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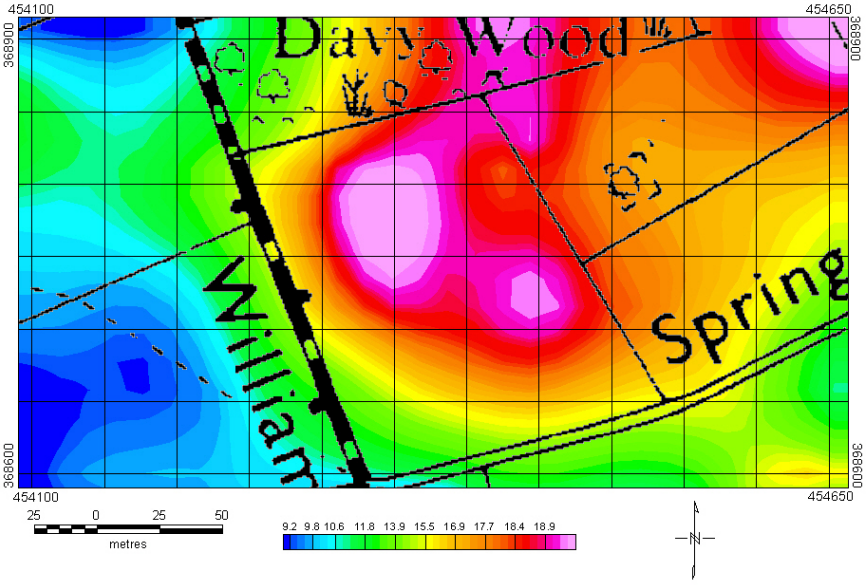
Airborne and ground-based radiometric investigation of colliery spoil near Shirebrook, the English Midlands

National Geoscience Framework and Chemical and Biological Hazards Programmes

Internal Report IR/05/059



BRITISH GEOLOGICAL SURVEY

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INTERNAL REPORT IR/05/059

Airborne and ground-based radiometric investigation of colliery spoil near Shirebrook, the English Midlands

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Keywords

Gamma spectrometry, HiRES-1, colliery spoil, natural radioactivity.

Front cover

Total counts per second data from the GTK survey at Warsop Vale.

Bibliographical reference

C EMERY, J R DAVIS, E HODGKINSON AND D G JONES. 2006. Airborne and ground-based radiometric investigation of colliery spoil near Shirebrook, the English Midlands. *British Geological Survey Internal Report*, IR/05/059. 60pp.

Keyworth, Nottingham British Geological Survey 2006

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Foreword

This report was produced as a contribution to the National Geoscience Framework and Chemical and Biological Hazards Programmes (2005-2010). Most of the work was carried out under the Environmental Radioactivity Project of the former Environmental Protection Programme and the HiRES-1 Project in the former Sustainable Energy and Geophysical Surveys Programme (2000-2005).

Acknowledgements

The authors would like to acknowledge all those who contributed to funding, organising and executing the HiRES-1 and GTK surveys. The surveys provided valuable airborne radiometric data sets, upon which this report is based.

Thanks to the East Midlands Development Agency, who very kindly provided access to the site at Shirebrook and provided background information on site history and reclamation. Thanks also go to the Coal Authority who kindly provided access to the site at Warsop Vale. In addition, the authors would like to thank the Warsop Vale Local History Society who provided useful background information.

Many thanks to Neil Breward for contributing to the geochemical description in section 1.2 and to the BGS staff who provided sample preparation and X-Ray Fluorescence data.

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Summary

As a result of the UK's industrial past, spoil heaps are widespread through our environment and encompass the waste products from a variety of different industrial processes.

Two high-resolution airborne geophysical surveys, conducted over parts of the UK in 1998 and 1999, identified four radiometric anomalies in the East Shirebrook region corresponding to four areas of colliery spoil. These areas which are composed of shales, siltstones and sandstones extracted during mining mixed with left over coal displayed elevated potassium, uranium and thorium, relative to their surroundings.

Ground follow-up work was undertaken at Shirebrook and Warsop Vale, the two disused collieries from the four radiometric anomalies identified by the airborne surveys in the East Shirebrook area. Active extraction ceased at these mines in the late 1980s and early 1990s and both have been undergoing reclamation and re-landscaping. The ground survey entailed ground-based continuous gamma spectrometry traverses over accessible parts of each site, ground-based static gamma spectrometry measurements over transects of the sites, surface soil sampling at these transect points for laboratory XRF analysis and short core sampling at Warsop Vale for gamma spectrometry analysis on a core logger. The ground survey confirmed, and more accurately delineated the anomalies observed by the airborne surveys. The ground-based continuous gamma spectrometry highlighted the marked difference between the radiometric signature of the spoil heaps and neighbouring agricultural land. A high degree of spatial variation was apparent in the ground-based gamma spectrometry data and highlighted that the spoil itself is not a uniform mass.

Ground survey static gamma spectrometry results and XRF analysis of surface soil samples correlate significantly. Comparison of these surface soil samples (0- 15 cm) obtained at both Shirebrook and Warsop Vale to regional Geochemical Baseline Survey of the Environment (G-BASE) surface soil samples obtained up to 1 km from the spoil tips showed that thorium was the element most elevated above background.

The radiometric data from the two airborne surveys correlate at the 99.95 % level for uranium, thorium and potassium. The values from the Geological Survey of Finland (GTK) airborne survey, flown in 1999 are slightly higher however, and this is probably the result of using the Finnish calibration range with very different rock types to those found near Shirebrook. The two surveys showed very similar patterns of radioactivity. They both distinguish well between areas of spoil and adjacent agricultural land. The higher spatial resolution (closer line spacing) of the GTK survey delineated the anomalies with a higher degree of detail. The GTK survey flying height was also lower than that of the High Resolution Airborne Resource and Environmental Survey Phase 1 (HiRES-1) airborne survey flown in 1998, thus reducing the sampling area, or 'footprint' from which the gamma radiation was averaged. This too increased the level of detail visible in the GTK survey results.

The extent of the spoil tips, as indicated by the radiometric data, does not always coincide exactly with the mapped boundaries of the spoil on Ordnance Survey 1: 50 000 or 1: 25 000 maps. This suggests that, in the absence of up-to-date maps, or where spoil extends beyond the mapped boundary, the radiometric data may be used to accurately define the current extent of the spoil. This may usefully be linked to EM data that show the extent of conductive (and potentially contaminated) groundwater (e.g. Beamish, 2002b, Klinck et al, 2004).

Although reclamation of the two former collieries has been successful in terms of the appearance of the site and in the return of wildlife to the areas, it appears that the radiometric anomaly still exists post-reclamation. However, the uranium concentrations encountered are well below Generalised Derived Limits and the maximum external annual gamma dose is less than 50 μSv.

There does not appear to be a significant radiological hazard to members of the public from an external dose perspective.

1 Introduction

Two high-resolution airborne geophysical surveys were conducted over parts of the UK in 1998 and 1999. As a result of these surveys, described in the following sections, areas were identified for further study. A ground follow-up survey was conducted in the Shirebrook area during late 2001 and 2002, which is the subject of this report. In order to set the results from these surveys in context, after a résumé of the airborne surveys, a brief geological and geochemical description of the area is given.

1.1 AIRBORNE SURVEYS

1.1.1 THE HIRES-1 SURVEY

The High Resolution Airborne Resource and Environmental Survey Phase 1 (HiRES-1) was flown between May and September 1998 acquiring 50, 434 line-km of data covering approximately 14 000 km^2 of central England (Peart *et al*., 2004) (Figure 1). The survey acquired 1024 channel gamma spectrometer, magnetic and Very Low Frequency ElectroMagnetic (VLF-EM) data. The aircraft flew at a height of 90 m in 'open' areas and at 240 m over developed zones. Flight line separation was generally 400 m with tie line spacing at 1, 200 m, except over three infill areas of special interest where flight line and tie line separations were effectively reduced to 200 m and 600 m respectively.

The HiRES-1 airborne survey area was chosen originally as it, amongst other things, encompassed areas of relatively high radioactivity related to industrial processes and radioactive waste.

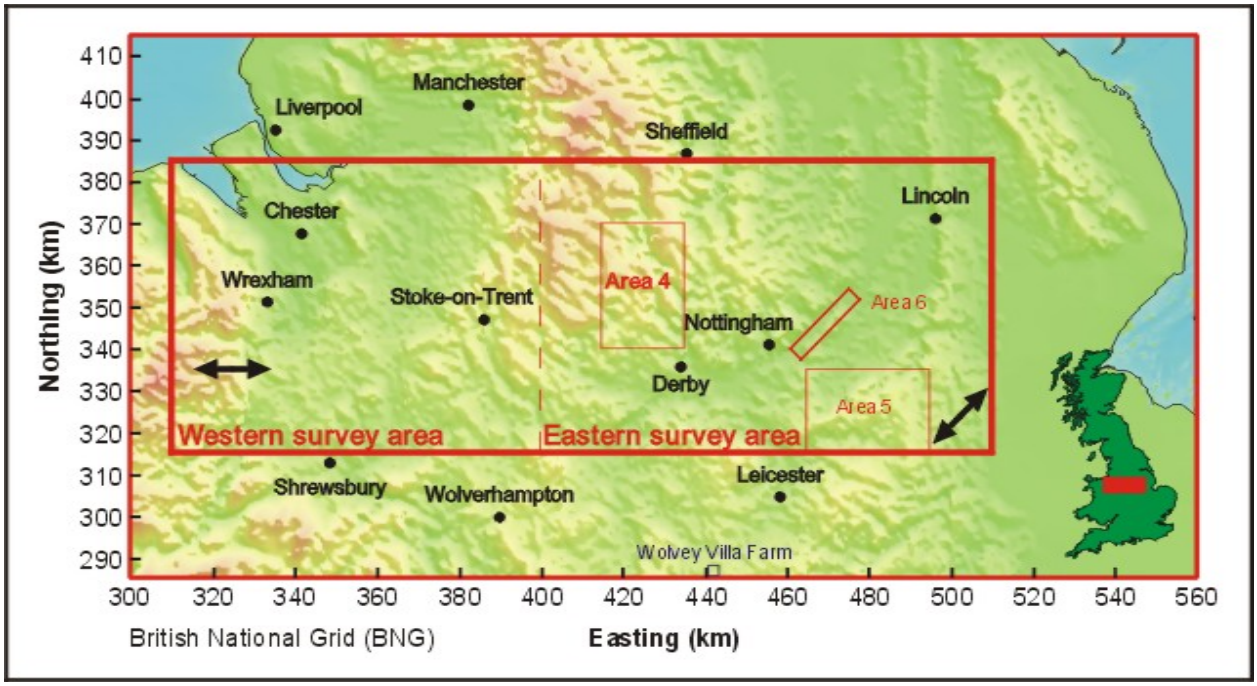


Figure 1 - The HiRES-1 survey area in central England (Area 4 is the Derbyshire dome infill area, Area 5 the Melton district and Area 6 the Trent Trough. All were chosen for more detailed study due to observed radiometric anomalies. Black arrows on diagram indicate the flight line direction in the eastern and western survey areas.)

Complete technical specifications of the equipment and systems employed during the airborne survey can be found in the survey logistics report by World Geoscience (UK) Ltd (2000). Outlined below are the specifications of the gamma ray spectrometer installed in the survey aircraft, Short Skyvan VH-WGL.

Gamma Ray Spectrometer

Model	-	Picodas PGAM 1000 Ver. 6.11
Detector Volume (downward)	-	33.56 litres
Energy Channels Recorded	-	1024 Individual Channel Data
	-	256 Channel (summed) Spectrum
Lower Energy Threshold	-	180 KeV
Cycle Rate	-	1 Hz (equates to about 70 m traverse distance)

Navigation and subsequent data location were achieved using a differential global positioning system (DGPS).

In the case of all the gamma spectrometry measurements it should be noted that an equivalent uranium (eU) value is determined from the ^{214}Bi gamma peak and an equivalent thorium (eTh) value is determined from the ^{208}Tl gamma peak. This is because adequate gamma peaks cannot be recorded directly from ^{238}U or ^{232}Th themselves, so daughter products are used to determine an equivalent value and equilibrium with the parent U or Th is assumed. This assumed equilibrium, however, may not always hold true due to the differing geochemical behaviour of the members of each decay chain. ^{226}Ra is a member of the ^{238}U decay chain and has a half-life of 1600 years. Radium's geochemical behaviour is markedly different from uranium, being generally much less mobile in the oxidising secondary environment. This property may therefore lead to uranium being removed during weathering, leaving radium. Therefore the ^{214}Bi gamma peak could indicate higher eU values than the ^{238}U levels actually present. Due to the long half-lives involved, should ^{238}U be separated from its daughter products, it would require about 1 million years to achieve 91% of secular equilibrium, in which state the daughter products are produced at the same rate that the ^{238}U decays. In contrast, the time taken for secular equilibrium to be achieved in the ^{232}Th decay series is about 70 years (Ball *et al*., 1991). This results in the ^{208}Tl gamma peak providing a more reliable estimate of the ^{232}Th values as the decay chain is more often in equilibrium.

