFP.

Table 5 Components of measurement uncertainty expressed as percentages of the total site variance, for the Canberra, Exploranium and *ex situ* surveys. Data reproduced from (Rostron et al., 2012).

		Contribution to total variance (%)		
		Sampling	Analytical	Measurement
Zone 12	Canberra *in situ*	0	92.5	**92.5**
	Exploranium *in situ*	53.9	56.2	**82.7**
	Ex situ 0-10cm	34.1	14.2	**48.2**
	Ex situ 10-20cm	54.6	5.0	**59.6**
	Ex situ 0-20cm	43.7	8.1	**51.8**
Barrier 31	Canberra *in situ*	0.5	0.3	**0.8**
	Ex situ 0-10cm	12.6	0.2	**12.8**
	Ex situ 10-20cm	43.9	0.1	**44.0**
	Ex situ 0-20cm	33.2	0.2	**33.4**

3.3 Systematic components of uncertainty.

3.3.1 Comparison of Canberra *in situ* and *ex situ* measurements.

Traditionally, *ex situ* measurements are considered to be the most reliable. This is partly because they are processed (e.g. dried and ground) in a controlled environment, and measurements are made using equipment that has been calibrated with standard (and often traceable) reference sources (or CRMs). However, *in situ* measurements can have significant advantages in time and cost. Of the four *in situ* methods used in these surveys, only the Canberra *in situ* measurements can be evaluated against the *ex situ* measurements, because a) the Canberra measurements were converted to activity concentrations, and b) the two measurement sets were acquired during the same time period, whereas the SEC survey activity concentrations were measured in 2003/4.

To examine whether the Canberra *in situ* measurements are good estimates of the average activity concentration over the two site areas, it is necessary to use non-parametric tests, as none of the measurement distributions were good fits to normal distributions, judged using a Ryan-Joiner test ($p<0.01$). For both surveys, a Mann-Whitney test for independent samples was performed to test for differences between the *ex situ* measurements and the Canberra *in situ* measurements that were made at the *ex situ* locations. No significant differences were found in the Zone 12 survey ($n=8$, $p<0.05$) or the Barrier 31 survey ($n=20$, $p<0.05$), suggesting that the median values of the *in situ* measurements were not significantly different from those of the *ex situ* measurements.

3.3.2 Systematic components of uncertainty in the Zone 12 measurements.

To test whether there is significant bias between Canberra *in situ* measurements and *ex situ* measurements, regressions can be performed to examine the relationships between the two measurements sets. For Zone 12. correlation coefficients (Pearson's r and the non-parametric Spearman's r_s) were found to be significant ($p<0.05$) for Zone 12 when the large outlying value at measurement location E11 (see Fig 3) was included. Spearman's r_s is still significant ($p<0.05$) but r is not significant ($p>0.05$) when the outlying value was removed from the dataset (Fig 5). Note that in this case the Canberra *in situ* measurements have been plotted against the *ex situ* measurements for the 0-10cm layer only, as *ex situ* soil samples at 10-20cm depth were acquired at only 8 out of the 20 *ex situ* measurement locations. Although neither the slope nor the intercept are significant ($p>0.05$), which would indicate that no significant bias exists between the two datasets, the non-significant correlation coefficient r and the apparent scatter in the data (Fig 5) may suggest that individual *in situ* measurements, which average small-scale heterogeneity over a large volume, are not good estimates of *ex situ* measurements in this case. This is probably a result of the high levels of random error in the Zone 12 measurements.

