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Comparison between *in situ* and *ex situ* gamma measurements on land areas within a decommissioning nuclear site: a case study at Dounreay.

Authors: Peter D Rostron¹, John A Heathcote² and Michael H Ramsey¹

¹University of Sussex, Falmer, Brighton, United Kingdom BN1 9QG.

²Dounreay Site Restoration Limited, Dounreay, Thurso, United Kingdom KW14 7TZ

Abstract. Measurements made *in situ* with gamma detectors and *ex situ* measurements of soil samples in a laboratory can have complementary roles in the assessment of radioactively contaminated land on decommissioning nuclear sites. Both *in situ* and *ex situ* methods were used to characterize ¹³⁷Cs contamination within an area at the Dounreay site in Scotland. The systematic difference (bias) between estimates of the mean activity concentration was found to be non-significant when *in situ* measurements were interpreted using a linear depth model, based on *ex situ* measurements made at two different depths. An established method of evaluating the random components of measurement uncertainty was used. The random component of analytical uncertainty in the *in situ* measurements, made in field conditions, was found to exceed that for the *ex situ* measurements, made in the controlled conditions of a laboratory. However, contamination by the target radionuclide was found to be heterogeneous over small spatial scales. This resulted in significantly higher levels of random sampling uncertainty in individual *ex situ* measurements. As *in situ* measurements are substantially less costly, a greater number of measurements can be made, which potentially reduces the uncertainty on the mean. Providing the depth profile of contaminants can be modelled with confidence, this can enable estimates of mean activity concentration over an averaging area to be made with lower overall uncertainties than are possible using *ex situ* methods.

1. Introduction

Regulation of radioactively contaminated land on nuclear licensed sites in the UK is the responsibility of the Office for Nuclear Regulation, an agency of the Health and Safety Executive (HSE). The immediate concern is risk arising from exposure to radiation of workers and/or the general public. This risk is managed via the nuclear site licence. Dounreay's mission is complete and the long-term ambition is to terminate the nuclear site licence. De-licensing of land areas requires the site licence holder to demonstrate that "there is no danger from ionising radiations from anything on that part of the site" (NIA, 1965). On areas that have been subject to radioactive contamination, this requires characterisation of the inventory and concentration distributions of contaminants in the ground (HSE, 2005; HSE, 2008).

Both *in situ* methods and conventional laboratory measurements of field samples (*ex situ* methods) can be used for land characterisation, and both are often used at the same site. When there is a high probability of gamma-emitting radionuclides existing at or near the ground surface, then *in situ* gamma-ray detection by scintillation or semi-conductor detectors can offer several advantages over the *ex situ* analysis of soil samples. *In situ* measurements are taken in real time and can be interpreted immediately, substantial cost savings can be made when compared to expensive laboratory procedures, and a larger sampling mass may give a more representative picture of the distributions of radionuclides over the site area. However, conversion of the activity counts recorded by an *in situ* detector into activity concentration units (e.g. Bq g⁻¹) requires knowledge of the depth distributions of contaminants. In contrast, *ex situ* measurements of samples acquired from different soil depths provides explicit information on depth distributions at discrete locations. It is usually necessary to use *ex situ* methods for the direct identification of alpha and beta emitters, because of the lower penetration of these types of emissions through intervening media such as soil and air. They may also be prescribed by regulatory authorities (IAEA, 1998).

Several studies have been performed where one of the objectives has been to compare *in situ* and *ex situ* gamma measurements of radioactivity in land areas. These have been performed in a variety of contexts, e.g. using distributions of fallout ^{137}Cs as a method of assessing soil erosion and deposition (He and Walling, 2000; Junjie et al., 2010); assessment of existing or novel techniques of obtaining reliable *in situ* measurements, considering radionuclide variability with depth (Baeza and Corbacho, 2010; Kastlander and Bargholtz, 2005; Korun et al., 1994; Korun et al., 1991; Tyler et al., 1996); experiments to assess the reliability of *in situ* measurements when compared to *ex situ* (Golosov et al., 2000; Macdonald et al., 1996; Sadremomtaz et al., 2010), and an inter-comparison exercise (Lettner et al., 1996). Some of these studies quote uncertainty in the measurements (both *in situ* and *ex situ*), while one (Golosov et al., 2000) reports an empirical estimate of the random component of uncertainty in measurements made *in situ*.