Man-made sources, such as ^{137}Cs and ^{60}Co, can also be identified and quantified. The depth of investigation of the technique is restricted to the ground surface layer to a depth of about 30cm.

1.1.2 The GTK Survey

A series of 4 trial airborne environmental surveys were conducted during a one-week period in June 1999. The survey was flown by the Geological Survey of Finland (GTK) in conjunction with the British Geological Survey (BGS). The four areas chosen for this survey (Figure 2) were:

A) Shirebrook, due to the colliery spoil heaps and the shallow Sherwood Sandstone aquifer which is vulnerable to contamination from percolating fluids;
B) The Trent Valley, due to numerous landfills in abandoned gravel pits containing power station fly-ash and domestic waste;
C) Wolvey Villa Farm, due to a well documented contamination plume in a shallow and thin sandy aquifer;
D) Langar, which contains two landfill sites in limestone quarries but has no known pollution problem.

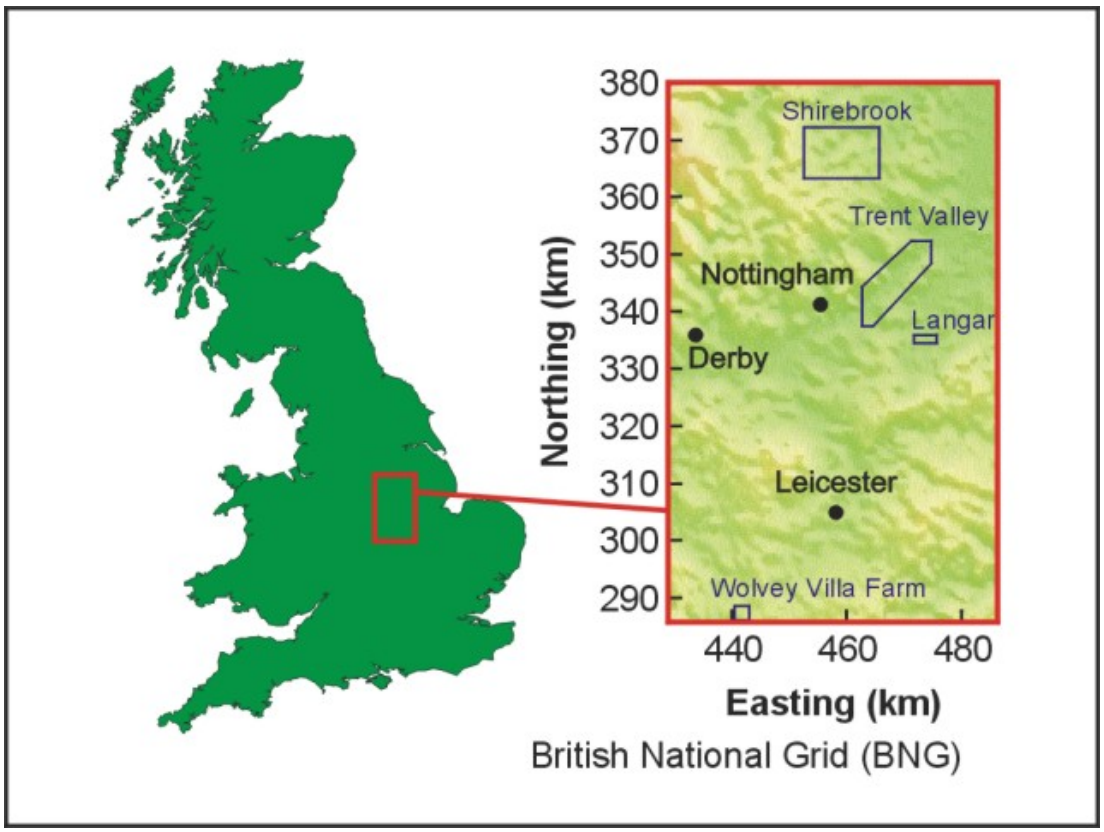


Figure 2 - The GTK survey areas: Shirebrook, Trent Valley, Wolvey Villa Farm and Langar

Like the HiRES-1 survey, radiometric, magnetic and electromagnetic data was acquired. Full technical details of the survey are outlined in Beamish *et al*. 2000c and d, but are briefly summarised below:

Data acquired	-	Radiometric, Magnetic and EM data
Line Spacing	-	50/100/200m
Flying Height	-	40/90m

Gamma Ray Spectrometer

Gamma Spectrometer model	-	Exploranium GR-820/3
Detector Volume	-	42.0 litres (downward and upward looking crystals)
Energy range	-	256 channels, each 12 keV and cosmic window >3.0 MeV.
Lower Energy Threshold	-	100 KeV
Cycle Rate	-	1 Hz (equates to about 50 m traverse distance)

1.1.3 Airborne Survey Data: identification of anomalies

Similar geological and environmental features were observed in the GTK survey data as in the HiRES-1 results. Of particular interest were 4 radiometric anomalies in the East Shirebrook region associated with colliery spoil. Four colliery spoil tips (Shirebrook, Thoresby, Warsop Vale and Welbeck) all stand out clearly as areas of elevated K, U and Th, relative to the surroundings (areas of white in Figure 3, refer to Figure 4). This is most probably a reflection of the strong radiometric contrast between the predominantly shaley colliery spoil and the sandstone or dolomitic limestone on which the colliery spoil tips are sited. To the West of the area depicted in Figure 4, U and Th are more dominant representing the geochemical characteristics of the Zechstein Group (the argillaceous rocks of the Edlington Formation and the Dolomitised limestone and dolomite of the Cadeby Formation; see Section 1.2). The majority of the area depicted in Figure 4 shows a K-dominance, reflecting the geochemical characteristics of the Sherwood Sandstone Group. The radiometric ternary image (Figure 3) also shows forested areas as low activity areas (shown as black), superimposed primarily on the Sherwood Sandstone Formation. These low values appear to reflect a canopy effect on the radar altimeter giving too low an altimeter reading and thus leading to incorrectly height-corrected data (Beamish 2002a).

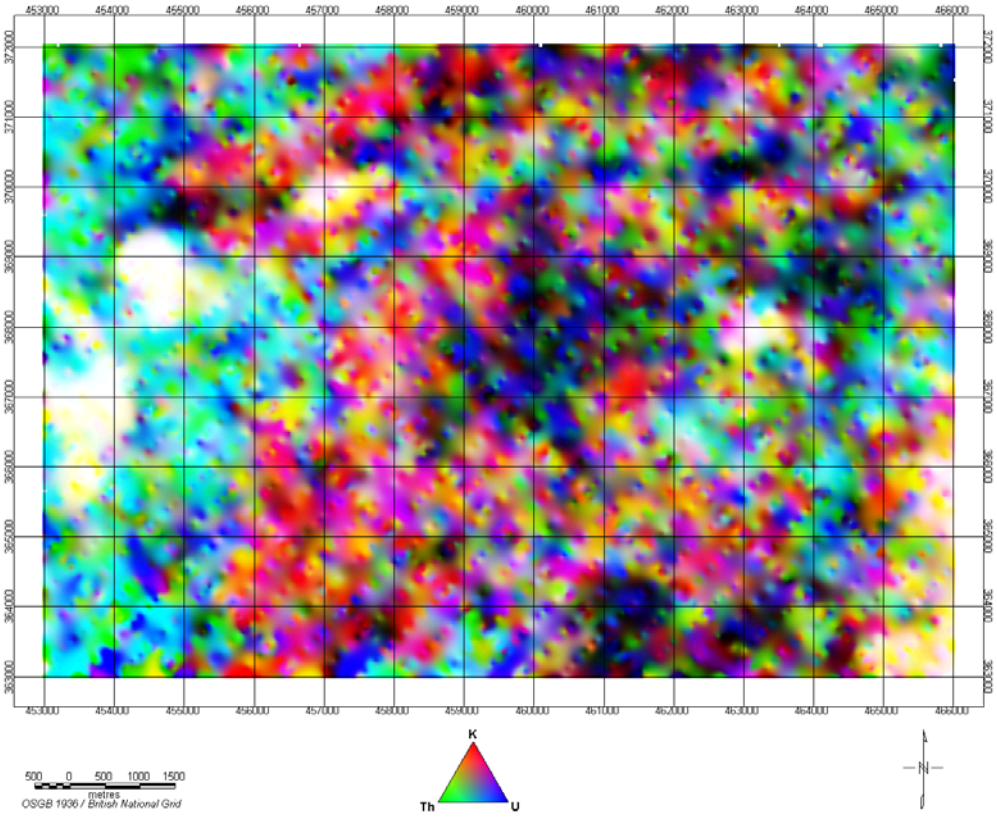


Figure 3 - HiRES-1 ternary image of the Shirebrook area showing relative K, U and Th distribution.

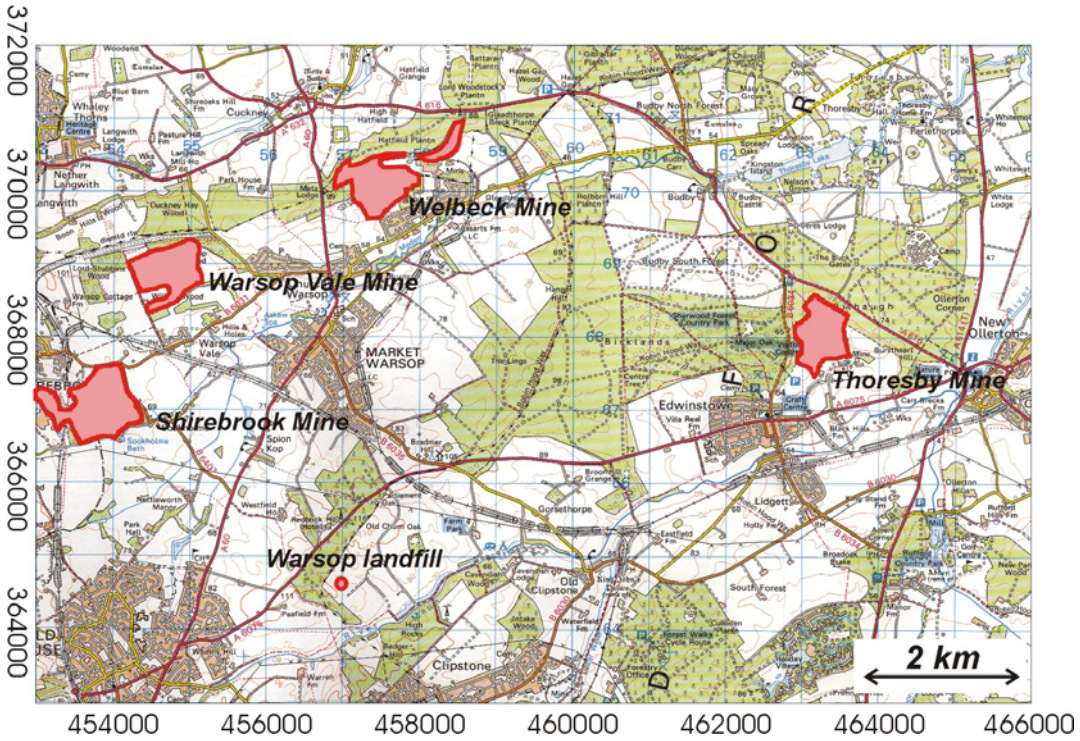


Figure 4- Locations Shirebrook, Warsop Vale, Welbeck and Thoresby colliery spoil sites, the cause of anomalous radiometric features in the Shirebrook area.

The extent of the spoil tips, as indicated by the radiometric data, does not always coincide exactly with the mapped boundaries on Ordnance Survey 1: 50 000 or 1: 25 000 maps (Figure 5). This suggests that, in the absence of up-to-date maps, or where spoil extends beyond the mapped boundary, the radiometric data may be used to accurately define the current extent of the spoil.