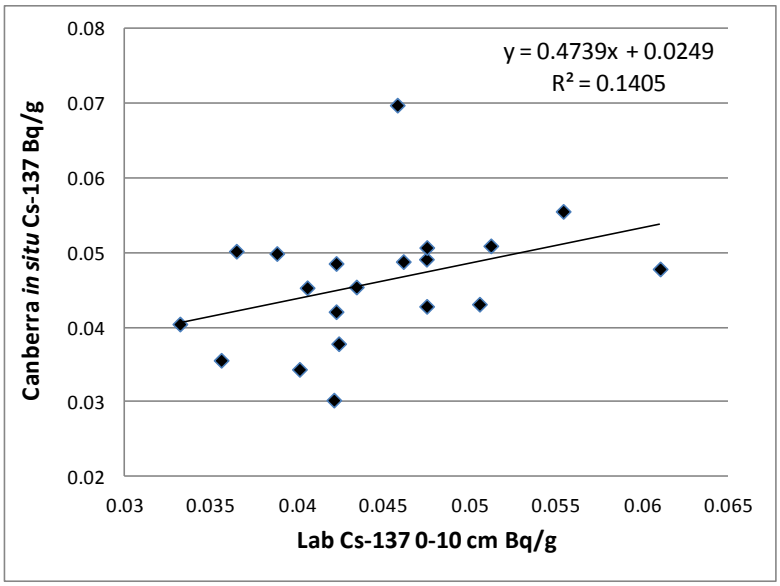

Fig 5 Relationship between Canberra *in situ* and *ex situ* measurements of ^{137}Cs activity concentration for Zone 12, for the 20 *ex situ* measurements acquired for the 01-10cm soil layer (Outlying value at measurement location E11 has been excluded). Reproduced from (Rostron et al., 2012).

3.3.3 Systematic uncertainty in the Barrier 31measurements.

Similar regressions of Canberra *in situ* against *ex situ* measurements for Barrier 31 were performed, both with the high outlying measurements at location C3 (Fig 4) included, and also with it excluded. In both cases r and r_s are significant ($p<0.05$), and the coefficients of determination ($R^2 = 0.777, 0.635$) show a fairly strong relationship between *in situ* and *ex situ* measurements. The intercept coefficient is not significantly different from zero, and the slope coefficient is not significantly different from unity ($p>0.05$), when the outlying value at C3 is excluded (Fig 6). This strong correlation and lack of significant bias suggest that the Canberra *in situ* measurements were good estimates of the *ex situ* measurements.

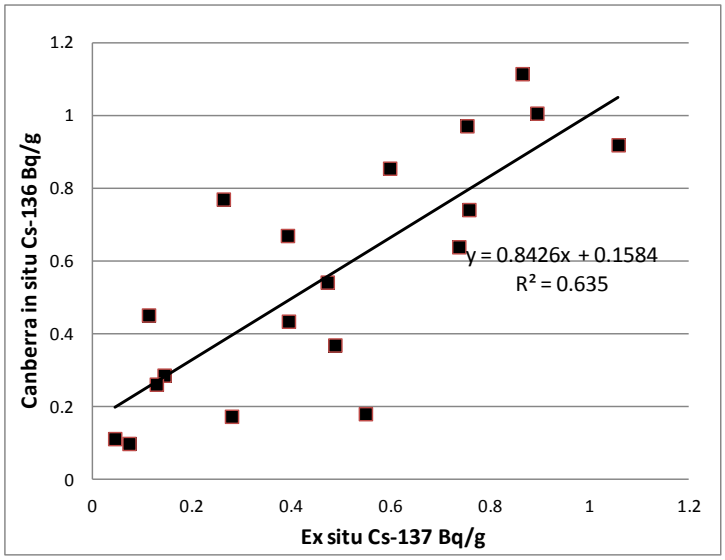

Fig 6 Relationship between Canberra *in situ* and *ex situ* measurements of ^{137}Cs activity concentrations in Barrier 31. Excluding the outlying measurement at location C3. Reproduced from (Rostron et al., 2012).

3.4 Shine from external sources

In situ measurements may be affected by shine from radiation sources external to the survey area, such as buildings and drains. There were no potential sources of external radiation apparent near the Zone 12 survey area, however, the Barrier 31 area was adjacent to both active drains and buildings. The most probable source of any significant radiation shine from an external source was from a silo containing historic waste, located immediately to the West of the survey area. It is clear that *in situ* measurements of activity concentrations show an increasing gradient from East to West towards these tanks. This effect could also be explained by higher actual activity concentrations in the ground, however. It would not be expected that shine would affect *ex situ* measurements. Further work needs to be done to investigate the issue of shine in this area, and how it could affect *in situ* measurements (Rostron et al., 2012).