Both laboratory and field measurements of gamma-emitting radionuclides use the same physical principles for the identification and quantification of radionuclides. The use of gamma-spectra analysis to make quantitative estimates of specific radionuclides introduces uncertainties, such as Poisson variance in the source counts, and uncertainties in the interpretation of spectra. For *in situ* measurements, uncertainties also arise from the model chosen to convert raw detector counts to activity per unit mass or volume, while the response of the detector might be significantly affected by environmental conditions such as temperature and soil moisture content. When *ex situ* laboratory measurements of extracted soil samples are used, there are potential uncertainties in the internal geometries of the samples, and also in the responses of different detectors. In contaminated land investigations, it is important to obtain estimates of the magnitudes of these uncertainties, in order to be able to use the measurements to make reliable decisions. Uncertainty estimates can also be used to evaluate if specific measurement methods are fit for their intended purposes (IAEA, 1998; Ramsey and Argyraki, 1997; Ramsey et al., 2002).

Radioactive contamination in the general environment typically (although not always) arises from aerial deposition, which would be expected to result in relatively homogenous spatial distributions of contamination. In contrast, contamination on a nuclear site could arise from a number of different sources. It is often the case that concentrations of contaminants over the majority of the land area are relatively low, and may be close to the detection limits of the measurement systems. Both of these factors, i.e. heterogeneity of contaminant distributions and low contamination levels, are likely to increase the overall uncertainties in characterisation measurements. The objectives of this paper are to:

- a) Report on the systematic differences between *in situ* and *ex situ* measurements made on an area of radioactively contaminated land on a decommissioning nuclear site;
- b) Make empirical estimates of the random components of uncertainty;
- c) Discuss the implications of these findings to the estimation of mean activity concentrations and spatial mapping of contamination within the investigated area.

2. Methods

Caesium ^{137}Cs was chosen as the target radionuclide to illustrate the general issues. Although this radionuclide does not occur naturally, it is widespread in the UK following fallout from the 1986 Chernobyl incident. It is also a very important part of the contamination inventory from nuclear power generation. With a half-life of ~30 years, it is sufficiently long-lasting and radioactive to be present in measurable quantities more than one decade after its production. It is fairly easy to detect and identify using gamma-ray spectroscopy.

2.1. *Ex situ* (laboratory) measurements

Soil samples of approximately 0.25kg were extracted using a bulb planter from 0-100 mm and 100-200 mm depths at each *ex situ* sampling location. A stony layer was encountered between ~150-200mm, which set the practical maximum depth at 200mm using this method. Samples were analysed using HPGe detectors housed in 100 mm lead shielding with a quoted resolution of 2 keV (FWHM) at

1.33 MeV. Calibrations were performed using a geometry standard comprising a sample pot filled with dry soil, spiked with a certified mixed nuclide gamma standard. During the analysis, Q.C. checks were performed daily using a standard that included a known amount of ^{137}Cs . Background and interference corrections were applied by the counting software. Samples were analysed 'as received' with a counting time of three hours. Thus samples were measured in the laboratory at similar moisture contents to the field measurements. A subset of samples was selected, dried and the loss on drying recorded. These measurements indicated a typical moisture content of 34% by mass in the 'as received' samples. The soil was noted to be wet at the time of sample collection and this is considered to be the upper limit of expected moisture content.

2.2. *In situ* measurements

In situ measurements were made using a Canberra 3" × 3" NaI detector, fitted with a 90° lead collimator with a base thickness of 25 mm, wall thickness of 20 mm, and an internal aperture of 25 mm diameter. The collimator / detector assembly was positioned on the top shelf of a wheeled trolley. A circular hole of 115 mm diameter was cut into this shelf so as not to obscure the Field-Of-View (FOV) of the collimator. With the detector in place, the lower face of the collimator was measured at a height of 895 mm when the trolley was on a hard, level surface. This gave a detector height of 920 mm, which allowed full coverage of the ground surface by the FOV of the collimator to be obtained within a reasonable time-scale. The detector was connected to a laptop PC and spectra were recorded using Canberra's Genie-2000 software (Canberra, 2009a). All *in situ* measurements used a counting time of 600 seconds.

The spectra obtained from the Canberra detector were analysed using Genie 2000 software. Activity levels of ^{137}Cs were inferred from an energy peak located in the region of 662keV, which is a characteristic energy line in the decay of $^{137\text{m}}\text{Ba}$, a short-lived daughter of ^{137}Cs . Energy calibrations were performed during acquisition, using a ^{137}Cs source at the start of each half day, or more frequently if significant drift in the 662keV peak was observed. Interferences from energy peaks of other radionuclides may occur, in particular from the naturally occurring radionuclide ^{214}Bi , which emits an energy line at 609keV. The effect of ^{214}Bi peaks on estimates of ^{137}Cs activity is less than would be implied by a comparison of their activity concentrations, due to the lower emission probability of the 609keV energy line (46.9%) compared to the 662keV energy line (85.12%), and also because the peaks may not fully overlap. The spectra were checked during analysis for possible interferences, but Peaks at 609 keV were not clearly distinguishable from background noise. All measurements of ^{137}Cs were above the reported MDA calculated by the software.

