^{137}Cs Measurement Uncertainties and Detection Limits for Airborne Gamma Spectrometry (AGS) Data Analysed Using a Spectral Windows Method

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Abstract
The uncertainties associated with Airborne Gamma Spectrometry (AGS) measurements analysed using a spectral windows method, and associated detection limits, have been investigated. For individual short measurements over buried ^{137}Cs activity detection limits of 10 kBq m^{-2} are achieved. These detection limits are reduced for superficial activity and longer integration times. For superficial activity, detection limits below 1 kBq m^{-2} are achievable. A comparison is made with the detection limits for other data processing methods.

1. Introduction

Airborne Gamma-ray Spectrometry (AGS) is a technique in which the distribution of radionuclides in the environment can be rapidly determined using sensitive γ-ray detectors mounted in low flying aircraft. Typically the detector would consist of 16 litres, or more, of NaI(Tl) scintillator, often with supplementary data from one or more germanium (Ge) semiconductor detectors. The technique depends on the penetrating nature of γ-rays in air, with, for example, the 662 keV gamma-ray from ^{137}Cs having a half distance in air of approximately 70 m. Thus, such radiation can be readily detected at ground clearances of ≈ 100 m or less.

A recent research project (Sanderson *et al*. 2001), principally funded by the Department of the Environment, Transport and the Regions (DETR) and other bodies, has been conducted to investigate some of the effects of survey parameters on data reproducibility. It was necessary for such an investigation that the uncertainties within the AGS method were more fully understood. To this end, the uncertainties in a small sample of data collected during a survey conducted in April 1999 as part of the project were analysed. This data was collected over a section of Rockcliffe Marsh in the Inner Solway near Carlisle, and calibrated using data from

Caerlaverock Marsh which had been extensively characterized by ground based sampling. The estuarine salt marshes of the Solway, and elsewhere along the Irish Sea, are contaminated by relatively high levels of ^{137}Cs discharged from the Sellafield complex since the 1950s. This activity is generally well buried, with the peak activity levels some 10-20 cm below the surface, resulting in an attenuation in full energy count rates compared to more superficial activity distributions. This results in the detection limits determined in this paper being approximately 3-4 times those that would be observed for measurements of freshly deposited ^{137}Cs activity.

The data from this project has been analysed using gross counts within defined spectral windows, from which background values and interferences between spectral windows are removed. The method is based on the IAEA Recommended Method for determining uranium, thorium and potassium activity concentrations (IAEA 1991, 2003), extended for measurements of anthropogenic nuclides by the definition of additional spectral windows (ICRU 1994). The resulting isotope specific count rates are calibrated using altitude correction and sensitivity constants determined from data collected over a site at Caerlaverock, Dumfries and Galloway, which was extensively sampled using a spatially representative expanding hexagonal pattern (Tyler *et.al.* 1996).

The spectral windows method used here is a simple, easy to program method that produces near instantaneous results. As noted, it is the method recommended by the IAEA, and is widely used. Recently, several other processing methods have been developed. These tend to be more computationally intensive and require more time to process survey data. Some of these are discussed in detail later.

2. Determination of Measurement Uncertainties

The spectral windows method for the analysis of AGS data is defined by the (IAEA 1991, 2003) and (ICRU 1994). Spectral windows corresponding to anthropogenic ^{137}Cs and ^{60}Co γ-rays and naturally occurring ^{40}K, ^{214}Bi (Uranium series) and ^{208}Tl (Thorium series) activity were defined. The windows used for this work are slightly modified compared to those recommended by the IAEA, including windows for anthropogenic nuclides and being slightly broader. The broader windows are used to reduce susceptibility to gain shift and resolution degradation, and to maintain continuity with earlier analysis procedures. Table 1 lists the spectral windows used

here, and those recommended by the IAEA and ICRU. For each measured spectrum the gross counts in each spectral window, *i*, were determined, and divided by the integration time to give a gross count rate, C_{gi}. Background count rates for each spectral window, C_{bi}, were determined from spectra recorded over open water, and subtracted from the gross count rate to give a net count rate for each window, C_{ni}.

$$C_{ni} = C_{gi} - C_{bi} \quad (1)$$

Scattering processes in the soil and air paths, and within the detector, result in a lower energy scattered background associated with each full energy photopeak. In addition, the U and Th decay series have a large number of different γ-rays associated with them. These result in interferences between spectral windows which must be subtracted from the net count rates. This is accomplished using a stripping matrix, $\boldsymbol{S}$, giving the fractional interference for each nuclide window in the other nuclide windows. It is determined from laboratory spectra recorded over a series of doped concrete pads with a number of perspex absorber sheets to simulate an airpath of approximately 70 m. The inverse of the stripping matrix is applied to a vector containing the net count rates in each of the radionuclide channels, $\boldsymbol{c_n}$, producing a vector containing the stripped counts in these channels, $\mathbf{c_s}$.

