Evaluation of uncertainties in *in situ* and *ex situ* gamma measurements on land areas with low contamination levels.

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Abstract.
Previous work on the characterization of land areas with moderate contamination levels showed that *in situ* measurements made with a gamma detector can achieve lower levels of the random component of uncertainty than laboratory measurements of extracted samples. This was found when the variance caused by small-scale lateral heterogeneity of contaminants was included in the uncertainty estimation. The present paper documents the results of applying the same techniques of uncertainty estimation to an area with contamination levels that were lower by a factor of 10. If the same counting times were used, it would be expected that both measurement types would be affected by higher levels of random uncertainty in the individual measurements because of increased uncertainty from counting statistics and other factors such as interpretation of gamma spectra. However, when uncertainty due to sampling was included, it was found that both measurements methods were subject to similar combined uncertainties at individual locations. Using an assumption of the depth distributions of radionuclides that was supported by *ex situ* measurements, *in situ* measurements were able to produce averaging estimates with an approximate reduction of 50 % in the standard error on the mean at ~50 % of the cost of the *ex situ* measurements.

1. Introduction

Surveys of land areas are carried out to satisfy the requirements of the site licence holder to ensure that risk constraints are not exceeded for workers. In the longer term, there is also the objective of releasing land areas from the terms of the site licence. Typically, a case for release would demonstrate that there are no detectable areas of localised contamination, and that the average level of contamination in the area concerned was less than some agreed value. Absence of localised contamination can be demonstrated only by total coverage surveys, either using a scanning detector or an appropriately designed survey with a stationary detector (Rostron *et al*, 2014).

Comparison of an area average with an assessment level requires a statistical approach. Methods for making such an assessment are well established (USEPA, 2000a; USEPA, 2000b) and software (Visual Sampling Plan, or VSP) is able to recommend a sampling grid based on tolerable error rates and the variance of the measurements (USDOE, 2014). Measurements on the sampling grid could be made by either *in situ* or *ex situ* techniques, or some combination. The confidence interval on the mean could be defined as:

$$\bar{x} \pm t_{1-\alpha,n-1}\frac{s}{\sqrt{n}}$$

where $\bar{x}$ is the sample mean, *t* is the t-distribution, α is the tolerable error rate, *n-1* is the degrees of freedom, *s* is the sample standard deviation and *n* is the number of measurements. The confidence interval can be reduced, permitting a less conservative assessment, by increasing *n* or reducing *s*. VSP solves for *n* given a confidence interval width and tolerable error rate α. *In situ* measurements are relatively inexpensive, making it possible to increase *n*, although they would be expected to have increased variance (s^2) compared with *ex situ* measurements.

When applied to an area with moderate contamination levels (~0.5 Bq g^{-1} ^{137}Cs), it was found that estimates of mean activity concentrations were not significantly different between *in situ* and *ex situ* methods, given necessary assumptions about the depth profile of contamination. Empirical estimates of uncertainties at individual measurement locations showed that *ex situ* measurements were severely

affected by small-scale heterogeneity. *In situ* measurements were much less affected by this factor, due to their larger spatial coverage. This has potential implications for spatial mapping of the lateral distributions of contamination (Rostron *et al*, 2014). In this paper, we examine the use of *in situ* measurements at lower activity concentrations, where making *in situ* measurements is potentially more challenging.

A significant factor in comparisons between measurements made *in situ* and *ex situ* is the differences in counting times that would typically apply. Once samples have been acquired and processed, laboratory measurements are largely "hands-off" operations carried out under controlled conditions. Counting times are then limited by costs and priorities only. Most laboratories support multiple instrumentation, and times of between 3 and 12 hours are often practically achievable. In contrast, *in situ* measurements are carried out during normal working hours in environmental conditions. They require at the very least a notional attendance by trained personnel, and counting times are likely to be measured in minutes, not hours. However, because no sample extraction, storage and processing is required, the use of comparatively short counting times at individual locations has the advantage that several *in situ* measurements can be made for the same cost as a single laboratory measurement. This means that a denser sampling pattern is possible within the same budget.