3.5 Identification of hotspots of activity

The Groundhog system as used at Dounreay has been aimed at the *in situ* identification of hotspots of ^{137}Cs activity, which is a primary component of particle activity where this is found on the site. As radioactive particles may be buried beneath the ground surface, the ability of an *in situ* system to detect these particles can be influenced by attenuation due to overlying material. An *in situ* system of hotspot detection, then, may depend on identifying areas of activity that are elevated compared to their immediate surroundings, rather than direct comparison of activities or activity concentrations to an intervention threshold. Interpretation of a measurement set where the intention is to locate such elevated areas may be aided by interpolation techniques. (Mabit and Bernard, 2007) found that in trials of three different interpolation methods, ordinary Kriging was the best method for mapping fallout ^{137}Cs redistribution. Kriged maps of Groundhog measurements on zone 12 are shown in Figs 8 and 9. In each, five areas have been highlighted where Groundhog measurements appear to be concentrated into hotspots of approximately 0.5-1.5 m diameter. These are then overlaid by measurements from a) the 0-10cm *ex situ* soil samples (Fig 8) and b) Canberra *in situ* measurements (Fig 9). It can be seen that both techniques identified the elevated concentration at measurement location E11, but not the other four locations. In both cases this appears to be a result of the small (~1m) hotspots being missed by the sampling pattern. In the

case of the *in situ* measurements this is likely to be due to the low coverage factor (6.2% of ground covered by the field-of-view of the collimated Canberra detector). Similar results were found for the Exploranium data, which gave a clear indication of measurement point E11only. Investigation of SEC survey data revealed none of the hotspots found by Groundhog, however this survey was carried out in 2003/2004, so the radionuclide content of the site may have changed significantly in the intervening time period. It is not thought that any activities in the area were likely to have significantly affected the radionuclide content of the site between August 2009 (Groundhog) and May 2010 when the surveys were carried out by the authors.

As previous *in situ* data was not available for the Barrier 31 site, the same comparison could not be carried out here. It could be assumed that the high coverage survey performed with the Canberra detector, when 100% of the ground area was covered by the collimator's field-of-view (FOV), would be the more reliable method to locate spots of higher activity. However, when using an *in situ* method, a very small particle (e.g. size <10% of the FOV area) will yield significantly fewer counts within the detector volume if the particle is positioned at the periphery of the FOV, compared to a particle positioned in the centre of the FOV, due to differences in source/detector geometry. In Barrier 31, individual areas of significantly higher activity than their surroundings were suspected from the data used to create the dot map in Fig 3, which were subsequently confirmed to have elevated activity by two sub-surveys close to measurement points C3 and N6. The seemingly elevated concentration at point Q3 was not investigated. It is unlikely that the *ex situ* measurements as shown in Fig 4 would have prompted investigations of the area around N6 or Q3, but may have initiated investigation at location C3, as this was the highest *ex situ* measurement recorded. However, it is not known whether this outcome would have changed had the same number of *ex situ* measurements been made as *in situ* (Rostron *et al*., 2012).

4.0 Future work

4.1 Optimal *in situ* survey design for the location of hotspots.

When a regular square grid sampling pattern is used, the circular field-of-view (FOV) of a collimated *in situ* detector, subtended at the ground surface, must be overlapped between adjacent measurements in order to achieve 100% coverage of the survey area (Fig 7). This represents more than 100 percent coverage overall as there is overlap between the individual measurements. It should be noted that a triangular grid is a more efficient way to give complete coverage of the survey area.