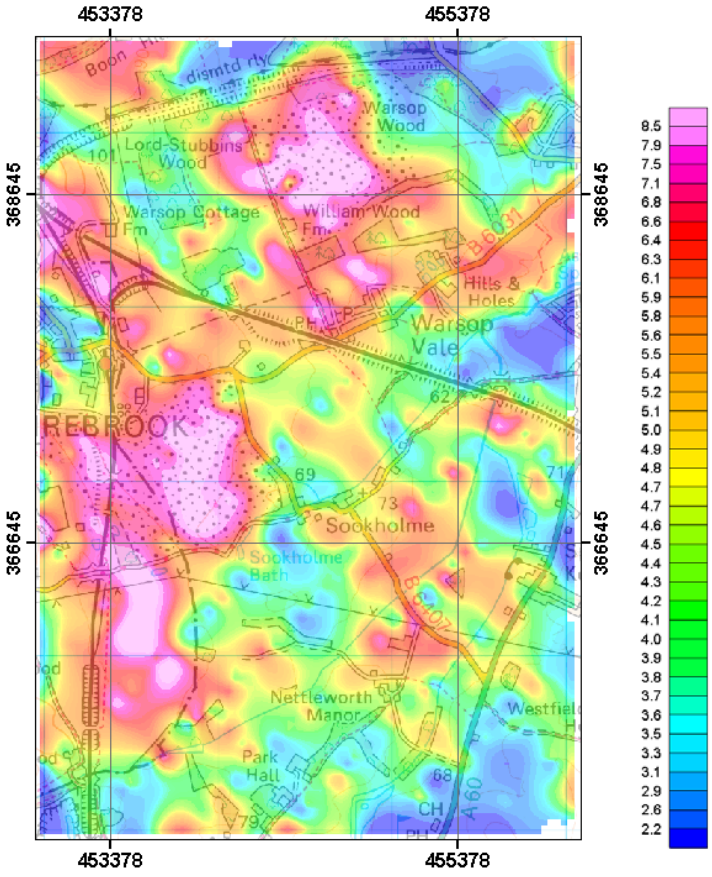
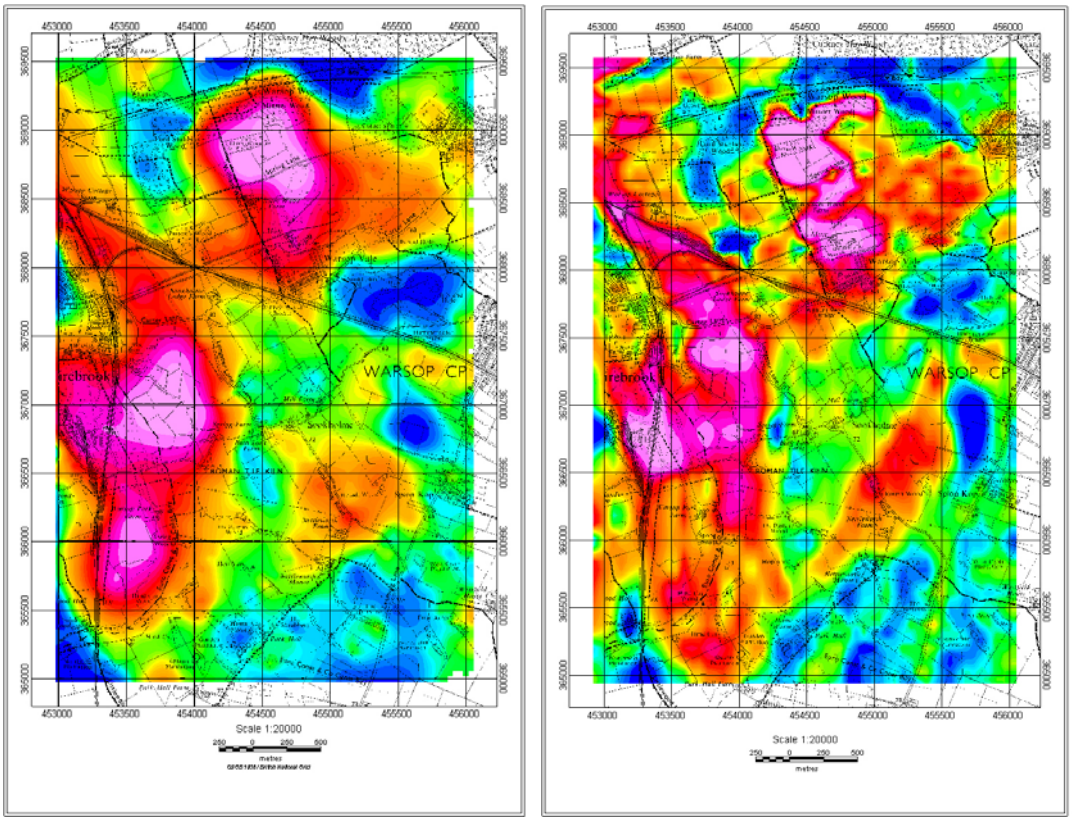


Figure 5- HiRES-1 Thorium data (ppm) at the Shirebrook and Warsop Vale sites. The anomalous area extends beyond the OS mapped boundaries.

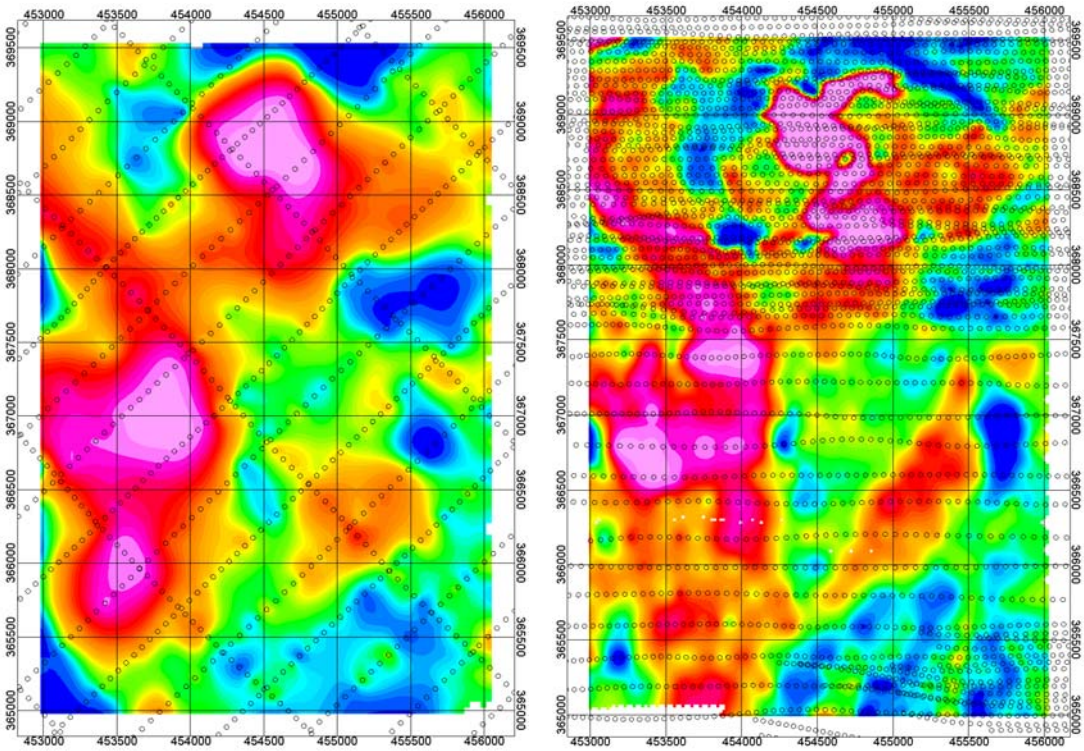
Of the four distinct radiometric anomalies found in the East Shirebrook area, two were working collieries, Thoresby and Welbeck, and two were colliery spoil tips due for reclamation (Warsop Vale and Shirebrook). Further study focussed on the Warsop Vale and Shirebrook sites as access to the working mines was restricted.

Total count data from both the HiRES-1 and the GTK airborne surveys encompassing both colliery spoil areas is displayed in Figures 6 and 7. This demonstrates good agreement between the two surveys at the regional scale with similar patterns of concentrations of radioactivity. They highlight the general features, distinguish well between areas of spoil and adjacent agricultural land and have corresponding areas showing relatively high surface activity. The higher spatial resolution of the GTK survey, evident from the higher density of data points shown in Figure 7, delineates the anomalies with a higher degree of detail. In addition to the higher density of sampling points, the GTK survey flying height was lower than that of the HiRES-1 survey, thus reducing the sampling area, or ‘footprint’ from which the gamma radiation was averaged. This too increased the level of detail visible in the GTK survey results.



a) HiRES-1 **b) GTK**

Figure 6 - Total count data showing radiometric anomalies at both Shirebrook and Warsop Vale spoil tips for a) the HiRES-1 survey flown in 1998 and b) the GTK survey flown in 1999.



a) HiRES-1 **b) GTK**

Figure 7 - Total count data at both Shirebrook and Warsop Vale spoil tips for a) the HiRES-1 survey and b) the GTK survey with locations of data points (o).

1.2 BRIEF GEOLOGICAL AND GEOCHEMICAL DESCRIPTION OF STUDY AREA

1.2.1 Cadeby Formation

This Late Permian member of the Zechstein Group is a grey to buff dolostone (Figure 8). It is commonly oolitic or granular, with subordinate mudstone, dolomitic siltstone and sandstone. It is up to 100 m thick and thickens in the subsurface to the east.

Soil geochemical data over this outcrop gives a background value of around 0.1 % K_2O, <1.9 ppm U and <5.0 ppm Th, which are all low by regional standards (BGS, 2005).

The definition of the lower boundary comes at the base of the dolostone or dolomitic siltstone overlaying organic-rich mudstone (Marl Slate) or where that is absent, Permian basal sands and breccias. The upward boundary is a transition that is usually sharp into the Edlington Formation, or farther east in the subsurface, the Hayton Anhydrite (Smith *et al*., 1986).

1.2.2 Edlington Formation

The Edlington Formation is a Late Permian mudstone, which is red-brown in colour. It contains subordinate siltstone and sandstone with greenish-grey sandstone more common in Nottinghamshire. Dolostone and gypsum/ anhydrite is locally common. Soil geochemical data over the Edlington Formation shows somewhat elevated levels of each element, typically 0.2-0.3% K_2O, 2.5- 3.0 ppm U and 7-10 ppm Th, which are moderately high values regionally, consistent with the lithology (BGS, 2005).

In the Shirebrook area the Edlington Formation passes upwards into the Lenton Sandstone Formation.

1.2.3 Lenton Sandstone Formation

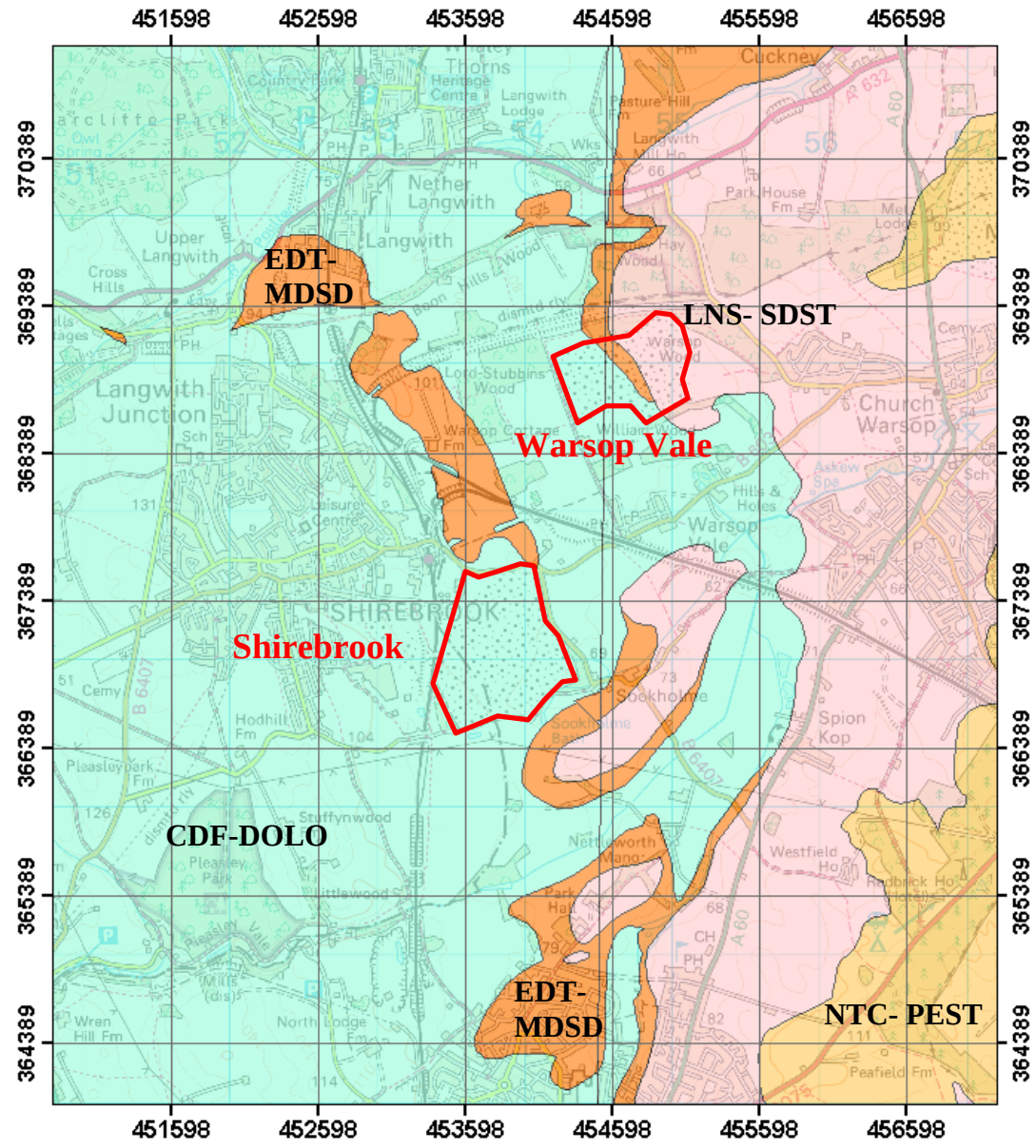
This Late Permian to Scythian sandstone, belonging to the Sherwood Sandstone Group, is very fine to medium-grained, argillaceous and is red-brown with buff mottles. It contains subordinate beds of red-brown mudstone and conglomerate. The lower boundary of the Lenton Sandstone Formation is a gradational upward passage by interdigitation from mudstone of the Edlington Formation.