The use of shorter counting times implies that *in situ* measurements will be subject to a higher degree of uncertainty from counting statistics than *ex situ* measurements. Combined with uncertainties in measurement interpretation and making measurements in the environment, this means that individual *in situ* measurements are likely to be affected by significantly higher values of the random component of uncertainty than can be achieved using a laboratory detector. When applied to moderately contaminated land, it was found that this factor was offset by the reduction of sampling uncertainty (caused by small-scale heterogeneity) at individual measurement locations (Rostron *et al*, 2014). However, this raises the question of how the two measurement methods would compare on areas with contamination levels that are expected to be very low but that still need to be characterized in order to verify that they meet regulatory and local site requirements. In this case, the purpose of an investigation is primarily to verify that activity concentrations in the target area are sufficiently below regulatory thresholds that no further action is required to protect workers or the public, and that a longer-term case can be made for release of the land from the terms of the site licence. If fewer *ex situ* measurements can be used, and replaced with less expensive *in situ* methods, then this has the potential to reduce the total costs of land characterization substantially.

The objective of this study was to evaluate and compare the following uncertainties in *in situ* and *ex situ* gamma measurements on a land area with very low contamination levels:

1. Systematic uncertainty on the mean;
2. The random component of uncertainty at individual measurement locations;
3. Random uncertainty on the mean.

2. Methods

An unused area of land was selected at the Dounreay site in Caithness, Scotland. It had no previous history of processing or storage of nuclear materials, but had been subject to aerial deposition from authorized discharges to sea and air. It had also been used as a temporary storage area for rubble from demolished buildings, and so there was also the potential for some localized contamination to have arisen by this route. Previous high-coverage *in situ* surveys using vehicle mounted detectors had indicated the presence of a single spot of ^{137}Cs activity, that was thought to be below regulatory concern but which was elevated compared to the average activity on the site. The area was chosen to be representative of one that might be supposed to be free of contamination, either because of its usage history or post-remediation, but which requires demonstration that activity concentrations are consistent with regulatory objectives. This area was expected to be free of background caused by shine from adjacent areas.

In situ measurements were made using a Canberra 3" × 3" (76 x 76mm) NaI detector fitted with a 90°, 20 mm lead collimator. This was placed on a wheeled platform with the lower face of the detector at a height above ground of 280mm.Measurements were acquired at 88 locations on a systematically spaced survey grid, using a counting time of 600 seconds per measurement (Figure 1). The grid was marked out using GPS so that one location (J5) corresponded with the highest level of ^{137}Cs activity from previous high-coverage *in situ* surveys. The survey can thus be considered to include one judgmentally placed location.

Peak area analysis of the *in situ* measurements was carried out using Canberra's Genie 2000 software (Canberra, 2009a). This provided an estimate of the counts above background of the peak in the region of 662keV, which is a characteristic energy line in the decay of ^{137m} Ba, a short-lived daughter of ^{137}Cs. A calibration process was then required to convert peak counts to activity concentration units. This was achieved using ISOCS^{TM} calibration software, which requires the definition of an appropriate source model (Canberra, 2009b). ISOCS uses mathematical algorithms to calculate the absolute massimetric detection efficiency for the defined geometry (Canberra, 2001). A circular plane source of 20 m diameter and 200 mm thickness was used, since it had been found previously that a proportion of the detector signal can be expected to arise from outside the volume defined by the FOV of the collimator (Rostron *et al*, 2014). The use of a planar model can only be expected to provide an approximation of actual activity concentrations because it takes no account of variations in topography, surface roughness or overlying vegetation. This large diameter may also result in contributions from beyond the area of immediate interest.

Soil samples were acquired at location J5 and also at a total of 20 randomly assigned locations on the survey grid. These 20 samples are indicative of the sample density that might arise from DQO considerations, with a subsequent choice to collect the required number of samples randomly. The samples were taken from the top 100 mm of soil, using a coring tool, and stored in sample pots. Soil samples were subsequently analysed using HPGe detectors with a quoted resolution of 2 keV (FWHM) at 1.33 MeV, in Dounreay's laboratory. Calibrations were first performed with a sample pot filled with dry soil that was spiked with a certified mixed nuclide gamma standard. During the analysis, quality control checks were performed daily using a standard that included a known amount of ^{137}Cs. Background corrections were applied by the counting software. Samples were analysed 'as received' with a counting time of three hours. Moisture content was gravimetrically estimated to be approximately 30%.