2.3. Conversion of *in situ* activity counts to activity concentration units

Counts from *in situ* measurements require a calibration process to convert them to activity concentration units, because the source volume in *in situ* measurements is not clearly defined. This was achieved using ISOCS™ calibration software, which requires the definition of an appropriate source model (Canberra, 2009b). It also requires an assumption to be made about the depth distributions of target radionuclides. The *in situ* measurements were converted to activity concentrations using an assumption of a uniform depth distribution of radionuclides. This was based on comparisons between the *ex situ* samples at the two different soil depths. No significant difference was found between the mean activity concentrations of ^{137}Cs in the 0-100 mm and the 100-200 mm *ex situ* samples ($p > 0.05$, Student's paired t-test). Clearly this will not always be the case, and could potentially result in systematic error (bias) in the *in situ* measurements. Establishment of the depth profile to use in interpretation might be based on previous information about the site, but will often require collection of a number of *ex situ* samples at different soil depths in order to have confidence in the model. A number of methods have also been proposed which take the depth of activity into account when processing spectra from *in situ* measurements of soil (MacDonald *et al*, 1997), although these were not used in this survey.

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Changing the ISOCS model from 200mm to 500mm soil depth decreased calculated activity concentrations by ~3.3% and so this could be considered an additional component of uncertainty in the *in situ* measurements. Further modelling experiments showed that an insignificant amount of radiation at the 662keV energy level would be expected to reach the detector from depths beyond 500mm.

It might be expected that the source volume could be defined by the assumed maximum depth and the FOV of the collimator. This is incorrect. It has previously been found that even with a more effective 44 mm lead collimator, a significant amount of radiation will pass through the sidewalls, contributing 22-27 % of the instrument response (Kalb *et al.*, 2000). This is because the component of radiation passing through the side-walls of the collimator arises from a much greater expanse of ground than the relatively small area defined by the nominal FOV. A similar method was applied to define the source model dimensions with the 20mm collimator, by increasing the ISOCS model dimensions until response reached ~97% of the response from a semi-infinite planar source. It suggests that dimensions of 25 m diameter and 0.2m depth would be a reasonable assumption for the modelled source volume in the *in situ* measurements (figure 1), as increasing the model beyond these dimensions would have a relatively low impact on the conversion of counts to activity concentration units.

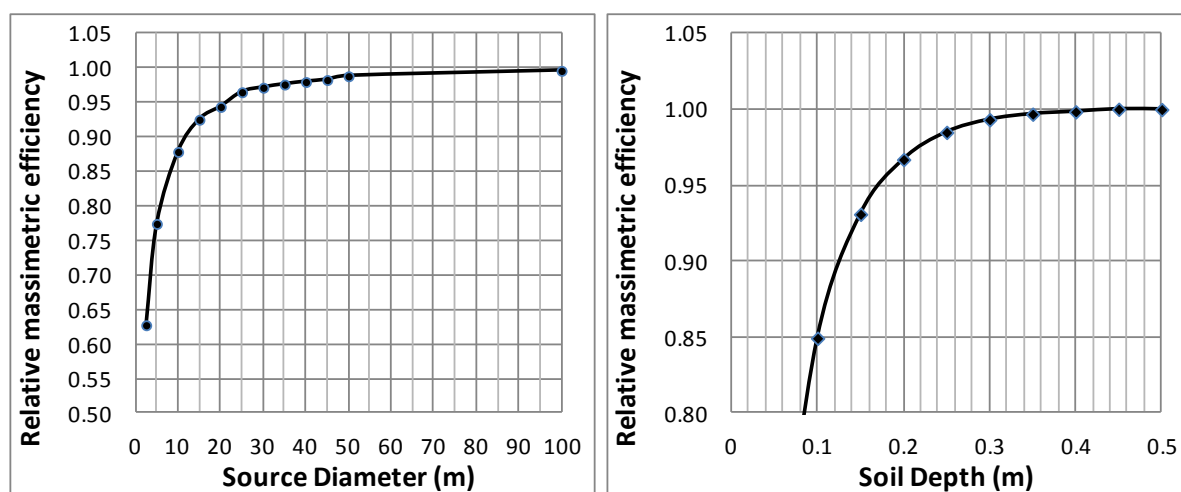


Figure 1. Relative massimetric efficiency (absolute efficiency/mass) calculated by ISOCS plotted against increasing model diameter (left) and increasing model depth (right). Assumed soil density = 1.7 g cm^{-3} and mass attenuation coefficient = $0.0788 \text{ cm}^2 \text{ g}^{-1}$ at 662 keV. Based on these results, an ISOCS source model of 25 m diameter and 0.2m depth was used in the conversion of activity counts to activity concentration units.