The definition of the upper boundary is a marked but gradual upward increase in grain size and pebble content into the overlying Nottingham Castle Sandstone Formation (Warrington *et al*., 1980; Charsley *et al*., 1990).

1.2.4 Nottingham Castle Sandstone Formation

Also part of the Sherwood Sandstone Group, this Scythian sandstone is pinkish-red or buff-grey, medium to coarse-grained, pebbly, cross-bedded and friable. Additionally it contains subordinate lenticular beds of reddish-brown mudstone. It ranges in thickness between 70- 170 m and its upper boundary is a sharp non-sequence marked by mudstone at the base of the Mercia Mudstone Group (Warrington *et al*., 1980; Charsley *et al*., 1990; Elliott, 1961).

The Sherwood Sandstone Group lithologies are difficult to distinguish geochemically. Both the Lenton Sandstone Formation and the Nottingham Castle Formation show higher values of K_2O in soils, typically 0.3-0.4 % K_2O, though levels are considerably higher over the younger Mercia Mudstone Group. Uranium values in deeper profile soils over the Sherwood Sandstone Group are variable, but mostly lie in the 1.8-4.1 ppm range. Thorium values are again variable but mostly moderately low on the regional scale, being typically in the 5-9 ppm range (BGS 2005).



CDF-DOLO Cadeby Formation- Dolomitised limestone and dolomite

EDT-MDSD Edlington Formation- Mudstone and sandstone

LNS-SDST Lenton Sandstone Formation- Sandstone

NTC-PEST Nottingham Castle Sandstone Formation – Pebbly sandstone

Figure 8 – 1: 50 000 scale bedrock geology showing location of the Shirebrook and Warsop Vale colliery spoil sites.

2 Ground-Based Investigation

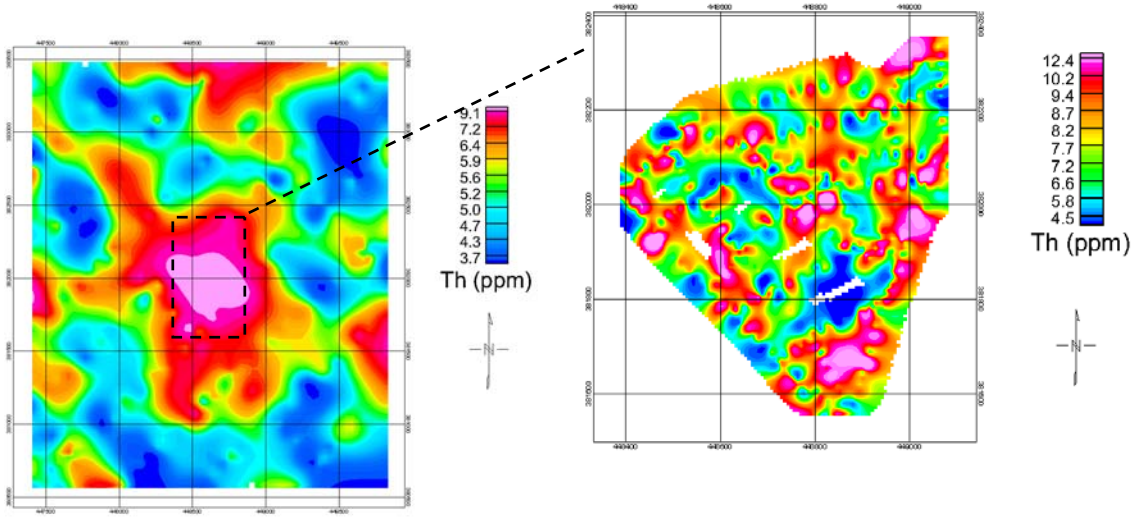
Of the four distinct radiometric anomalies found in the East Shirebrook area, only the disused colliery spoil areas at Shirebrook and Warsop Vale were accessible, as active coal extraction was still going on at Thoresby and Welbeck. A ground survey of these two areas was therefore undertaken to follow up the findings from the airborne survey. This ground survey included:

- Ground-based continuous gamma spectrometry traverses over accessible parts of each site
- Ground-based static gamma spectrometry measurements on selected transects across the sites
- Surface soil sampling at these transect points for laboratory XRF analysis and short core sampling at Warsop Vale for gamma spectrometry analysis on a core logger to investigate changes with depth.

In order to set the results from the ground-based survey in context, previous research on colliery spoil radiometric anomalies is outlined and site history information for both Shirebrook and Warsop Vale is given. The ground-based equipment used and the methods deployed are then detailed.

2.1 PREVIOUS RESEARCH

Kestell (2000), as part of an MSc in association with the BGS, found a good correlation between radiometric anomalies identified by the HiRES-1 survey and colliery spoil tips (Figure 9). Thorium was found to be most elevated above background and the presence of the naturally occurring radionuclides was attributed to the dominance of shale in the spoil. The higher resolution ground survey displays a higher degree of detail, relative to the HiRES-1 survey (Figure 9). Patterns of radionuclide activity displayed in the ground survey were interpreted as weathering of the plateau region resulting in the deposition of radionuclides on the slopes of the spoil heap. Total annual effective whole body equivalent dose rate at the site was calculated as 0.80 ± 0.03 mSv yr^{-1}. Kestell concluded that this dose level could conceivably significantly increase dose to members of the public who were regularly in close contact with the area (Kestell, 2000).



a) **b)**

Figure 9 - Kiverton Colliery Spoil radiometric anomaly showing a) HiRES-1 thorium (ppm) and b) Ground-based continuous gamma spectrometry thorium data (ppm). The hatched line marks the relative positions of both surveys.

2.2 COLLIERY SPOIL SITES IDENTIFIED FOR GROUND INVESTIGATION

2.2.1 Shirebrook

In 1896 the Shirebrook Colliery Company began sinking 2 shafts at Shirebrook, and coal was reached a year later at a depth of about 600 yards (549 m). By 1909 the Company was exporting coal to France, Russia, Italy, Spain, Germany, Norway and Sweden. In 1969 Shirebrook produced its first one million tons of coal but the colliery was closed in April 1993.

Reclamation in the area began in April 1998. Some spoil has been removed and sewage sludge has been applied at a rate of 200-500 tonnes per hectare on the site. This was completed in June 1999, the time of the GTK airborne survey. Planting on the western side of the site was completed by October 1999 and was still being carried out during the current investigations.

The East Midlands Development Agency (EMDA) and Forest Enterprises, owners of the Shirebrook site, have given the land east of the railway line, the main anomalous area, to Nottinghamshire County Council, for public recreational use. The benefits of the ongoing reclamation were obvious to see. Reed-beds for water purification were evident (Figure 10). Lagoons had nesting bird-life including swans, there was a fishing pond, and other grassed areas open to public use. Some areas were still being re-landscaped, or undergoing other treatment such as spraying of agrochemicals. Although reclamation appeared very successful, colliery spoil was still evident in places where a surface cap had been removed, or perhaps where the spoil had never been properly covered (Figure 11).

In addition, the area west of the railway also shows a radiometric anomaly (Figures 6 and 7). This area was being reclaimed when fieldwork was undertaken, and a large section of the spoil was smouldering at a temperature between 30°C and 100°C (Figure 12). Health and safety considerations, due to the close proximity to the smouldering spoil, had to prevail and this area was not surveyed.

Figure 10 – Reed-bed water purification at the Shirebrook reclaimed colliery spoil tip

Figure 11 – Colliery spoil evident at surface, post reclamation at Shirebrook.

Figure 12 – Smouldering spoil heap in area undergoing reclamation at Shirebrook

2.2.2 Warsop Vale

Warsop Main Colliery was the largest of the Staveley Coal & Iron Company collieries. Warsop Main was sunk in 1893 and production started in 1895 in the Top Hard Seam. By 1911, 2700 miners were producing around one million tons of coal a year. Warsop Main closed in 1989 and the headstocks and surface buildings were demolished in 1991.

At the time of the ground survey, the majority of the site had been 'reclaimed' and work was apparently no longer ongoing. The spoil did not appear to have been as well covered as at the Shirebrook site and, despite having had more trees planted (Figure 13), there were still very large areas of exposed spoil (Figure 14), where presumably top soil had disappeared, or was incompletely spread over these areas.

Figure 13 – Young tree plantation at the reclaimed Warsop Vale colliery site. Looking west onto neighbouring agricultural land.

Figure 14 – Exposed spoil at the surface was common at the Warsop Vale reclaimed colliery spoil site.

2.3 METHODS AND EQUIPMENT

2.3.1 Ground-based continuous gamma spectrometry

Continuous ground-based gamma spectrometry was carried out using an Exploranium GR-320 portable gamma spectrometer with a 76 x 76 mm NaI (Tl) detector. This was mounted in a backpack at a height of 1 m while the operator walked slowly over the field survey sites. With the detector at a height of 1 m, it can detect gamma rays from an area approximately within a 10-metre radius to a depth of approximately 30 cm (Atomic Energy Commission, USA, 1972). At both Shirebrook and Warsop Vale, ground-based continuous gamma spectrometry not only covered the areas identified as anomalous by the airborne surveys, but was also extended into surrounding areas to provide background levels. Occasionally access to certain areas was not possible due to ongoing remediation works.

The spectrometer was set to acquire a spectrum every 10 seconds recording counts due to K, U, Th and ^{137}Cs, in pre-set Regions of Interest (ROI's). At the Warsop Vale site, the position of each measurement was recorded using a Magellan ProMark X differential GPS receiver. On return to the office the data streams from the GPS and Spectrometer were merged. At Shirebrook the radiometric data was recorded on a Husky fex21 palm-top computer and merged, in real time, with positional information from the GPS receiver using a modified version of PocketGIS software.

At Warsop Vale 36 line-km of data were acquired and at Shirebrook 27 line-km. As was previously outlined, an equivalent uranium (eU) value is determined from the ^{214}Bi gamma peak and an equivalent thorium (eTh) value is determined from the ^{208}Tl gamma peak. This is because adequate gamma peaks cannot be recorded directly from ^{238}U or ^{232}Th themselves, so daughter products are used to determine an equivalent value and equilibrium with the parent U or Th is assumed.

2.3.2 Ground-based static gamma spectrometry

Ground-based static gamma spectrometry was carried out using the same Exploranium GR-320 and 76 x 76 mm NaI (Tl) detector. Ten minute counts were obtained at a height of 1 m by mounting the detector on a tripod (Figure 15) (Atomic Energy Commission, USA, 1972; Lovborg and Kirkegaard, 1974; Jones *et al*., 1999). Again, with the detector at a height of 1 m it can detect gamma rays from an area approximately within a 10 metre radius to a depth of approximately 30 cm (Atomic Energy Commission, USA, 1972).

The locations of the static gamma spectrometry sites were chosen with reference to the airborne data in order that the measurement sites would transect areas of interest. Measurements were made along transect lines with sampling points approximately 100 m apart. The start and end points of these transect lines were situated in areas that displayed background levels of radioactivity in the airborne data. The position of each measurement point was recorded using the Magellan ProMark X differential GPS receiver.

Approximately 20 static measurements were made at each of the two sites.

Figure 15 – Ground-based static gamma spectrometry

2.3.3 Shallow core and soil sampling

In order to further characterise the radioelement concentrations of the colliery spoil and to compare the gamma spectrometry data to laboratory X-Ray fluorescence measurements (XRF), soil samples were taken at each static measurement site. In addition, in order to partially assess the vertical distribution of radionuclides at Warsop Vale, seven short vertical cores were taken at that site.

Twenty-nine soil samples were taken for XRF analysis (for a wide range of elements) from Warsop Vale and Shirebrook. Soils were collected using a hand-held Dutch auger from the surface horizon (0-15 cm) in order to be most relevant to the radiometric techniques, which can only measure surface concentrations.

Core samples were taken using 6 cm diameter polycarbonate pipe, which was driven into the ground by hand to depths between 21 cm and 45 cm, dependent on ground conditions. On careful removal the cores were sealed, labelled and returned to the laboratory for gamma spectrometry analysis on the BGS Gamma Spectrometer Core Logger. Cores were counted in 1 cm increments by the NaI gamma spectrometer. Full 256 channel spectra were recorded over a specific count time (generally 30 minutes) for each 1 cm step. Regions of Interest (ROIs) were also set up covering parts of the spectrum that allow relative K, U, Th and total count to be evaluated.