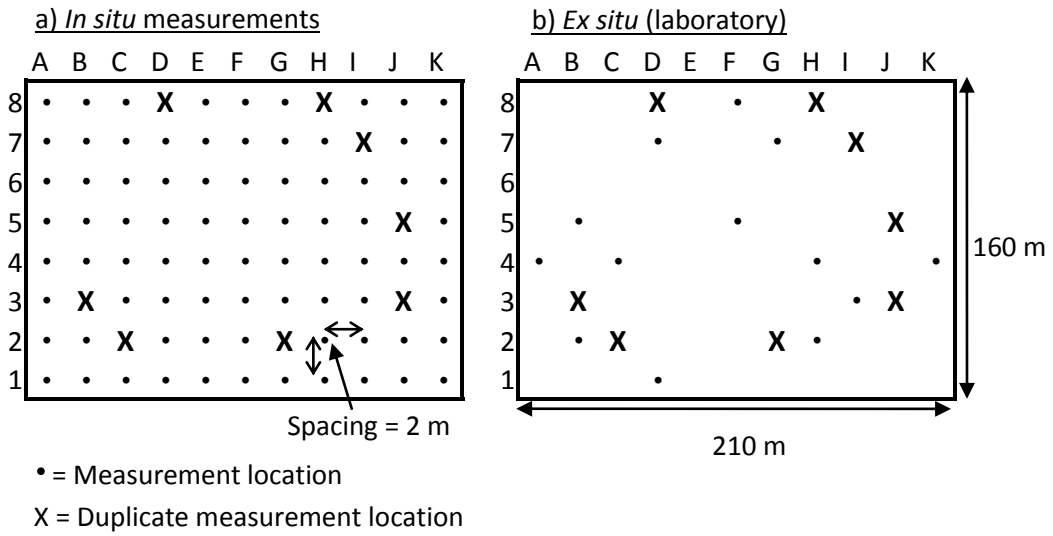

Figure 1. Measurement grids for *in situ* and *ex situ* surveys showing the primary and duplicate measurement locations.

In order to estimate the random component of uncertainty at individual measurement locations, eight measurement locations were randomly assigned as duplicate measurement locations. These have been represented by the symbol 'X' in Figure 1. The random component of uncertainty was estimated using the *duplicate method* (Ramsey and Ellison, 2007). At each of the duplicate measurement locations, a duplicate sample location was also assigned at a distance of 200 mm (10% of the measurement spacing) in a random direction from the primary location. At each duplicate measurement location, two *in situ* measurements were acquired at the primary location, and an additional two at the duplicate sample location. Additional soil samples were also extracted from the duplicate sample locations, and the primary and duplicate soil samples were both analysed twice in the laboratory. The random components of sampling and analytical uncertainty were then estimated using robust ANOVA (Ramsey et al., 2002). The estimated sampling uncertainty in this case can be considered to be an estimate of the variance in the measurement set caused by small-scale lateral heterogeneity of the analyte, while the analytical uncertainty is an estimate of the uncertainty that would be obtained by repeated measurements of the same sample.

An additional subset of soil samples was taken from the depth of 100-200 mm, or as deep as could practically be achieved before a stony layer was encountered. These were extracted from the duplicate measurement locations, both at the primary and duplicate positions.

3. Results and discussion

Dot maps of the *in situ* and the *ex situ* measurements are shown in Figure 2. Both maps use the same scale to represent the magnitudes of the measurements at individual locations. The elevated measurement at location J5 is clearly visible in both cases.

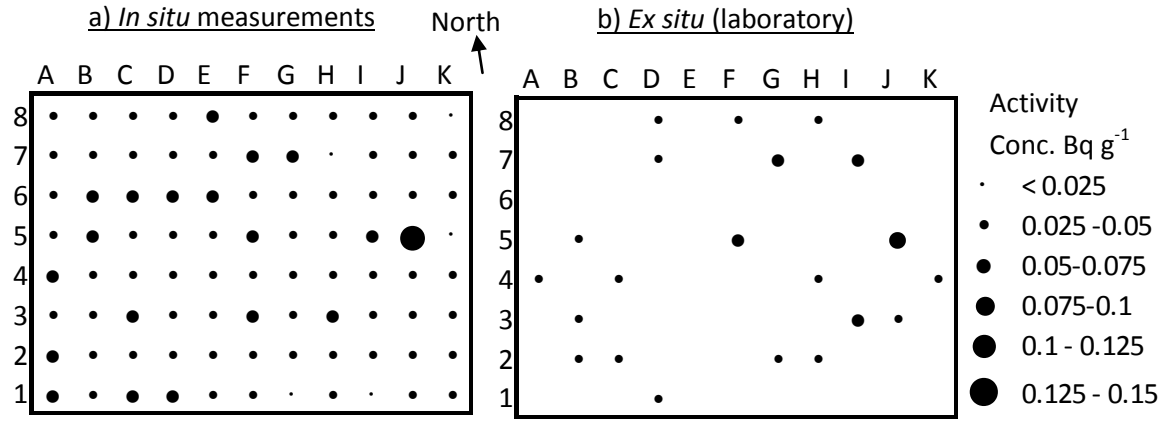

Figure 2. Dot maps (equal divisions) for ^{137}Cs activity concentrations. a) *In situ* measurements; b) Laboratory measurements of *ex situ* soil samples. The values at the duplicate measurement locations are averages of the four measurements acquired at each.