$$\boldsymbol{c_s} = \boldsymbol{S^{-1}c_n} \quad (2)$$

This is coded simply as a series of linear equations in which the elements of $\boldsymbol{c_s}$, C_{si}, are the sum over all radionuclide channels of the product of the elements of $\boldsymbol{c_n}$ and the elements s_{ij}^{-1} of the inverted stripping matrix.

$$C_{si} = \sum_j C_{nj} S_{ij}^{-1} \quad (3)$$

The stripping matrix does show small changes for different ground clearances, which could in theory be accounted for by coding the elements of the stripping matrix as functions of the ground clearance. In practice, this effect is reduced by tightly constraining the ground clearance of the survey flights, which over basically flat terrain is a simple task for the pilot. The stripping matrix would also only be valid for while the detector has the same performance (particularly

gain and resolution) as it had during the stripping measurements. Good quality control procedures, especially regular checks on detector performance and corrections as necessary, would minimise these effects. The effect of ground clearance and resolution on stripping has been explored more fully in Allyson and Sanderson (2001).

The final stage of data processing is to apply altitude correction, a_{ci}, and sensitivity calibration factors, s_i and s_i'. These were determined from measurements conducted over a salt marsh at Caerlaverock, Dumfries and Galloway. A spatially representative expanding hexagonal pattern (Tyler *et.al.* 1996) was used in extensively sampling this calibration site to characterise the levels and distribution of activity. The altitude correction uses an exponential relationship with the ground clearance, A, determined by radar altimetry to produce count rates, C_{ai}, normalised to a nominal ground clearance of 100 m. The use of data from a calibration manoeuvre eliminates systematic errors in the ground clearance measurements, for example miscalibration of absolute ground clearance.

$$C_{ai} = C_{si} e^{(a-100)a_{ci}} \tag{4}$$

The altitude normalised count rates are converted to calibrated activity per unit area (kBq m^{-2}) for anthropogenic radionuclides or activity per unit mass (Bq kg^{-1}) for naturally occurring radionuclides. If more than one calibration site is used a slope, s_i, and intercept, s_i', can be determined from a plot of altitude normalised count rates against activity concentration. The intercept terms allow for any systematic errors in the stripping due to differences between laboratory and field geometries. With only one calibration site, these intercept terms are set to zero.

$$A_i = s_i C_{ai} + s_i' \tag{5}$$

To determine the uncertainties on the calibrated activity per unit area or per unit mass measurements, the uncertainties associated with each stage of the analysis process are determined, and propagated through later stages of analysis. For a single measurement the uncertainty on the gross and net count rates for each spectral window are simply:

$$\Delta C_{gi} = \frac{\sqrt{N_i}}{T} \tag{6}$$

$$\Delta C_{ni} = \sqrt{\Delta C_{gi}^2 + \Delta C_{bi}^2} \quad (7)$$

For a 16 l NaI(Tl) detector gross count rates for naturally occurring radionuclides are typically 50-150 cps for ^{40}K and 10-30 cps for ^{214}Bi and ^{208}Tl. For ^{137}Cs count rates range from approximately 100 cps for weapons testing levels (less than 5 kBq m^{-2}) to 500 cps for Sellafield contaminated saltmarshes (typically above 100 kBq m^{-2}). Thus, for 2 s measurements, uncertainties on the gross count rates are typically 15% for uranium and thorium series activity, 7% for ^{40}K and 3-7% for ^{137}Cs.

Generally, by determining the background from several spectra at regular periods during a survey, the uncertainties associated with the background measurements are insignificant compared to the uncertainty on the gross count rate for a single measurement and can be neglected.