In addition to the soils collected during the field survey described in this report, data from previous soil sampling under the BGS Geochemical Baseline Survey of the Environment (G-BASE) project from within a kilometre of the colliery sites provided a further small data set from the area for comparison with the new material. The G-BASE samples were also collected using a hand held Dutch soil auger and were taken from the surface (0 - 15 cm) from a composite of five holes distributed within an area of approximately 20 m x 20 m.

3 Results and discussion

3.1 SHIREBROOK

3.1.1 Airborne and ground-based continuous gamma spectrometry data

HiRES-1 survey data (Figure 16), GTK survey data (Figure 17) and continuous ground-based gamma spectrometry results (Figure 18) are presented as gridded data, displayed as Geosoft Oasis montaj plots. All grids only include data from within the areas shown.

Within the boundaries of the selected areas surveyed by continuous ground-based gamma spectrometry, there are approximately 2700 ground-based gamma spectrometry sample points. This compares with approximately 100 measurement points from the GTK survey and only 15 from the HiRES-1 survey.

Anomalies are, therefore, broader in the HiRES-1 survey data, whilst the GTK survey allows better definition of anomalous areas. For example, the GTK survey defines several areas high in uranium to the north-east of the area and close to the railway to the west of the area (Figure 17), whilst the HiRES-1 survey identifies one larger anomaly between the two areas identified by the GTK survey. Potassium, uranium and thorium values appear slightly higher in the GTK survey than both the HiRES-1 and the ground surveys. This is probably because the GTK survey was calibrated using a range in Finland, unlike the HiRES-1 survey, which was calibrated on a local range in the Vale of Belvoir. The differing ground conditions between Finland and the East Midlands are probably the reason for the discrepancy. This could be checked by examining results for the GTK flights over the Belvoir range. A wider degree of variability, and therefore higher overall values would be expected in the ground survey due to the smaller ‘footprint’, or area over which a measurement averages. The total count values for the two airborne surveys cannot be directly compared as the lower energy thresholds used were different (180 KeV for HiRES and 100 keV for GTK) and the detector volumes differed. Also the HiRES data were provided in counts per second whilst the GTK results were converted to UR (units of radioactivity by weight, where 1 UR is the total count given by 1 ppm of U in equilibrium with its daughters).

The continuous ground data showed a much higher degree of detail, with uranium and thorium showing the highest degree of variability. The two most northerly bands of data were most elevated in all three radioelements (as is highlighted in the total count map, Figure 18). The area to the east, which showed the most elevated potassium levels in the ground-based data, corresponded to an anomaly observed in the GTK data.

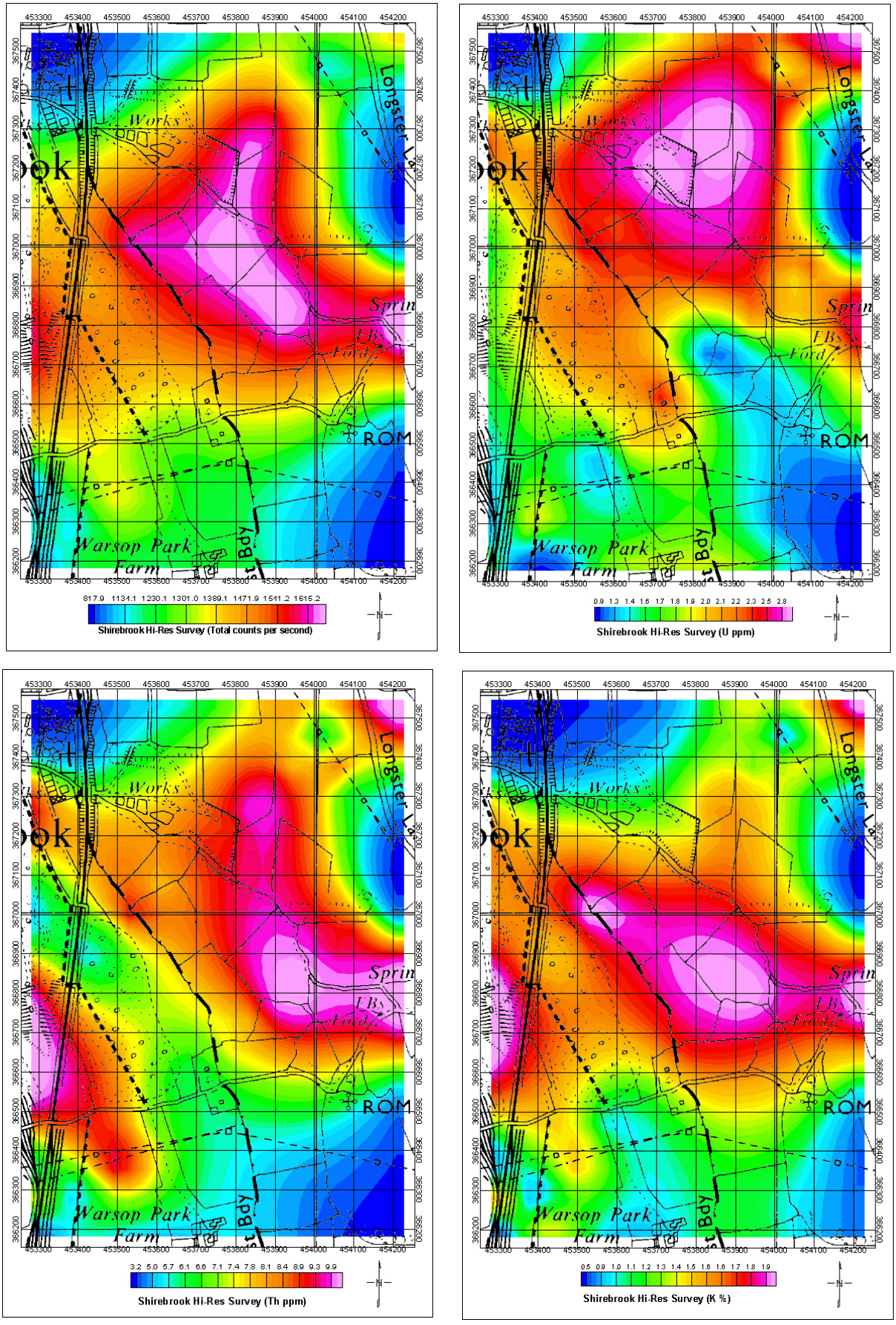


Figure 16 – Total counts per second (cps), uranium (ppm), thorium (ppm) and potassium (%) from the HiRES-1 airborne survey at Shirebrook.

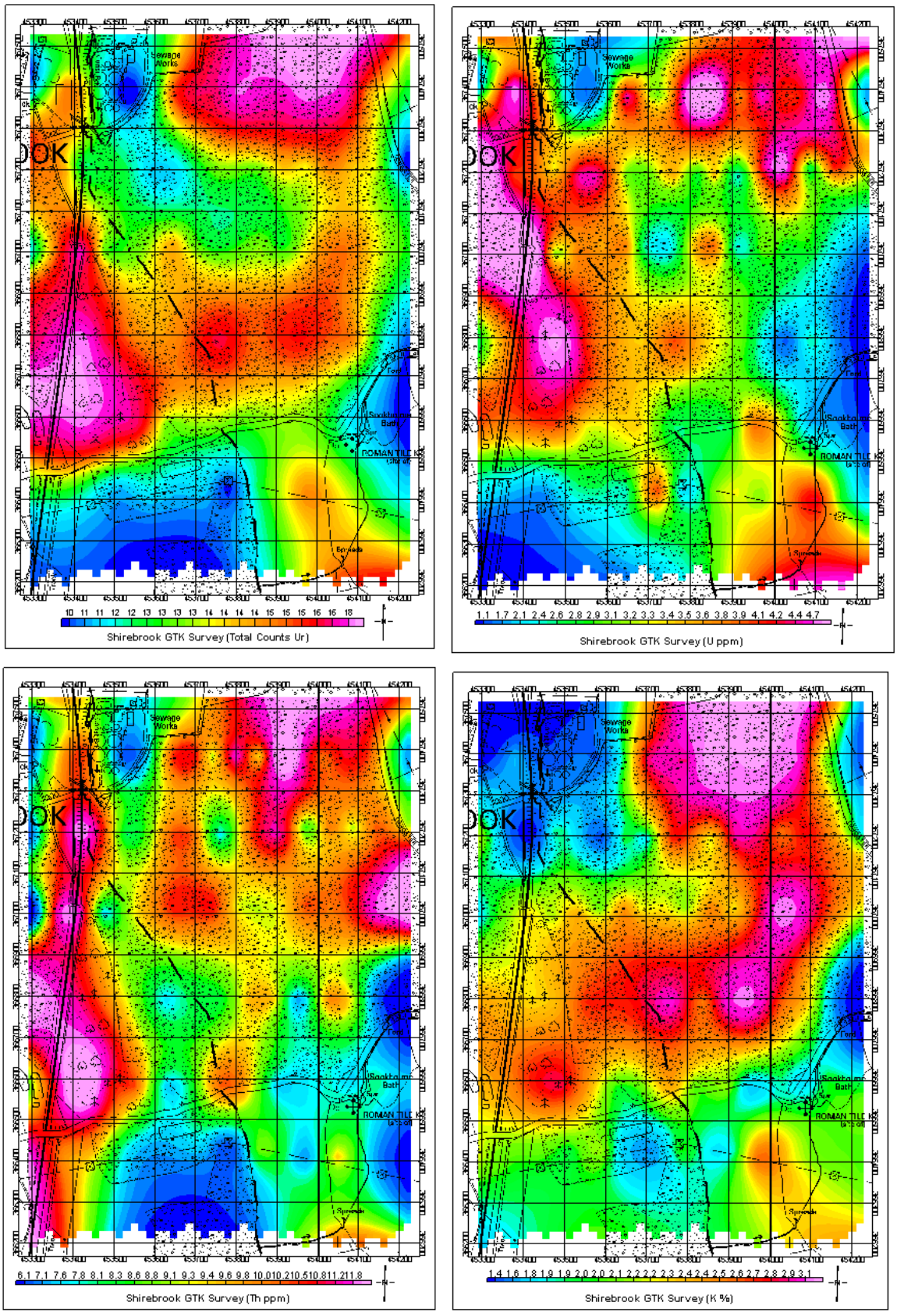


Figure 17 – Total counts (Ur units; 1 Ur is total count from 1 ppm U in equilibrium), uranium (ppm), thorium (ppm) and potassium (%) from the GTK airborne survey at Shirebrook

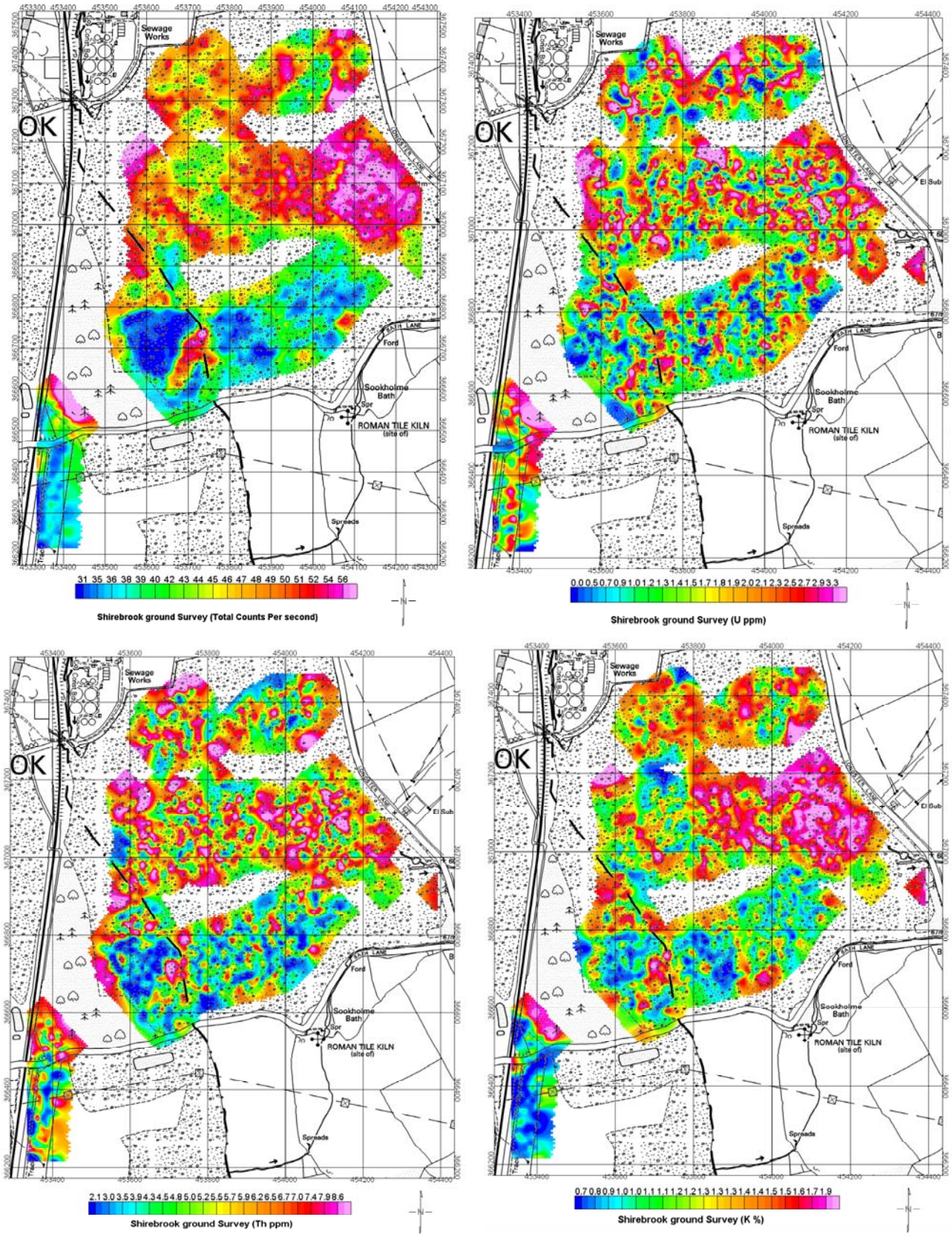


Figure 18 - Total counts per second (cps), uranium (ppm), thorium (ppm) and potassium (%) from the continuous ground-based gamma spectrometry survey at Shirebrook.