Statistics for both measurement methods are given in Table 1.

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})
Ex situ (0 – 100 mm)	21	0.047	0.013
In situ (Ex situ locations)	21	0.051	0.024
In situ (all)	88	0.043	0.015

3.1 Systematic differences between the means

None of the three measurement sets in Table 1 were found to be normally distributed (Anderson-Darling, $p<0.05$).Removal of the high measurement at the judgementally positioned location at J5 (Figure 2) reduced the differences between the means (Table 2). In this case the three measurement sets were normally distributed ($p<0.05$).The means of the upper (0 - 100 mm) and lower (100 – 200 mm) layers were found to be 0.045 Bq g^{-1} and 0.052 Bq g^{-1} respectively, using the 14 *ex situ* measurements that were extracted from the deeper soil layer after removal of J5 from the data (note that soil samples were not acquired at every *ex situ* location). Differences between these means were not found to be significant at the 95% confidence level using a paired 2-tailed t-test, $t(13) =-1.84$, $p=0.09$. This provides some justification for assuming a homogeneous distribution with depth, as was used by the ISOCS model in the estimation of *in situ* activity concentrations.

Table 2. Mean values excluding outlying value at location J5.

	No. of locations (N)	Mean activity concentration (Bq g^{-1})	Standard deviation s (Bq g^{-1})
Ex situ (0 – 100 mm)	20	0.044	0.007
In situ (Ex situ locations)	20	0.046	0.009
In situ (all)	87	0.042	0.010

Differences between the mean values of these sets (Table 2) were evaluated using the Student's *t*-test. The difference between the means of the *ex situ* measurements and the 20 *in situ* measurements at the same locations were not found to be significant using a 2-tailed paired *t*-test, $t(19)=0.76$, $p=0.46$. There was also no significant difference found between the means of the 20 *ex situ* measurements and the 87 *in situ* measurements using a 2-tailed independent *t*-test (equal variances), $t(105)=0.91$, $p=0.36$.

3.2 Random components of uncertainty at individual measurement locations

The random components of uncertainty at individual measurement locations were estimated using the duplicate method, the results of which are shown in Table 3. Although the sampling and analytical components of uncertainty were found to be significantly different between the two methods, it is interesting that the combined measurement uncertainty, evaluated as the root of the sum of the squares of these components, was approximately the same. The random component of sampling uncertainty has been calculated as near zero for the *in situ* measurements, while it is the dominant component of uncertainty in the *ex situ* measurements. This is a result of the substantially larger sampling volume in the *in situ* measurements, which has the effect of smoothing out the small-scale lateral heterogeneity of ^{137}Cs activity. The *ex situ* measurements, however, were severely affected by heterogeneity because of their small (~ 100 mm) spatial extent.

Table 3. Summary of the random component of measurement uncertainty (95% confidence).

	Expanded relative uncertainty (%)		
	Sampling	Analytical	Measurement
Ex situ (0 – 100 mm)	34	22	**41**
Canberra *in situ*	0	44	**44**

These patterns of random uncertainty contrast with uncertainties estimated for an area of land with 10-fold higher levels of ^{137}Cs contamination. In that case, the analytical components of uncertainty were found to be significantly lower at ~ 5% for *ex situ* measurements, and 7.5% for *in situ* measurements (Rostron *et al*, 2014). The sampling component was also found to be higher for the *ex situ* measurements, and significant sampling uncertainty was found in the *in situ* measurements in that case. When these uncertainties were combined by the sums of their squares, the overall uncertainty in the *in situ* measurements was found to be less (by a factor of three) than for the *ex situ* measurements for the depth range 0 - 100 mm. The much greater analytical uncertainty found in the current survey probably arises from a number of factors, including statistical variances in the recorded counts at very

low activity levels, and uncertainty in the peak area analysis when the full energy peak is not well defined because of the low activity compared to background. The lack of significant *in situ* sampling uncertainty is partly a result of lower heterogeneity across the site compared with the previous case, excepting the one high value found at location J5.