2.4. Estimation of measurement uncertainty

Measurement uncertainty arises from a number of different sources, and potentially at any stage from sample collection to the final reporting of measurement results. Much effort has been put into the reduction and evaluation of analytical uncertainty in laboratory measurements, but there is increasing awareness that sampling uncertainty is often the largest component of the overall uncertainty (Boudreault *et al.*, 2012; IAEA, 2004; Ramsey and Argyraki, 1997; Ramsey and Ellison, 2007; Taylor *et al.*, 2004). Sampling uncertainty can be considered to represent the uncertainty in measurements that arises when a primary sampling process (using the same nominal protocol) is repeated. While there may be additional components of sampling uncertainty arising from differences in interpretation of the sampling protocol, and the processes inherent in sample transport and preparation, it is usually the case that heterogeneity of contaminants is the dominating factor (Ramsey *et al.*, 2013). Land that is contaminated by radionuclides has often been found to exhibit high levels of contaminant heterogeneity (Dale *et al.*, 2008; IAEA, 1998; IAEA, 2011). It is therefore likely that sampling uncertainty will be a significant factor in radioactively contaminated land investigations.

Estimation of systematic uncertainty, or bias, is typically carried out in a laboratory by comparing measurements of standard reference materials with their known values. For field measurements, and in the absence of a manufactured reference source, the method most commonly used is to compare these measurements with laboratory measurements of extracted samples from the same locations.

The method used to estimate the random components of uncertainty was the duplicate method, as recommended by the Eurachem guide (Ramsey and Ellison, 2007). This has previously been used in studies of chemically contaminated land (Boon *et al.*, 2007). Duplicated samples are taken at a proportion of the primary sample locations, and duplicated analyses are then carried out on each in a balanced design hierarchy. Where applicable, the spatial distance between the duplicated samples at the primary locations is determined by an estimate of the spatial separation that would have occurred had the same nominal sampling protocol been repeatedly applied (Ramsey and Ellison, 2007). Applying ANOVA to the results allows separation of the different contributions of analytical and sampling uncertainties. If a small number of outlying values is encountered, robust ANOVA is able to give estimates that are more representative of the main population (AMC, 1989; Rostron & Ramey, 2013). The magnitude of the random component of uncertainty is expressed as the expanded relative measurement uncertainty (U), in which the relative standard deviation of the measurement uncertainty is multiplied by a coverage factor of 2 to give a confidence level of approximately 95 %:

$$U\% = 200 \times \frac{\text{standard deviation}}{\text{robust mean}}$$

(Ramsey, 2004):

2.5. Survey area

Dounreay has a history of experimental and commercial power generation since the 1960s. Decommissioning of the site has been in progress since the last reactor was shut down in 1994. The test site was located alongside a subterranean storage tank containing intermediate level radioactive waste (ILW). Due to its location, this area was not thought to have been significantly affected by authorized discharges, but it was known that some ground contamination had resulted from historic leaks from an active drain that runs between the ILW store and the surveyed area. It may also have been contaminated during discharges of material to the ILW store. Previous surveys had indicated moderate levels of contamination by ^{137}Cs , including the presence of radioactive particles. When found, these had been removed for authorized disposal. The soil in this area appeared to be a silty clay loam with organic content in the upper 100 mm. From knowledge of the local geology, the mineralogy was assumed to be non-calcareous, comprising mainly quartz and feldspar (Milodowski *et al.*, 2006). The ISOCS models for *in situ* interpretation were based on an assumed single homogenous layer of density 1.7 g cm^3 , using a standard non-calcareous soil definition provided by the program. This assumed a linear mass attenuation coefficient of $0.078832 \text{ cm}^2 \text{ g}^{-1}$ at 662 keV.

Measurements were made within a total area of $\sim 200 \text{ m}^2$, using a regular square grid pattern with 1300 mm between adjacent measurements (figure 2). The spacing was calculated to provide full coverage of the ground area by the FOV of the collimated detector, with a total of 120 *in situ* measurements. A proportion of these locations were assigned as duplicate measurement locations, corresponding to 10% of the total (i.e. 12) for the *in situ* measurements. To reduce analysis costs, 8 of these were selected as *ex situ* duplicate locations (figure 2), which has previously been suggested to be the minimum number of locations for reliable application of the duplicate method (Lyn *et al.*, 2007). All *ex situ* duplicate soil samples were excavated at points which spatially coincided with *in situ* duplicate measurement locations.

A common approach to positioning the duplicate measurement is at the extremity of the expected error in the positioning of the primary measurement. The expected absolute positional error of the Trimble differential GPS unit used in these surveys was of the order of a few cm, which was less than

the diameter of the soil sampling device. It was therefore decided to take the duplicate measurements at a distance from the primary measurement location equal to 10% of the measurement spacing (130 mm). This approach was used to assess the small-scale lateral heterogeneity of contaminants, and its effect on the sampling component of measurement uncertainty (Ramsey et al., 2002).