Assuming that the uncertainties on the elements of the stripping matrix are negligible, again as a result of the significantly longer measurement times of the laboratory measurements, the uncertainties on the stripped count rate are determined by propagating the uncertainty on the net count rates through the stripping matrix, yielding a linear equation from equation (3).

$$\Delta C_{si}^2 = \sum_j (\Delta C_{nj} s_{ij}^{-1})^2 \quad (8)$$

As the altitude correction coefficients are determined from several measurements in a hover manoeuvre, the statistical and systematic uncertainties in these are negligible. The calibration site is characterised by a large number of measurements in a spatially representative pattern. Although each of these measurements is relatively precise, the spatial variability that is unavoidable in a natural environment results in some considerable uncertainties in the overall activities per unit area or per unit mass determined for the site. Also, the calibration will only strictly be valid for that site, other areas in the environment where the distribution of radionuclides (particularly the depth profile) differs from the calibration site will be incorrectly reported using calibration coefficients determined on a site that is not representative of those environments. These uncertainties are systematic rather than statistical. Although they affect the

confidence with which a result may be reported, and hence need to be considered in reporting any data, they do not directly affect the ability of the technique to detect a given level of activity. So, neglecting the uncertainties associated with the ground based measurements of the calibration site, the statistical uncertainty associated with each measurement of activity per unit area is given by:

$$\left(\frac{\Delta A_i}{A_i}\right)^2 = \left(\frac{\Delta C_{si}}{C_{si}}\right)^2 \quad (9)$$

Thus, an assessment of the uncertainties associated with the stripped count rates (equation (8)) is sufficient to determine detection limits.

3. Detection Limits

The analysis presented here is conducted using the fractional uncertainty in the stripped count rate for measurements of ^{137}Cs activity determined from a set of data recorded over the southern side of Rockcliffe Marsh and adjacent areas in April 1999 using a 16 l NaI(Tl) spectrometer with a 2 s integration time (Sanderson *et.al.* 2001). As noted above, this is equivalent to analysis of the calibrated activities per unit area. This is an area with a radiation environment dominated by ^{137}Cs activity derived from marine discharges from Sellafield, deposited on salt marshes. Activities per unit area of approximately 50-100 kBq m^{-2} have been measured on the salt marsh, with much lower levels on the surrounding mud flats and terrestrial environments, resulting in a wide range of count rates within this small data set. The ^{137}Cs activity is deeply buried, with a depth profile showing a distinct sub-surface maximum at around 15 g cm^{-2} mass depth. The natural activity levels in the area are average for the area, with mean activities per unit mass of 263±68, 9.9±6.8 and 5.6±1.6 Bq kg^{-1} for ^{40}K, ^{214}Bi and ^{208}Tl respectively.

By summing the spectra from consecutive measurements spectra were produced with integration times of 4, 8 and 16 s. The relationships between the fractional uncertainty in ^{137}Cs stripped count rates and the ^{137}Cs activity for these four different integration times are shown in figure 1.

Though a rigorous treatment of detection limits is possible, such treatments usually relate to

uncertainties in the background measurement (e.g. see Currie 1968 or Martin 2000) which in this case are very much smaller than those determined for the measurement due to the very much longer integrated live time for the background measurements. For this work, a realistic estimate of the detection limit is all that is required given that the actual detection limit for ^{137}Cs depends considerably on the local natural activities, source geometry and whether other anthropogenic radionuclides are present. From figure 1, it is simple to determine the activity per unit area that would result in any given level of uncertainty. A fractional uncertainty in the stripped count rate of approximately 30% is approximately equivalent to a signal level 3σ above zero. This would be slightly larger than the detection limit L_D defined elsewhere (Currie 1968, Martin 2000) as a 95% confidence level. For this work, this simple assessment of detection limit suffices, and hence has been used.

The detection limits determined for the different integration times are given in table 2. These detection limits are very high, which is largely due to the radiation environment from which the measurements were made. The ^{137}Cs activity on the salt marsh is deeply buried, less deeply buried activity would result in reduced uncertainties for a given activity. In this environment, the detector records a stripped count rate of approximately 3 cps per kBq m^{-2} of ^{137}Cs activity, whereas during an international exercise in Finland with a much shallower activity depth profile with a mean mass depth of 1.31 g cm^{-2} (Sanderson *et.al.* 1997a) gave a stripped count rate of approximately 10 cps per kBq m^{-2} (Sanderson *et.al.* 1997b). It is, therefore, to be expected that uncertainties and detection limits for superficial deposition of activity would be reduced by a factor of approximately 3-4 compared to those presented here.

Another substantial component of the uncertainty in the stripped count rates is the use of a stripping matrix that includes a window for ^{60}Co radiation, even though there is no significant quantity of ^{60}Co in the environment. This spectral window is included in the routine analysis as a result of earlier survey work, where ^{60}Co was present. The very small net signals in the ^{60}Co window result in negligible reduction in the stripped count rates in other spectral windows, though the increased uncertainty in those windows is slightly more significant. Removing this element from the stripping matrix would result in slight improvements to the uncertainties. However, it should be noted that there are many situations in which an additional anthropogenic radionuclide could be present in the environment in which case a spectral window and associated stripping matrix elements would be needed.