3.1.2 Static gamma spectrometry and laboratory XRF measurements

Static gamma spectrometry measurements were made in lines across the site, selected from the airborne data to cover areas of both high and low natural radioactivity. K, U and Th data is displayed in Table 1, with sample locations illustrated in Figure 19.

Soil samples (0-15 cm) were taken at these static measurement points and XRF analysis for potassium, uranium and thorium was carried out (Table 1). Figures 20, 21 and 22 show a comparison of gamma spectrometry derived and XRF derived thorium, uranium and potassium data, respectively.

The results obtained by gamma spectrometry and XRF analysis show a significant correlation, as is discussed in section 3.4.6. The results obtained by gamma spectrometry analysis are generally slightly lower than that of the XRF measurements, as the gamma spectrometry method averages the gamma radiation from an area with an approximate radius of 10 m, where the XRF analysis was made on a split from a much smaller sample volume. In addition, the gamma spectrometry method determines an equivalent uranium (eU) value from the ^{214}Bi gamma peak and an equivalent thorium (eTh) value from the ^{208}Tl gamma peak and equilibrium with the parent nuclide (^{238}U and ^{232}Th, respectively) is assumed. However, disequilibrium in the natural radioactive decay series, particularly for U, is a possible cause for differing results between gamma spectrometry and XRF analysis. The spoil sites have been worked in recent decades potentially altering any states of secular equilibrium. Accelerated radon gas escape, changes to the geochemical environment and hence preferential leaching of members of the decay chain would all contribute to disequilibrium.

Table 1- Potassium, uranium and thorium from static gamma spectrometry measurements and XRF analysis for Shirebrook soil samples (see Figure 19 for site locations). (For average value, <1 values entered as 0.5).

			Static gamma spectrometry			XRF analysis		
Site	**Easting**	**Northing**	**K %**	**eU (ppm)**	**eTh (ppm)**	**K_2O %**	**U ppm**	**Th ppm**
1	454050	366750	1.4	1.5	4.5	2.74	<1	8
2	453950	366750	1.4	1.2	5.4	2.39	<1	7
3	453850	366750	1.1	1.3	5.3	2.23	2	10
4	453750	366750	1.4	2.3	6.8	3.27	3	12
5	453385	366750	2	2.3	9.5	3.02	3	13
6	453396	366850	1.9	2.5	9.7	2.81	2	13
7	453450	366750	1.8	2.4	8.4	3.51	3	13
8	453385	366650	1.8	2.7	9.3	3.02	3	13
9	453382	366555	1.5	2.7	7.8	2.84	3	13
10	453384	366496	1.4	2.9	8.7	3.18	4	16
11	453387	366389	1	1.9	6.8	1.96	2	9
12	453384	366301	1	2	7.2	1.94	2	10
13	453573	367398	1.1	2.2	6.5	2.28	2	11
14	453670	367400	1.9	2.3	7	3.44	1	13
15	453770	367405	1.7	2.1	6.6	3.94	<1	11
16	453870	367400	1.6	2.1	6.4	3.56	<1	12
17	453969	367409	1.5	1.8	7.4	3.09	2	13
18	454071	367417	1.5	2.1	7.3	2.83	2	11
19	453650	366746	1.1	0.8	3.2	1.91	<1	5
Average Value			1.5	2.1	7.0	2.84	1.9	11.2

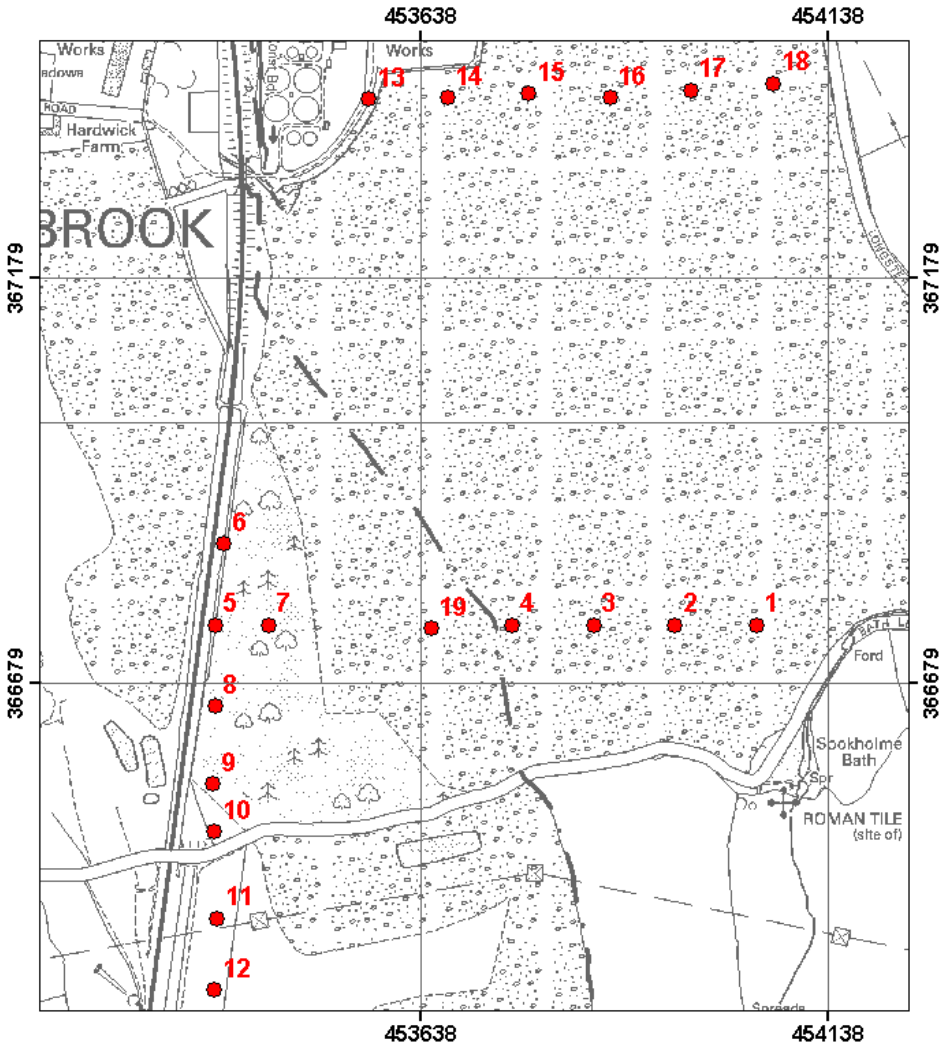


Figure 19 – Static gamma spectrometry and soil sample site locations at Shirebrook

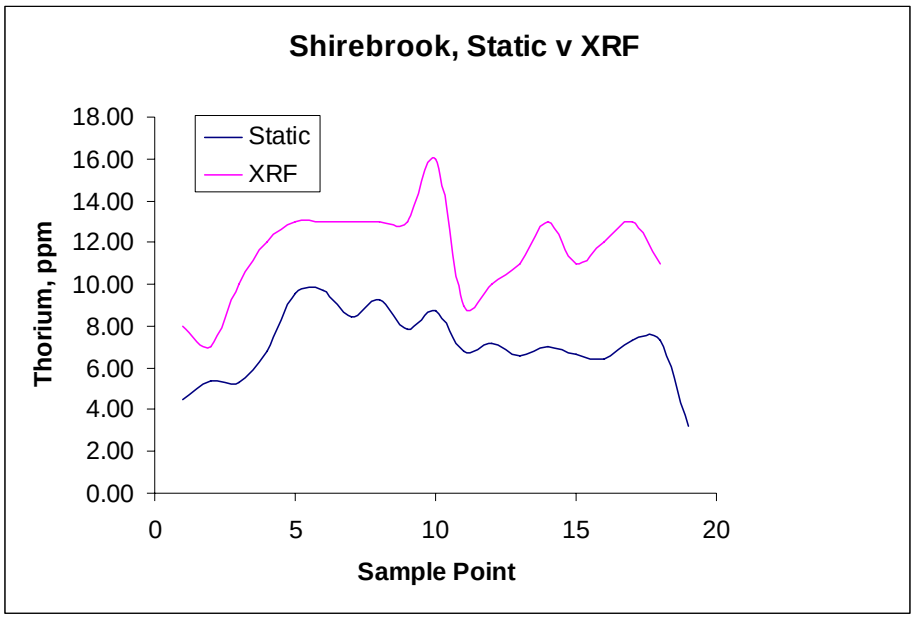


Figure 20- Comparison of thorium measured by static ground-based gamma spectrometry and XRF analysis across the Shirebrook transects (see Figure 19 for sample point locations).

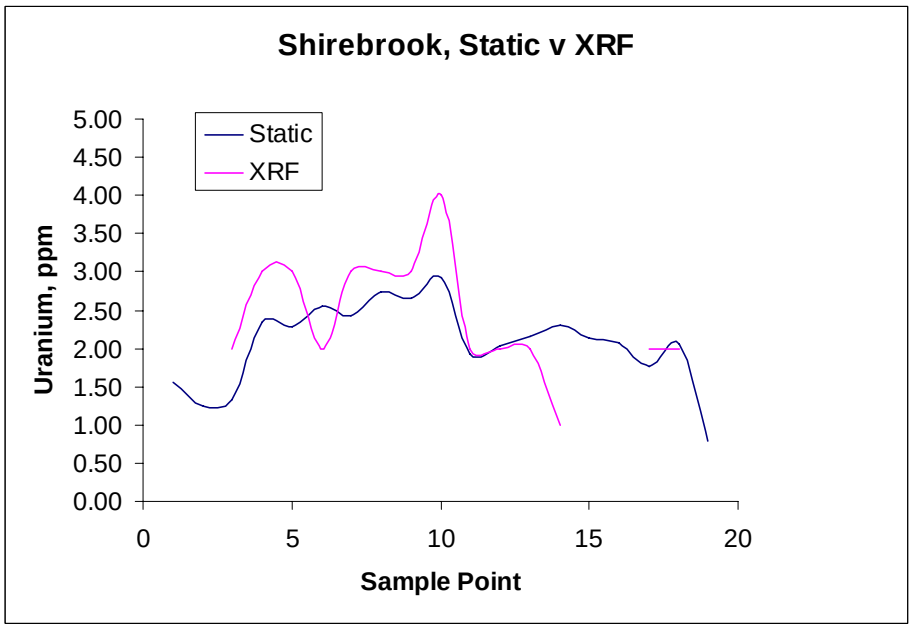


Figure 21 - Comparison of uranium measured by static ground-based gamma spectrometry and XRF analysis across the Shirebrook transects (see Figure 19 for sample point locations).

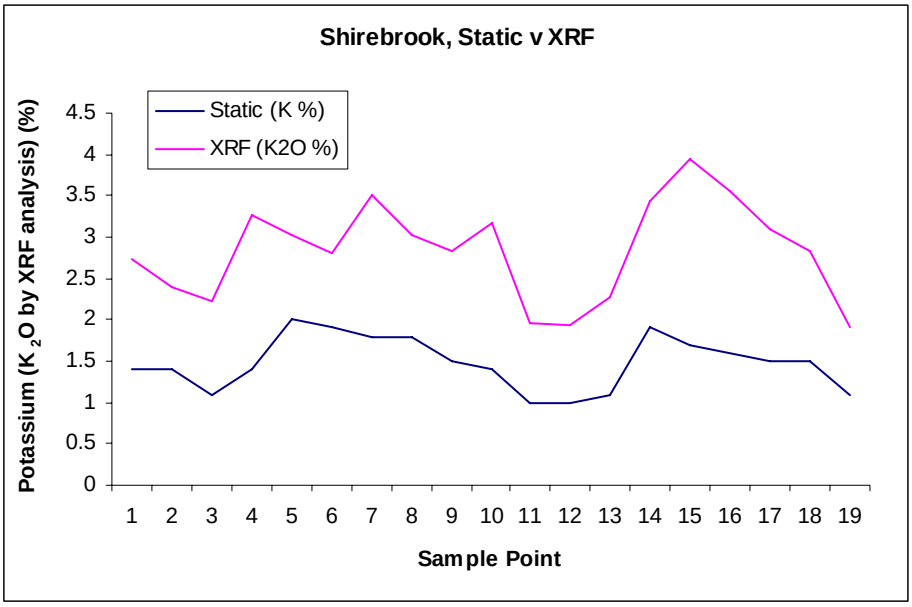


Figure 22 - Comparison of potassium, measured by static ground-based gamma spectrometry and potassium oxide by XRF analysis across the Shirebrook transects (see Figure 19 for sample point locations).