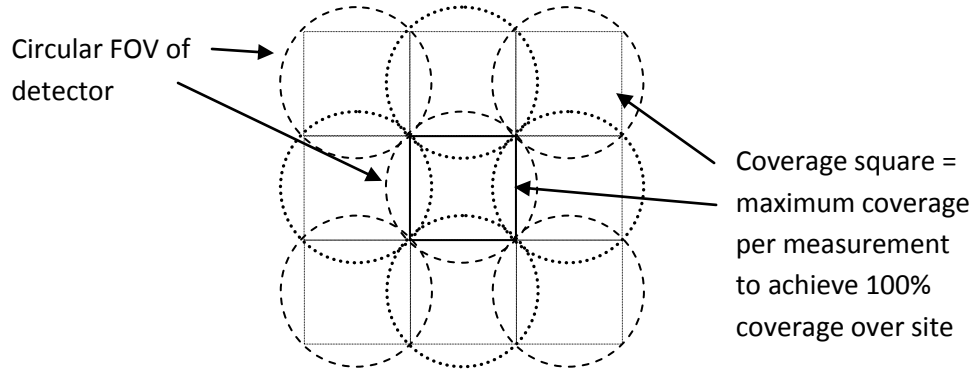

Fig 7 Illustrating the coverage required when using an *in situ* method with a circular field-of-view (FOV) to sample on a regular square grid, where 100% coverage of the ground surface is required.

The size of the FOV depends on two factors: a) the type of collimation used (e.g. angle of aperture) and b) the height of the detector above the ground surface, assuming that the collimator face is parallel to the ground surface. In order to locate a hotspot with a specified certainty, preliminary investigations using modelling techniques have indicated that in many cases it would be a more efficient use of time to increase the coverage factor of an *in situ* detector beyond the 100% coverage shown in Fig 7. This is because a small particle at the periphery of the FOV is not as easily detectable as a particle in the centre of the FOV. A possible future direction for investigation is an optimisation of the relationship between the size of the field-of-view, the degree of coverage, and the counting time when searching for small hotspots (i.e. hotspots that can be wholly contained within the field-of-view of the detector). It is suggested here that such an optimisation might be based either on a) fixed probabilities of obtaining false measurements; or b) on minimising the total **expectation of loss**. Minimisation of the expectation of loss, using a modified financial loss function, has previously been proposed