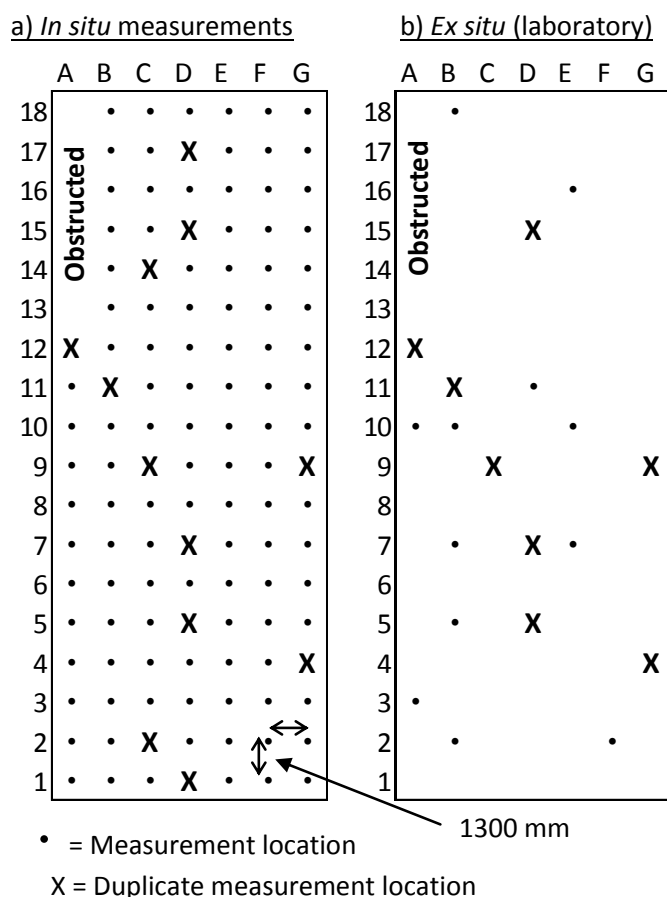


Figure 2. The survey grids showing the primary and duplicate measurement locations.

4. Results and discussion

4.1. Systematic differences between *in situ* and *ex situ* measurements

Measurement statistics obtained from both *ex situ* and *in situ* methods are summarised in table 1, which includes a calculation of the standard error on the mean (SEM). Maps of the measured activity concentrations using both methods are shown in figure 3. All measurements of ^{137}Cs were above the reported MDAs of the measurement systems.

Table 1. Summary statistics for both *ex situ* and *in situ* methods.

	No. of locations (N)	Mean activity concentration (Bq g^{-1})	Standard deviation s (Bq g^{-1})	SEM (Bq g^{-1})
<i>Ex situ</i> 0-100mm	20	0.67	0.83	0.186
<i>Ex situ</i> 100-200mm	20	0.53	0.46	0.103
<i>Ex situ</i> 0-200mm	20	0.60	0.64	0.143
<i>All in situ</i>	120	0.48	0.34	0.032
<i>In situ</i> at <i>ex situ</i> locations	20	0.63	0.44	0.098

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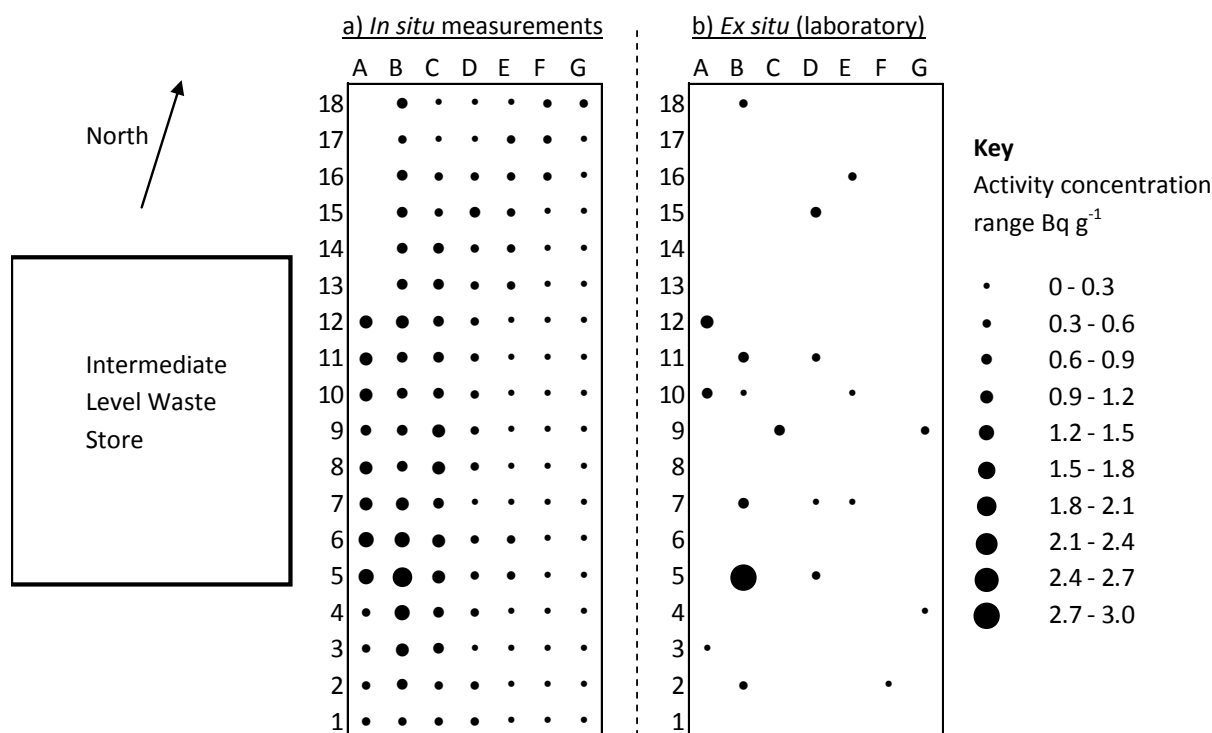