3.2 WARSOP VALE

3.2.1 Airborne and ground-based continuous gamma spectrometry data

HiRES-1 survey data (Figure 23), GTK survey data (Figure 24) and continuous ground-based gamma spectrometry results (Figure 25) are presented as gridded data, displayed as Geosoft Oasis Montaj plots. All grids only include data from within the areas shown.

Within the boundaries of the area surveyed by continuous ground gamma spectrometry, there are approximately 3600 ground-based sample points compared to 600 survey points from the GTK survey and only 50 from the HiRES-1 survey.

All three survey methods agreed well at this site. The HiRES-1 and GTK surveys showed elevated potassium, uranium and thorium over the spoil area itself with the woodland to the north displaying low values in all three elements. The ground-based continuous gamma spectrometry highlighted the marked difference between the radiometric signature of the spoil heap and neighbouring agricultural land to the west (Figure 25). This was particularly obvious in the potassium and total count data sets. This area of lower potassium, thorium, and to a lesser extent, uranium was better defined by the GTK survey than the HiRES-1 survey due to the higher density sampling and lower flying height. A high degree of spatial variation was apparent in the ground-based gamma spectrometry data, but interesting patterns are apparent, and are particularly well displayed in the total count map. High uranium was present to the north and south-east. Thorium mirrored uranium and potassium displayed a ‘banded’ appearance. This data highlighted that the spoil itself is not a uniform mass. The different stages of extraction may have removed several different rock types, which contributed to the spoil.

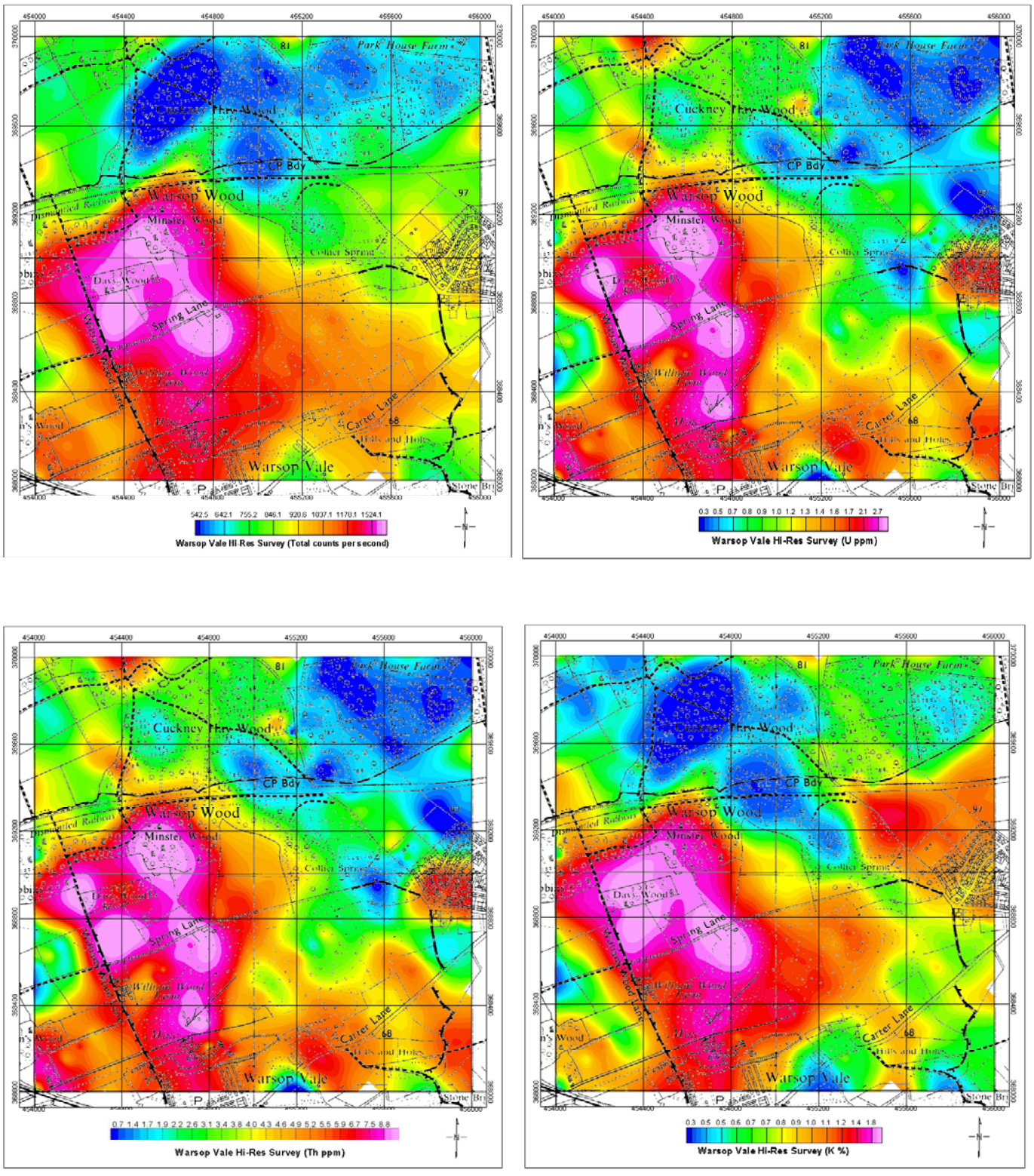


Figure 23 - Total counts per second (cps), uranium (ppm), thorium (ppm) and potassium (%) from the HiRES-1 airborne survey at Warsop Vale.

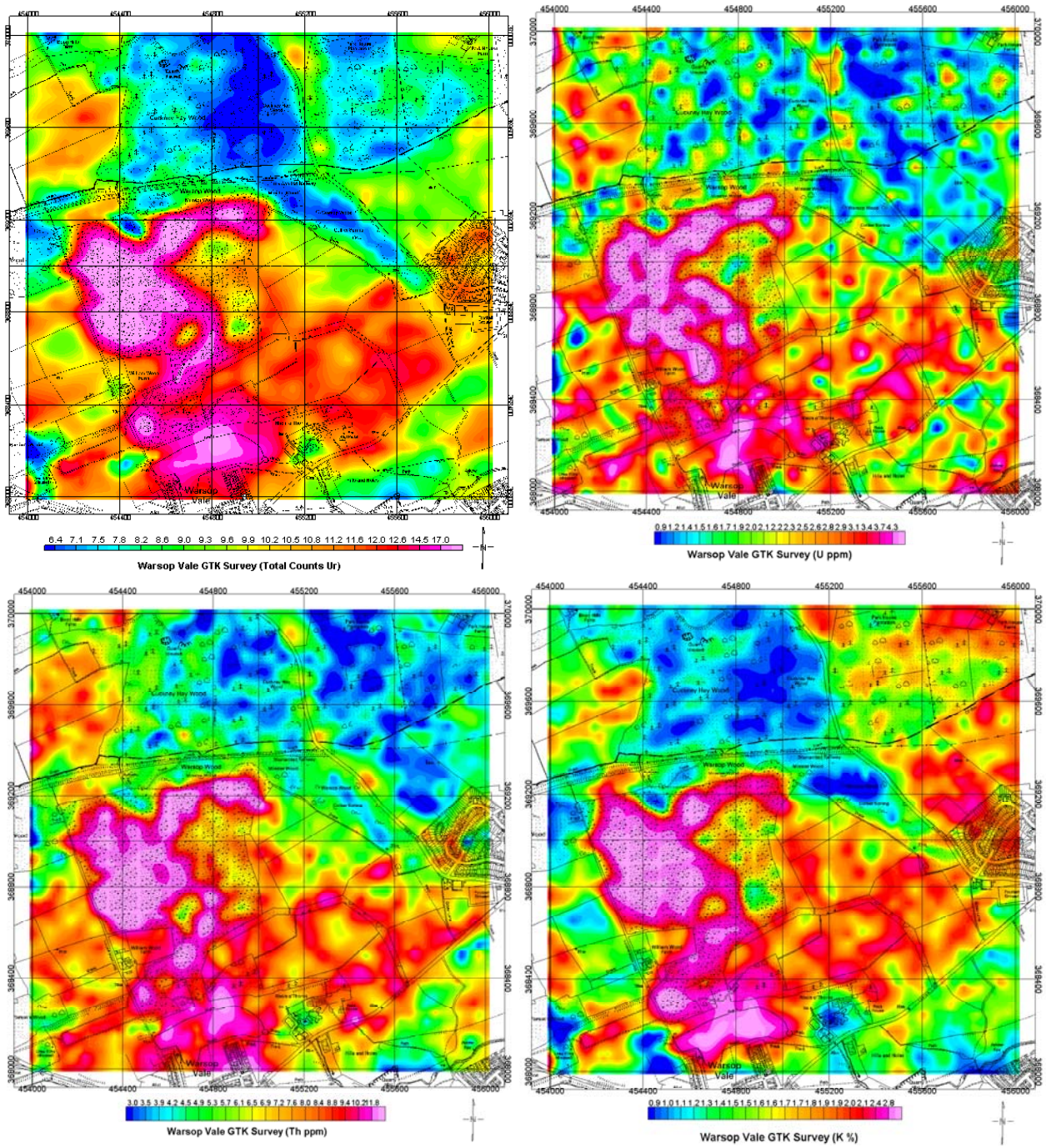


Figure 24- Total counts (Ur units; 1 Ur is total count from 1 ppm U in equilibrium), uranium (ppm), thorium (ppm) and potassium (%) from the GTK airborne survey at Warsop Vale.

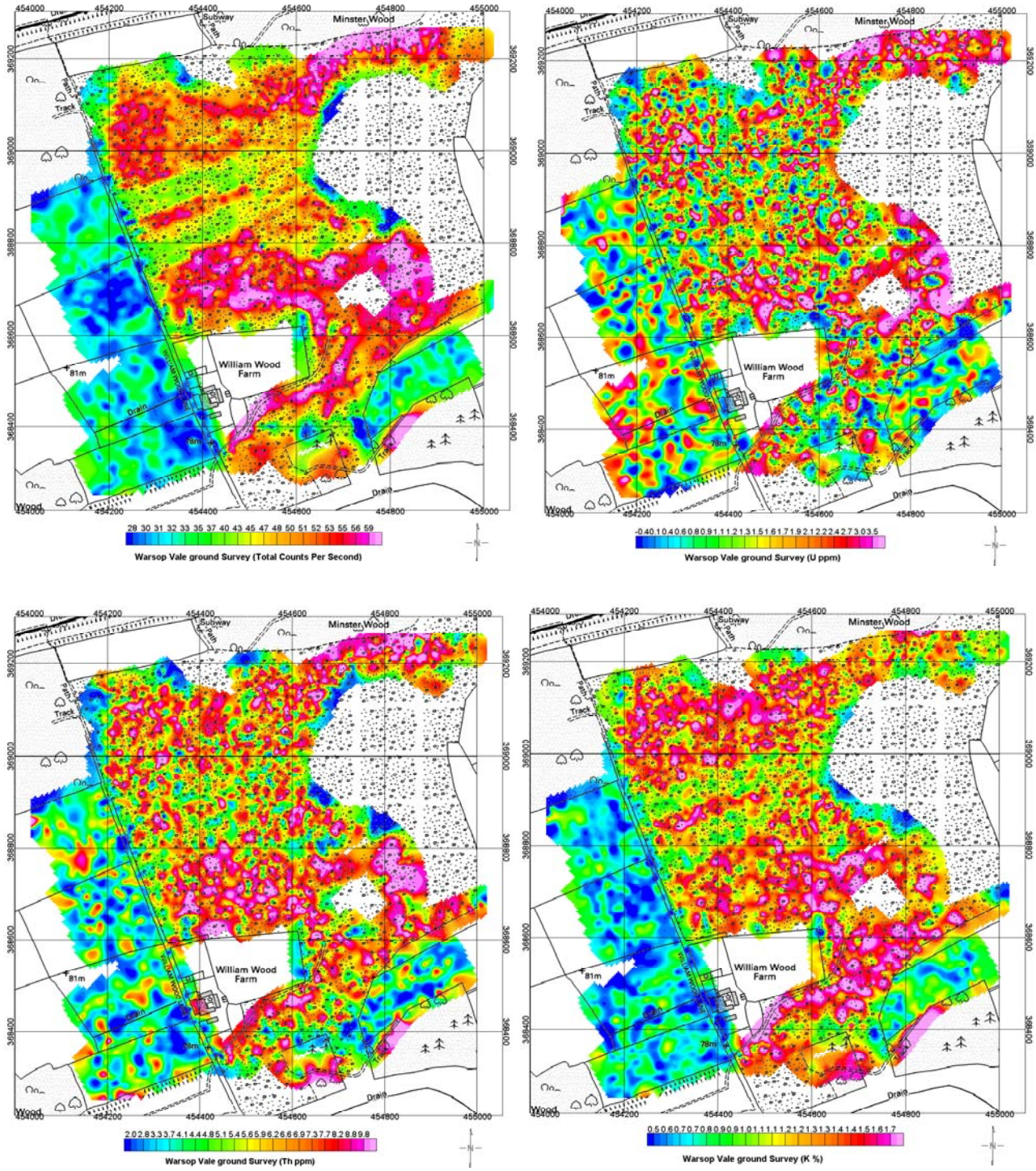


Figure 25- Total counts per second (cps), uranium (ppm), thorium (ppm) and potassium (%) from the continuous ground-based gamma spectrometry survey at Warsop Vale.