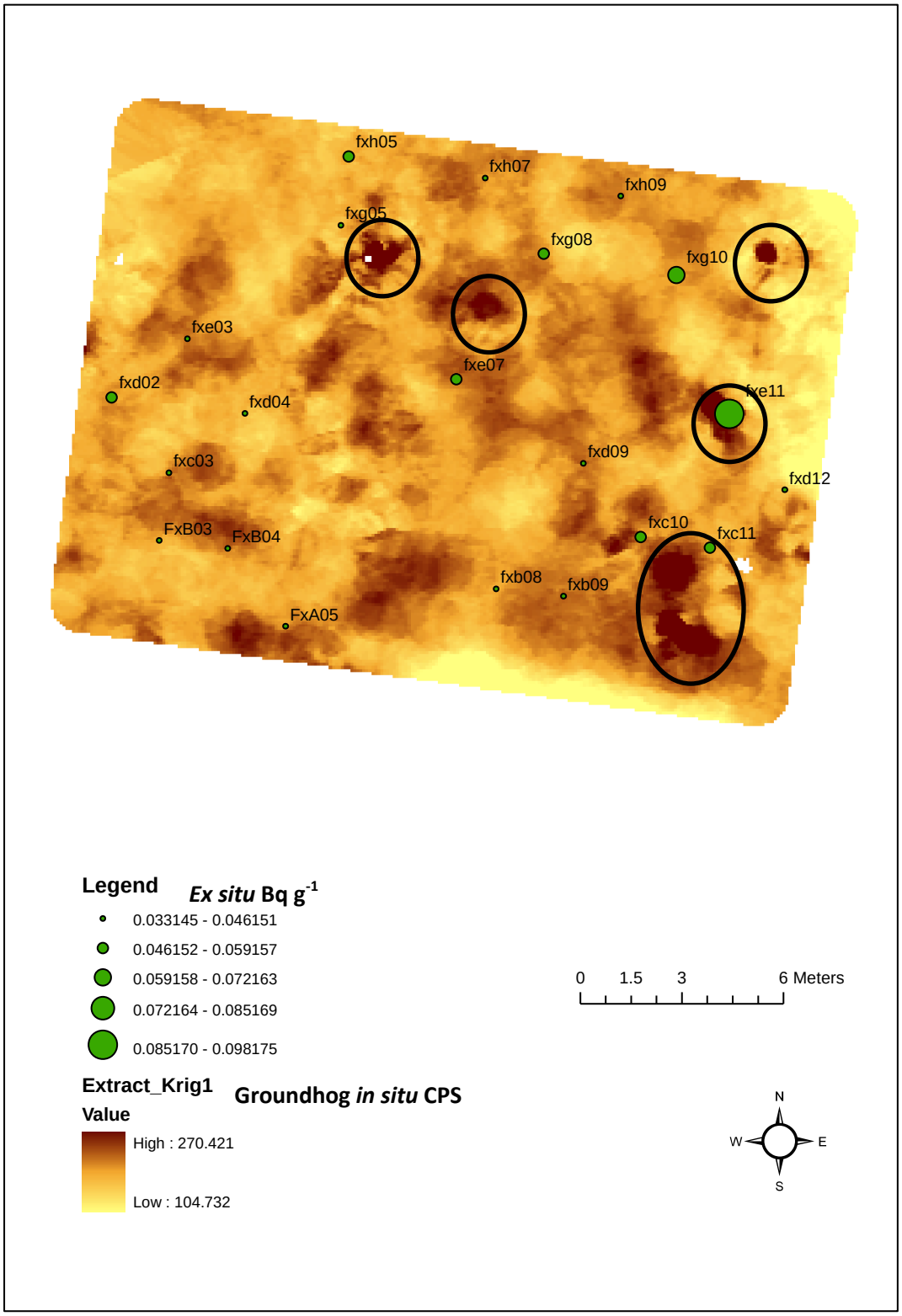

Fig 8 Krigged map of Groundhog measurements in the Zone 12 area, overlaid with proportional symbols (dots) representing ^{137}Cs activity from *ex situ* soil samples (0-10cm). The five black circles outline areas which appear to show the highest activity by Groundhog. Reproduced from (Rostron et al., 2012).