In addition, the interpolation routine used to generate radiometric deposition maps combines several data points, hence further reducing the fractional uncertainty for each location by increasing the effective integration time at each measurement point. Thus, for fresh airborne deposition of ^{137}Cs activity a detection limit of 2-3 kBq m^{-2} should be possible for spectra recorded using a 16 l NaI(Tl) detector using a 2 s integration time and less than 1 kBq m^{-2} for 16 s integration times, depending on background levels and assuming no interference from other anthropogenic radionuclides.

4. Comparisons with other data processing methods

The spectral windows method described in this paper is one of several methods for analysing spectra recorded by AGS systems. Other methods include spectral profile analysis (Guillot 1996, 2000, 2001) and Noise Adjusted Singular Value Decomposition (NASVD) (Hovgaard 1998, 2000).

The Peak Isolation Method (PIM) is a spectral profile analysis that uses a digital filter to account for the characteristics of the absorption peaks, and hence reduce statistical fluctuations. Analysis of such methods has given a detection limit for ^{137}Cs of approximately 1.2 kBq m^{-2} at one standard deviation, for natural backgrounds (Guillot 2001). Comparisons between the results of analysis of data using the spectral windows method and the PIM for data collected during international exercises has shown that the two methods produce highly comparable results (Guillot 2001, Bourgeois *et.al.* 2003), though at lower levels of contamination the PIM resulted in greater statistical uncertainty.

The NASVD method extracts a set of spectral components containing all the spectral information from the entire data set. Linear combinations of these components are produced to reconstruct the measured spectra allowing for a significant noise reduction. A pseudo-concentration method based on this method mixes the important components to exclude contributions from the natural background to give a synthetic spectrum for ^{137}Cs, or other artificial nuclides. This method has been used to successfully estimate ^{137}Cs Equivalent Surface Concentrations to below 1 kBq m^{-2} (Aage *et.al.* 1999). The Equivalent Surface Concentration is defined as the concentration of entirely superficial ^{137}Cs that would generate the same flux of 662 keV gamma rays in the detector as the actual source. It is always a smaller number than the

activity per unit area of the measurement site, because the ^{137}Cs present in the environment is never entirely superficial. The conversion between ESC and activity per unit area requires a knowledge of the depth profile of the measured activity.

It should be noted that in both of these cases the definition of detection limit differs from that used in this paper, and would give slightly higher values if the definition used here was applied.

5. Discussion and Conclusions

The uncertainties associated with AGS measurements analysed using a method based on spectral windows and stripping out spectral interferences has been investigated. It has been shown that for individual short measurements over buried ^{137}Cs activity, analysed using a simple spectral windows method using fairly wide spectral windows, detection limits, corresponding approximately to the 3σ above zero signal level, of 10 kBq m^{-2} are achieved. However, these detection limits are reduced for superficial activity, longer integration times, and denser measurements (from a reduced line spacing). For freshly deposited activity, detection limits for ^{137}Cs of 2-3 kBq m^{-2} would be possible for 2 s integration times. Detection limits below 1 kBq m^{-2} should be possible with integration times of 10-15 s, although with some loss in spatial resolution compared to shorter integration time measurements.

The contribution to the uncertainty due to the stripping of spectral interferences is a significant contribution to the overall statistical uncertainty in the final activity concentrations determined by this method. This contribution should be reduced if narrower spectral windows, with consequently smaller interferences, were used. However, to use narrower spectral windows greater control of the spectral gain and resolution would be needed to ensure the windows are always appropriate. Additional reductions in the uncertainty could be achieved by not using spectral windows that correspond to radionuclides not present in the environment. In the work presented here, a window for ^{60}Co has been used even though there is no evidence of ^{60}Co in the environment studied. It should be noted, however, that in the event of an accidental release of radioactive material spectral windows for additional radionuclides will probably be needed.

Other spectral analysis methods used for AGS data show similar detection limits to the simple windows stripping methods, after accounting for differences in the definition of detection limit

and deposited activity concentration reported in other work.