Figure 3. Dot maps (using equal divisions) for ^{137}Cs activity concentrations. a) *In situ* measurements, also showing the relative position of the ILW store; b) Laboratory measurements of *ex situ* soil samples, showing mean concentrations for the two sample depths. Values shown at the duplicate measurement locations (figure 2) are the mean values of the four measurements acquired at each.

The results in table 1 suggest that the mean of the 120 Canberra *in situ* measurements (0.48 Bq g^{-1}) differs from that of the 20 *ex situ* measurements calculated for the depth range 0-200 mm (0.6 Bq g^{-1}). There is no significant difference between the median values ($p > 0.05$), using the non-parametric Mann-Whitney test for independent samples. Examination of the data also indicates that the differences in mean values are largely caused by one comparatively high value measured at a single judgementally-positioned measurement location 5B (figure 3). This was incorporated into the survey design because prior information had indicated a raised activity level compared to the surrounding area. Given the substantial differences in sample volume between the *in situ* and *ex situ* measurements, and assuming that the raised activity level occurred in a small soil volume, a significant difference at this particular location might be anticipated. Removal of this outlying measurement from the data reduces the differences between the mean values (table 2). These measurement sets with the outlier removed are not significantly different from normality (Anderson-Darling, $p < 0.05$), and no significant difference between the means was found (Student's paired t-test, $p > 0.05$). This finding is supported by the much closer agreement between the mean of the *ex situ* measurements (0.6 Bq g^{-1}) and the mean of the *in situ* measurements (0.63 Bq g^{-1}) that were acquired at the same locations (table 1).

Table 2. Comparison of the mean of the *in situ* measurements with the mean of the *ex situ* measurements, after exclusion of the single judgmental measurement location.

	No. measurement locations (N)	Mean Activity Concentration (Bq g^{-1})
<i>In situ</i>	119	0.47
Mean <i>Ex situ</i> 0-200mm	19	0.47

A regression model of the *in situ* measurements against the *ex situ* measurements is shown in figure 4. The large differences in the sampling volume between the two methods limits the applicability of this approach, however in a systematic survey design, both methods are intended to represent the same sampling target. Estimations of the systematic differences between them are useful in evaluating the

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relative effectiveness of these methods for the purpose of spatial characterisation. The regression shown in figure 4 excludes the single high-value outliers from the judgmental measurement location 5B (figure 3). The slope is not significantly different from unity, nor the intercept significantly different from zero ($p > 0.05$), suggesting that there is no significant bias between the two methods with the outlying measurements excluded. This is partly a result of the scatter in the data, however, which may arise from the differences in sample mass combined with the relatively high level of the random component of uncertainty in the *ex situ* measurements.

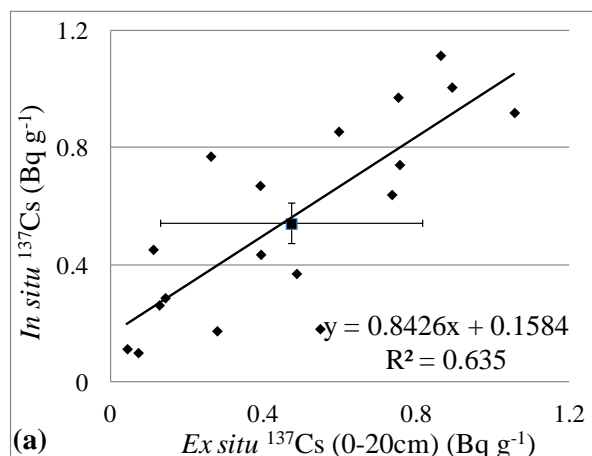


Figure 4. Regression of *in situ* against *ex situ* measurements of ¹³⁷Cs activity concentration. The error bars show the estimated random component of uncertainty (See Section 4.4) and for clarity are shown at a single point only.