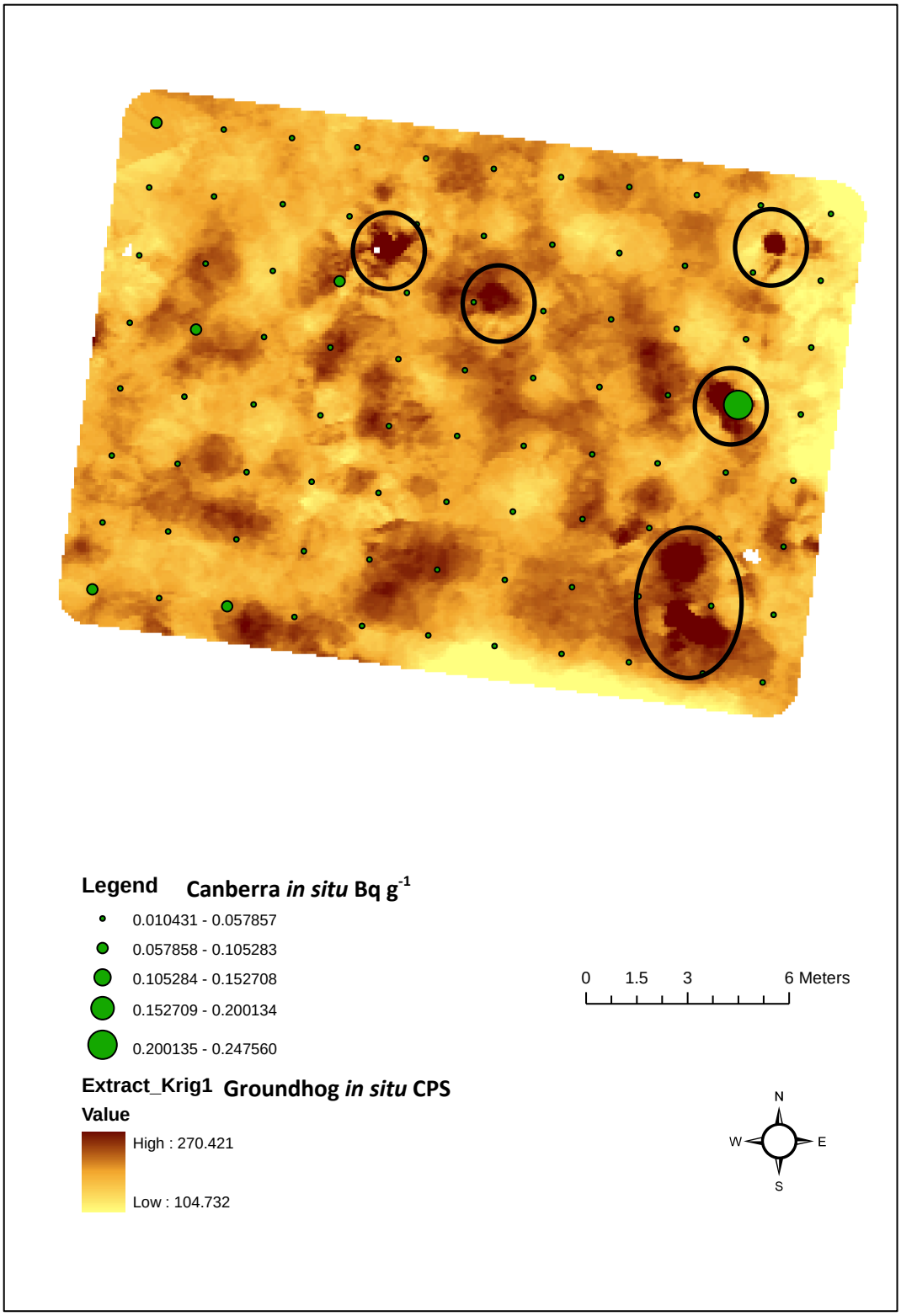

Fig 9 Krigged map of Groundhog CPS measurements in the Zone 12 area, overlaid with proportional symbols (dots) representing ^{137}Cs activity from Canberra *in situ* measurements. The five black circles outline areas which appear to show the highest activity by Groundhog. Reproduced from (Rostron et al., 2012).

as a method for evaluating whether surveys of chemically contaminated sites are fit-for-purpose (Ramsey et al., 2002, Thompson and Fearn, 1996).

5.0 Conclusion

The random components of measurement uncertainty were estimated for the two *in situ* and one *ex situ* measurement method used in the Zone 12 survey, and the single *in situ* and one *ex situ* methods used in Barrier 31. Estimates of random analytical uncertainty were also made for the Groundhog survey of Zone 12, and the site-wide SEC survey. Uncertainty in Canberra *in situ* measurements was significantly higher for Zone 12 (43.9%) than for Barrier 31 (12.6%), most likely due to proximity of the majority of the measurements to the MDA in the former case. Uncertainty in *ex situ* measurements was higher in Barrier 31 (72.6% for 0-20cm) than in Zone 12 (47.4%). Although the analytical uncertainty of *ex situ* measurements was lower in Barrier 31 than in Zone 12, there appears to be a much higher sampling uncertainty component. This is likely to be a result of the higher heterogeneity of ^{137}Cs activity concentrations in Barrier 31.

Using the fitness-for-purpose criteria proposed by Ramsey *et al* (1992), neither the Canberra, Exploranium or *ex situ* measurement methods would be considered fit for the purpose of describing the spatial distribution of ^{137}Cs in Zone 12. In the case of *in situ* measurements, extending the counting time to more than 10 minutes would most likely have reduced the analytical component of measurement uncertainty considerably. Extending the counting time of the *ex situ* measurements, which are more more severely affected by sampling uncertainty, would have a more limited effect on the overall measurement uncertainty. In both surveys, significant sampling uncertainty in the *ex situ* measurements is probably due to small-scale heterogeneity of ^{137}Cs activity concentrations. Expanded relative uncertainty for the Exploranium was estimated at 47% for ^{137}Cs and is approximately an even split of analytical and sampling uncertainty. More work would need to be done if there was a requirement to estimate activity concentrations using this detector.