3.3 Random uncertainty on the mean

The Standard Error of the Mean (SEM) was calculated as the standard deviation divided by the square root of the number of measurements for the total surveyed area. It is an estimate of the random error of the mean values. Calculated SEMs are shown in Table 4, which also includes estimates of the survey costs based on approximate costs of measurements at the case study site.

Table 4 Standard errors on the mean and survey costs estimated for the case study site

	No. of measurements	SEM (Bq g^{-1})	Cost (£) per measurement	Total cost (£)
Ex situ	21	0.0029	70	1470
In situ (Ex situ locations)	21	0.0052	8	168
In situ (all)	88	0.0016	8	704

The results of this survey demonstrate a good agreement between the *in situ* and *ex situ* measurements for establishing the mean activity concentrations. Although the SEM for *in situ* was approximately twice that for *ex situ* (Table 4) when the same number of measurements were used, it was significantly reduced when a large number of measurements (88) was used. As the costs of *in situ* measurements are also significantly lower, this shows that an improvement in this uncertainty (~50% lower) can be achieved at a substantially lower overall cost, i.e. a 50% cost reduction in this case, even for a situation where activity levels are towards the limit of the capability of the detector used. Reduced uncertainty permits a more precise comparison with the assessment level.

3.4 General discussion

Positioning the detector at a height of 280 mm in this survey implies that the field-of-view (FOV) of the 90˚ collimator would have subtended a circular area of 560 mm in diameter at the ground surface. As the measurement spacing was 2 m, the proportion of the total ground covered by the nominal FOV would have been approximately 6 %, which compares very favourably with the very small area of land that would be investigated by traditional *ex situ* samples. This is substantially different from the previously reported survey on more contaminated ground where 100 % of the ground area was covered by the nominal FOV. A separate approach would be necessary to ensure the absence of localised hot-spots, but this does not need to be fully quantitative – it would be sufficient to show no activity above the background activity that has been quantified with the point measurements.

In this survey, both *in situ* and *ex situ* methods would have been suitable for the purpose of establishing that individual measurement locations and the mean activity concentrations were below 1 Bq g^{-1}, which has previously been defined as a lower level criterion for ^{137}Cs that is within the scope of the radioactive substances regulation in the UK (DEFRA, 2011), and below the 0.1 Bq/g level defined by IAEA (2004). The equivalence of the combined random uncertainties at individual measurement locations (Table 3) suggests that neither method has a particular technical advantage for the spatial mapping of the distributions of radionuclides in smaller areas, although in this case this is unlikely to be a priority as there is no requirement for extensive remediation. However, because it is possible to make a much larger number of measurements within the same budget, there is a potential financial advantage and/or an improvement in decision-making. The higher value at location J5 would require further investigation if identified by an *in situ* survey, in order to establish whether this was a result of a higher level of activity beneath the ground surface.

4. Conclusions

Satisfying the conditions of the Nuclear Site Licence will require surveys of land areas that are expected to have very low levels of contamination, in order to establish that no further action is necessary. This may include the use of both use *in situ* and *ex situ* measurements. Previous work comparing the uncertainties in measurements using *in situ* and *ex situ* methods in an area with moderate contamination levels by ^{137}Cs reported a lower combined uncertainty in the *in situ* measurements. The combined uncertainties in the current work have been estimated to be approximately equal. The difference between this and the previous survey has mainly arisen due to a much higher analytical component of uncertainty in the *in situ* measurements, resulting from low levels of contamination in the current survey.

The use of *in situ* measurements in conjunction with a small number of *ex situ* measurements has the potential to reduce the costs of land surveys, although other factors such as shine from nearby sources must be taken into account. Additionally, some *ex situ* measurements will usually be required to characterize the depth profiles of contaminants. It will be necessary to demonstrate that these can be expected to be consistent throughout a discrete area, which is likely to be the case if the contaminating process was fairly uniform (e.g. fallout). If this is the case then *in situ* measurements are able to provide a lower cost method of characterization, with similar levels of uncertainty on individual measurements, and substantially improved levels of uncertainty on estimates of the mean. This is because a larger number of measurements are possible within a defined budget. A reduction in the uncertainty of the mean potentially enables a more precise comparison with assessment levels.

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