Clearly, an objective assessment of the different processing methods used for AGS survey data would need to include a comparison of detection limits. The recent ECCOMAGS Intercomparison Exercise (Sanderson *et.al.* 2003) collected a considerable quantity of data from different systems, but the majority of the data was analysed using the spectral windows method, with limited use of other methods. There is scope for future work to process subsets of this data set, and other appropriate data, using several methods with comparable calibration assumptions and definitions of detection limit, to more rigorously compare these methods. A full spectral exchange format has been defined (Guillot 2003) to facilitate the necessary production and exchange of such test data sets at a future date.

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Table 1: Spectral windows (keV) used in this study and recommended by IAEA (IAEA1991, 2003)

Nuclide	This study	IAEA
^{137}Cs	544-749	
^{60}Co	1009-1245	
^{40}K	1319-1629	1370-1570
^{214}Bi	1629-1927	1660-1860
^{208}Tl	2374-2932	2410-2810

Table 2: Detection limits for ^{137}Cs activity on Rockcliffe Marsh for 2, 4, 8 and 16 s integration times

Integration time (s)	Detection limit (kBq m^{-2})
2	9.9
4	6.9
8	4.9
16	3.5

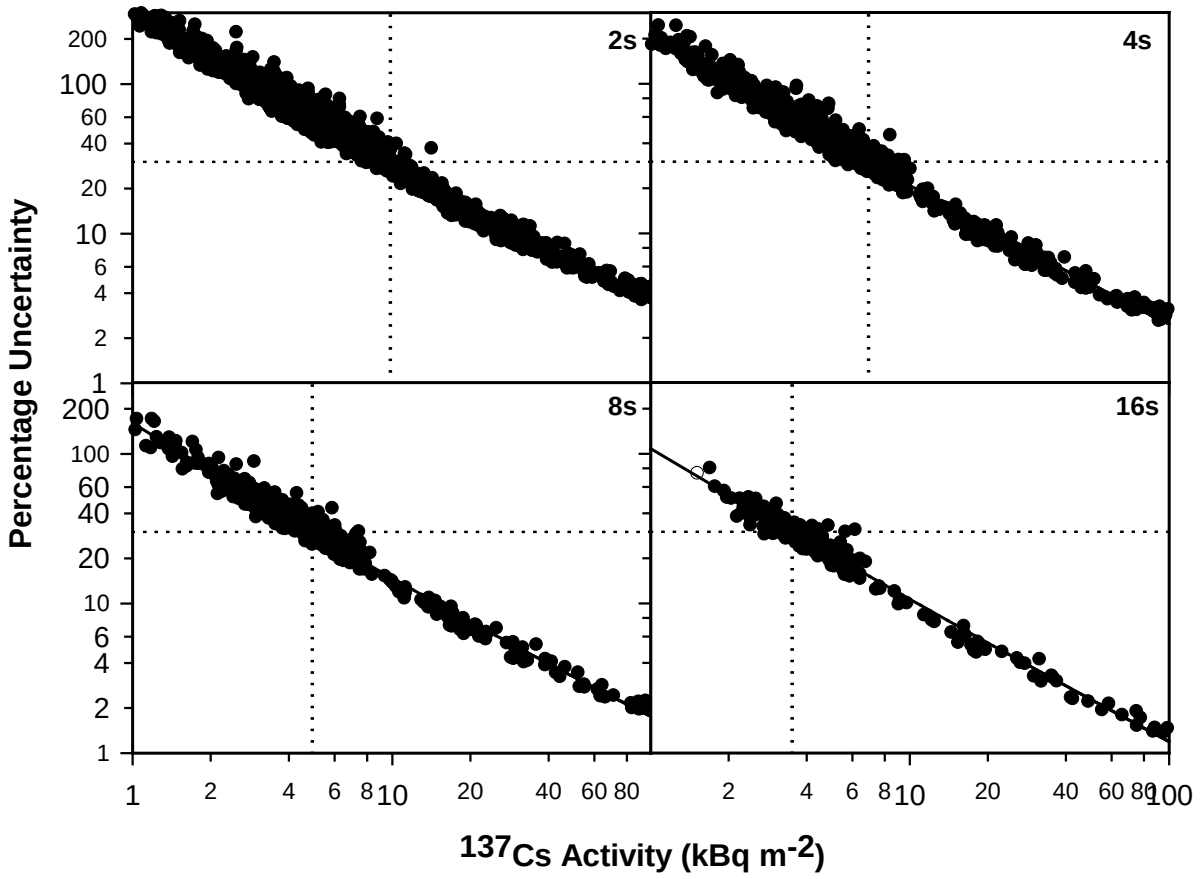

Figure 1: Percentage uncertainties on ^{137}Cs stripped count rates for 2, 4, 8 and 16 s integration times.