4.2. Systematic differences due to shine

In situ measurements may be affected by radiation ‘shine’ from sources external to the survey area, such as buildings and drains. In such situations the use of more effective shielding would be desirable, however this introduces handling problems, particularly on rough ground or in less accessible locations. In contrast, the 20 mm collimator used in this survey weighed ~14 kg and it was possible for one person to handle it in the field. The survey area was adjacent to buildings and drains that historically contained active material. A subterranean tank partially filled with intermediate level radioactive waste was a possible source of shine, located to the left of the survey grid (figure 3). It is clear from figure 3 that the *in situ* measurements show an increasing trend towards this waste storage tank. *Ex situ* measurements made in a remote laboratory would not be affected by the radiation field from the ILW store, but would be expected to show any gradient in mass activity concentrations of ¹³⁷Cs in the soil itself. These measurements also show an increase towards the tank. An estimate was made of the potential effects of shine by creating an approximate ISOCS geometry model of the ILW and intervening absorbers, using an estimate of the activity and mean density of the material contained in the tank. It was then possible to estimate a value for the absolute detection efficiency with the collimated detector placed at the position closest to the centre of the ILW (location 10A, figure 3). This suggests that a very small component of the measured activity concentrations would have arisen due to shine from the ILW (< 0.1%), although the possibility of shine arising from the remaining structures cannot be ruled out.

4.3. Standard error on the mean

Ex situ measurements are usually considered to be more reliable than *in situ* measurements, because the primary samples are processed and measured with known geometries in a controlled environment, and also because measurements are made using equipment that has been calibrated with standard (and often traceable) reference sources. However, *in situ* measurements can have significant advantages in time and cost. These costs have been estimated for the case-study site in the region of £70 for a single

laboratory analysis and £8 per *in situ* measurement. From table 1, the standard error on the mean (SEM) for the *in situ* measurements is similar to that for the *ex situ* measurements, where equal numbers of measurements were used (*in situ* SEM = 0.1 Bq g⁻¹, *ex situ* SEM = 0.14 Bq g⁻¹ for 0 – 200 mm depth). When all the *in situ* measurements are used in the calculation, the SEM reduces to 0.03 Bq g⁻¹ at an estimated cost of £8 x 120 = £960, compared to the estimated cost of the 20 *ex situ* measurements of £70 x 20 = £1400. These estimates do not take into account the additional costs of analysing *ex situ* samples from different depths, or of empirical uncertainty estimates (e.g. by the duplicate method), both of which would significantly increase the *ex situ* cost. The improved precision in the estimate of the mean from the *in situ* measurements also has the potential to reduce the likelihood of unnecessary remediation.

4.4. Random component of measurement uncertainty

Empirical estimates of the random components of sampling and analytical uncertainties, estimated by the duplicate method, are shown in table 3 for both *in situ* and *ex situ* measurements. The combined measurement uncertainty has been calculated as the root sum squares of the sampling and analytical components. High levels of the combined measurement uncertainty for *ex situ* measurements were estimated (40-96%), which is mostly a result of the sampling component of uncertainty that has been separated by robust ANOVA. The analytical component of uncertainty (5.1%) is approximately consistent with the mean value of the Poisson uncertainty reported by the on-site laboratory (5.4% at 2s). Where a high level of sampling uncertainty exists, it is indicative of a high level of small-scale heterogeneity of contaminants. This has the potential to seriously affect the precision of individual *ex situ* measurements when used for spatial characterisation.

Table 3. Summary of the random component of measurement uncertainty.

	Expanded relative uncertainty (%)		
	Sampling	Analytical	Measurement
<i>Ex situ</i> 0-100mm	40.1	5.1	40.4
<i>Ex situ</i> 100-200mm	96.1	4.9	96.2
<i>Ex situ</i> 0-200mm	72.5	5.1	72.6
Canberra <i>in situ</i>	10.2	7.5	12.6

The analytical component of uncertainty for the *in situ* measurements (7.5%) is approximately 50% higher than for the *ex situ* measurements. This would be expected, because of the shorter counting times used, and as a result of recording measurements in field conditions. The sampling component, however, is much lower in the field measurements, resulting in a lower combined measurement uncertainty of 12.6%. This is most likely due to the significantly larger sampling mass that applies to the *in situ* measurements.