Using the same FFP criteria for Barrier 31, the *in situ* and *ex situ* 0-10cm measurements would be considered FFP. This suggests that the 10 minute counting time used for the *in situ* measurements was adequate, given the levels of activity found in the area. Sampling uncertainty for the Canberra (10%) is very low compared to that typically found in chemically contaminated sites and this is likely to be a result the large sampling target arising from the remote detection of gamma-emiting radionuclides.

The estimate of analytical uncertainty for the Groundhog survey (approx 12%-19%) compares well to the other *in* situ methods used in Zone 12. As this system is aimed at identifying hotspots, and is intended to give 100% coverage of the ground area, it might be assumed that sampling uncertainty would be very low. Experiments would be needed to confirm this assumption. Estimates of the analytical uncertainty for the SEC survey, ranging from 53% to >300%. However, this estimate is somewhat tentative due to the reported accuracy of the positioning system used during the survey, and it might be significantly overestimated. The magnitude of this uncertainty suggests that the survey method used would not be adequate for characterising ^{137}Cs concentrations at the levels found in Zone 12 or Barrier 31.

Correlations between Canberra *in situ* measurements of ^{137}Cs activity concentration with *ex situ* measurements are not significant for Zone 12, except when the single high value at sampling point E11 is included. This is most likely due to the relatively high random uncertainty component of the total variance, which suggests that longer counting times would be needed for the *in situ* detectors to quantify reliably activity at individual locations. The same correlations are significant for Barrier 31, suggesting that here the *in situ* measurements were adequate for the purpose of characterising individual areas of the site. Based on the assumption that the laboratory measurements are reliable for small soil samples, the *in situ* measurements made by the Canberra detector appear to be good estimates of average activity concentrations over both sites. An additional factor of radiation shine from external sources was identified in the Barrier 31 *in situ* measurements, however, and this factor needs to be considered in any *in situ* investigation.

The *in situ* measurements taken, and those acquired by the previous Groundhog survey, suggest that potential hostspots of activity in the Zone 12 area tend to be small in extent, of the order of 0.5-1.5m. Comparison of the *in situ* and *ex situ* measurements in Zone 12 with previously acquired Groundhog data suggests that both methods were relatively inefficient at locating potential hotspots of activity, when compared with the continuous coverage obtained by the Groundhog vehicle. Of the three techniques used, *ex situ* soil sampling was found to give the weakest indication of the hotspot at sampling point E11, with the Canberra and Exploranium units both giving a strong indication of this hotspot. However, an apparent raised activity level was found by laboratory measurements of a soil sample from the 10-20cm soil layer at sampling point G10. When any radionuclide

contamination is located beneath the ground surface, attenuation of gamma radiation by the upper soil layers may prevent detection by *in situ* methods. In this case, an intrusive sampling method was the only means of detection, although longer counting times by the Canberra or Exploranium detectors may have identified raised activity levels thus prompting further investigation.

In Barrier 31, no previous continuous coverage data were available for comparison. As only 20 *ex situ* measurements were taken as opposed to 122 *in situ*, the two techniques cannot be convincingly compared for their suitability to hotspot detection. Hotspots again appear to be quite small in extent. It is therefore likely that *ex situ* soil samples taken on a regular grid (as per Fig 4) would miss some areas of raised activity. The continuous coverage provided by *in situ* measurements (e.g. Groundhog, or the Canberra measurements in Barrier 31, aided where necessary by interpolation), would appear to be the most reliable method of identifying small areas of raised activity.

This study suggests that the Canberra *in situ* detector can give estimates of activity concentrations for individual measurements, and for averaging areas, that are not significantly different to those obtained by *ex situ* surveys, providing a sufficient counting time is used. It is also likely that using a high-coverage *in situ* method will be more succesful at locating hotspots of activity at or near the ground surface. More work will be needed to invesitgate the relationships between counting time, coverage factor, angle of collimation and detector height above the ground surface, in order to devise a method which can optimise these parameters, based either on probabilistic criteria or an expectation of financial loss.

Although these conclusions have been drawn based on measurements of ^{137}Cs activity and activity concentrations only, it is probable that they will apply generally to other radionuclides that are detectable by gamma-ray spectroscopy.

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