4.5. Implications of uncertainty for spatial characterisation

The lower random component of uncertainty in the *in situ* measurements compared with *ex situ* (table 3) suggests that *in situ* methods with a collimated detector may be more reliable than *ex situ* methods for spatial characterisation. A significant advantage of *in situ* measurements is that full coverage of the ground area by the FOV of a collimator is achievable at costs that are similar to or lower than the costs of a typical *ex situ* survey. However, the potentially large extent of the source volume needs to be taken into consideration, because a significant proportion of the detector response is likely to arise from a ground area that is beyond the nominal FOV of the collimator. In the case of the 20 mm collimator and detector height of 920 mm used in this survey, ISOCS modelling indicates that ~63% of the instrument's response would be expected to arise from a ground area that is 2.5 m in diameter, and ~94% from an area 20 m in diameter (figure 1). A 20 mm lead wall will permit transmission of approximately 9% of incident radiation at 662 keV, and so the significant proportion of detected radiation that passes through the collimator walls is more dependent on the increase in ground area

from which radiation may emanate and arrive at the detector (which increases by the square of the distance from the detector), rather than on small-scale heterogeneity, which might include a limited number of activity “hotspots” outside the FOV. Therefore the true proportion of the instrument response coming from inside the FOV will be mostly dependent on large-scale variability of activity outside the FOV in the vicinity of each measurement. It will be less affected by the presence of spatially small hotspots, especially when these are more than a few metres from the detector.

A simple method of reducing the source volume measured by a collimated detector is to reduce the detector height. Using a similar approach to that described in Section 2.3, and assuming the same soil density and attenuation coefficient, the diameter of the source from which ~95% of the instrument response arises with the collimator at a height of 12 mm was calculated as approximately 2 m. A practical approach to spatial mapping might therefore be an initial, high-coverage *in situ* survey with a collimated detector at a height of ~1 m in order to generate an overall picture of site variability, followed by targeted measurements using the detector at very low heights, and/or *ex situ* measurements in areas of interest.

4.6. Areas with very low activity levels

The use of the duplicate method of uncertainty estimation has also been demonstrated in an area of very low contamination, where the mean activity concentration was found to be $\sim 0.06 \text{ Bq g}^{-1} \text{ }^{137}\text{Cs}$ (Rostron *et al*, 2012). It should be noted that this survey used a lower height for the *in situ* detector, so full coverage by the FOV of the collimator was not achieved. When the duplicates obtained in this survey were analysed by robust ANOVA, it was again found that the *ex situ* measurements were significantly affected by sampling uncertainty, whereas this component of uncertainty was effectively absent in the *in situ* measurements. This was probably a result of a much more uniform distribution of contaminants over the larger scale. The area investigated in this case did not have a history of processing or storage of radioactive materials, but is thought to have been affected by aerial deposition. The existence of significant sampling uncertainty in the *ex situ* measurements suggests that contamination was heterogeneous on a relatively small scale, e.g. within the 200 mm sample duplicate spacing that was used. The combined measurement uncertainty was ~45 % for both methods, because of a larger random component of analytical uncertainty in the *in situ* measurements. This is most probably a result of Poisson variability in the relatively low count levels recorded by the *in situ* detector with a 600 s counting time. Taking these uncertainties into account, both measurement methods could be considered suitable for establishing that the mean activity concentration levels were below regulatory concern with a similar level of precision, but with a cost advantage to the *in situ* method.

5. Conclusions

In situ and *ex situ* surveys of an area of radioactively contaminated land were performed. A uniform depth model based on *ex situ* measurements at 2 different depths was used to estimate *in situ* activity concentrations. Systematic differences between the mean values of *in situ* and *ex situ* measurements to a depth of 200 mm were found to be non-significant in statistical tests. *In situ* measurements were able to give a lower standard error of the mean for less cost than *ex situ*, enabling higher precision of the mean value over the survey area. In surveys of this type, a number of *ex situ* measurements of extracted soil samples would usually be necessary to support *in situ* measurements, specifically for depth profiling information, and also to investigate the potential effects of shine from any nearby structures.

An empirical method of estimating the sampling and analytical uncertainties of individual measurements found that the random component of analytical uncertainty in *in situ* measurements of gamma emitting radionuclides with a collimated detector was higher than that obtained by *ex situ* measurements made in a laboratory, by a factor of ~1.5. However, the *ex situ* measurements were subject to high sampling uncertainty (40-96%), which has the potential to seriously affect the

precision of individual measurements. In contrast, sampling uncertainty for the *in situ* measurements was estimated at 10%. This suggests that *in situ* measurements with a collimator can enable an improved understanding of the overall spatial distribution of contamination, although transmission of radiation through the side walls of the collimator is a significant factor that must be considered in the interpretation.

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