3.2.2 Static gamma spectrometry and laboratory XRF measurements

Static gamma spectrometry measurements were made in lines across the site, selected from the airborne data as they represented areas of both high and low natural radioactivity. K, U and Th data is displayed in Table 2, with sample locations illustrated in Figure 19.

Soil samples (0-15 cm) were taken at these static measurement points and XRF analysis for uranium and thorium was carried out (Table 2). Figures 27, 28 and 29 show a comparison of gamma spectrometry derived and XRF derived thorium, uranium and potassium data.

The results obtained by gamma spectrometry and XRF analysis appears to correlate better at Warsop Vale, than at Shirebrook (Figures 20- 22). This is discussed further in Section 3.4.6. The results obtained by gamma spectrometry analysis are again generally slightly lower than those produced by XRF measurement. As was discussed in section 3.1.2, this is due to the smaller volume sampled for the XRF analysis, possible disequilibrium in the natural radioactive decay series and perhaps differences in the methods of calibration between gamma spectrometry and XRFS.

Table 2- Potassium, uranium and thorium from static gamma spectrometry measurements and XRF analysis for Warsop Vale soil samples (see Figure 26 for site locations)

			Static gamma spectrometry			XRF analysis		
Site	**Easting**	**Northing**	**K %**	**U ppm**	**Th ppm**	**K_2O %**	**U ppm**	**Th ppm**
1	454805	369079	0.8	0.9	3.1	NA	NA	NA
2	454676	369064	0.7	1.6	4.6	NA	NA	NA
3	454618	369037	1.2	2.1	6.8	NA	NA	NA
4	454550	369017	1.8	2.2	9.3	NA	NA	NA
5	454476	368991	2	2.3	10	NA	NA	NA
6	454374	368946	1.9	2.2	10.3	NA	NA	NA
7	454297	368919	2	2.6	9.7	NA	NA	NA
8	454233	368893	1.1	1.4	6.7	NA	NA	NA
9	454205	368851	0.9	1.7	6.3	NA	NA	NA
10	454011	368665	0.9	1.2	5.6	2.28	<1	9
11	454096	368692	0.8	1.2	5.5	1.89	<1	8
12	454265	368696	0.8	1.2	5.9	1.99	1	10
13	454366	368699	1.7	3	10.5	3.12	3	14
14	454464	368707	1.8	3	11.7	3.14	4	16
15	454567	368733	1.7	2.5	9.4	3.33	3	14
16	454669	368744	2.1	2.4	10.5	3.40	3	13
17	454786	368756	2	3.1	11.4	3.30	4	14
18	455004	368771	1	1.3	5.5	1.96	1	8
Average Value			1.4	2.0	7.9	2.71	2.2	11.8

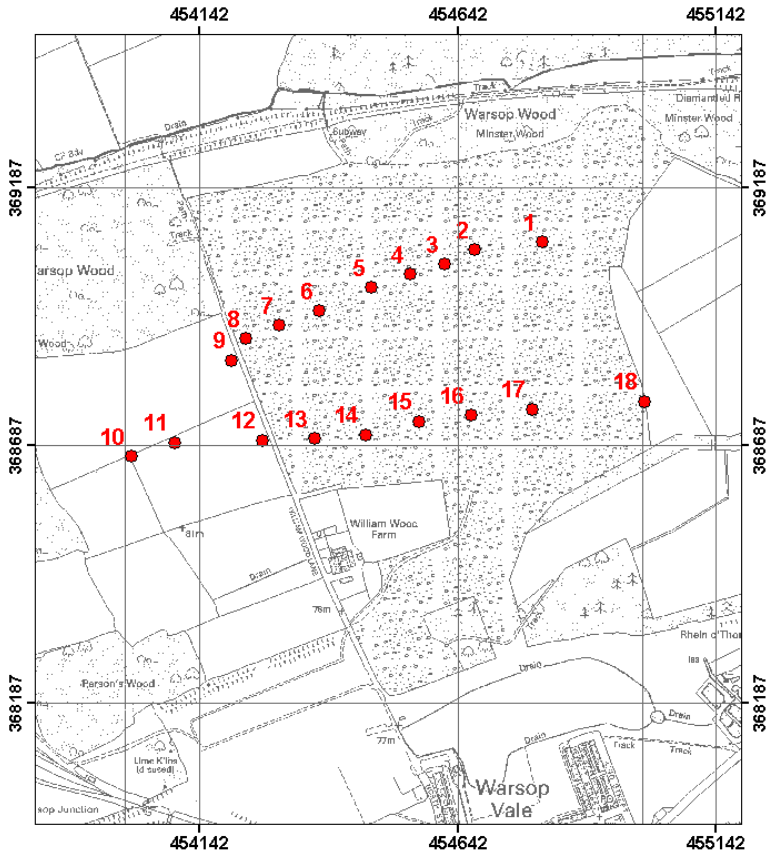


Figure 26- Location of static gamma spectrometry and soil sampling sites at Warsop Vale

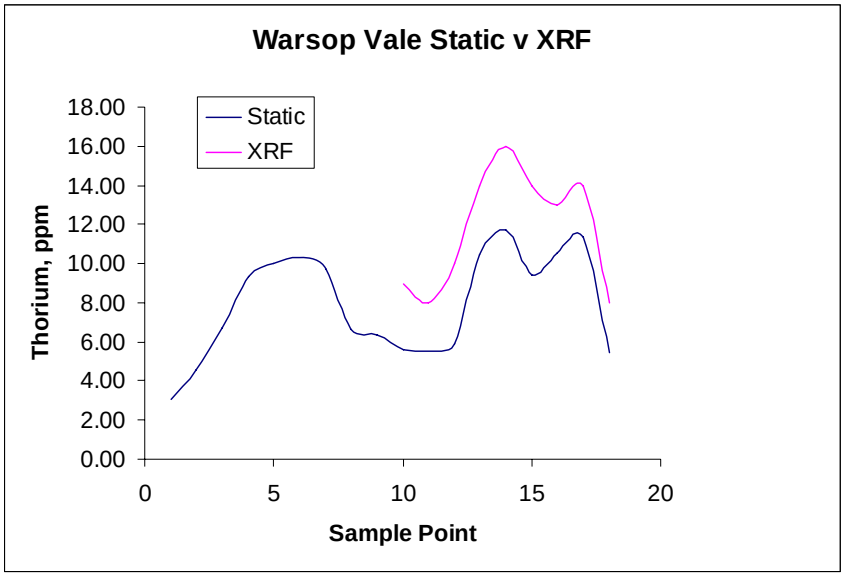


Figure 27- Comparison of thorium measured by static ground-based gamma spectrometry and XRF analysis across the Warsop Vale transects (see Figure 26 for sample point locations).

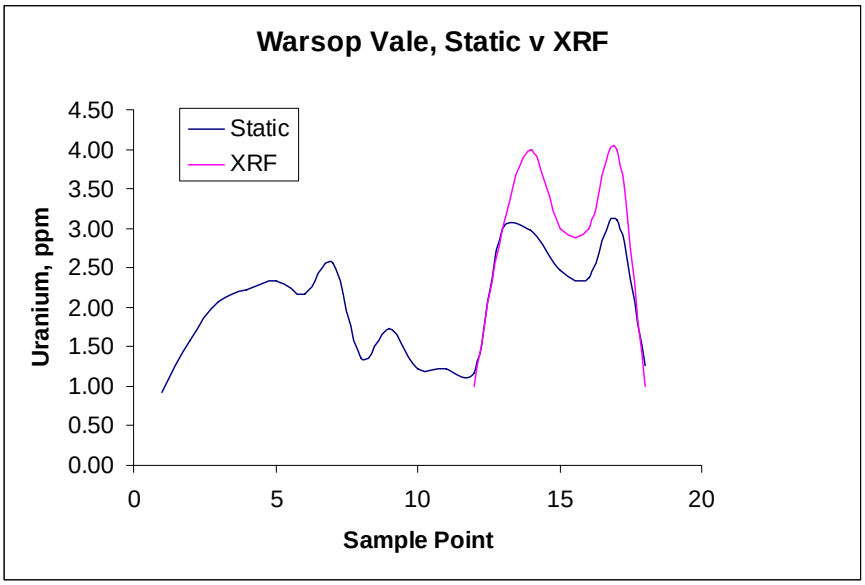


Figure 28- Comparison of uranium measured by static ground-based gamma spectrometry and XRF analysis across the Warsop Vale transects (see Figure 26 for sample point locations).

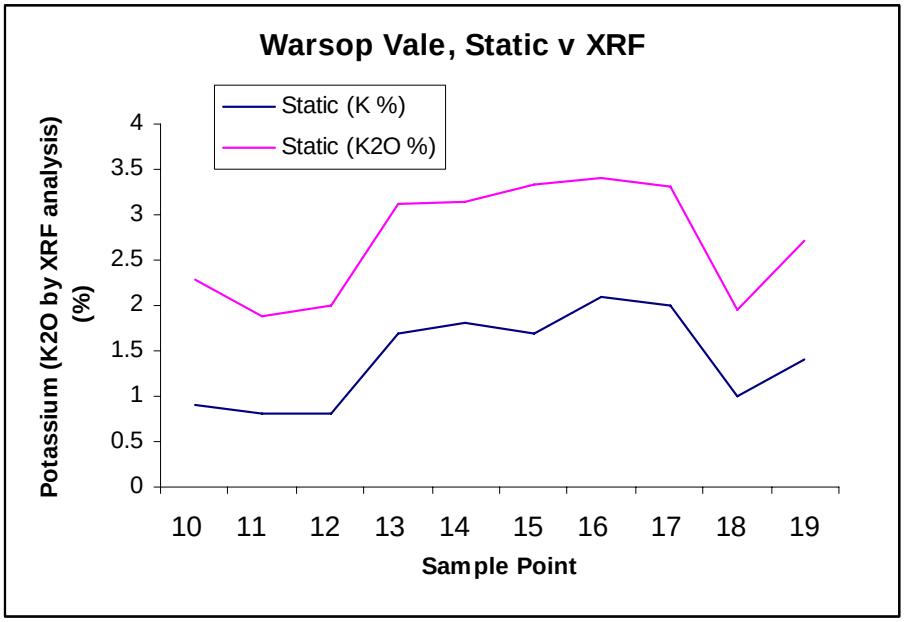


Figure 29 - Comparison of potassium measured by static ground-based gamma spectrometry and potassium oxide by XRF analysis across the Warsop Vale transects (see Figure 26 for sample point locations).

3.3 REFERENCE TO G-BASE ANALYSIS FROM SURROUNDING AREA

Reference was made to soil analysis obtained as part of the Geochemical Baseline Survey of the Environment from areas within 1 km of the spoil tips. Uranium and thorium values from the G-BASE soils (Table 3) were generally lower than those obtained by XRF analysis carried out on soil samples from the spoil tips at Shirebrook and Warsop Vale. Thorium values appeared most elevated, going from an average value of 7.5 ppm in the surrounding area to 11.2 and 11.8 ppm on the spoil tips. This agrees with Kestell's findings (2000), that thorium was the element most elevated above background levels.

Table 3- Small subset of G-Base soil data (0-15 cm), analysed by XRF from areas within 1 km of the spoil tips at Shirebrook and Warsop Vale.

Easting	Northing	U ppm	Th ppm
454280	365910	1.9	7.9
454960	367340	2.0	6.2
453950	366240	1.8	7.5
452360	365530	1.4	5.3
452190	367100	3.1	8.1
453900	368420	1.7	7.5
455120	368140	1.6	8.0
454380	369710	1.7	8.0
453900	368420	2.1	9.0
Average Value		1.9	7.5

3.3.1 Core Samples by gamma logger

A total of seven cores were taken and analysed by gamma spectrometry on the core logger (site numbers 1-6 and 8, see Figure 26 for site locations).

Total counts, relative U, Th and K from each core are displayed as box a whisker plots (Figure 30).

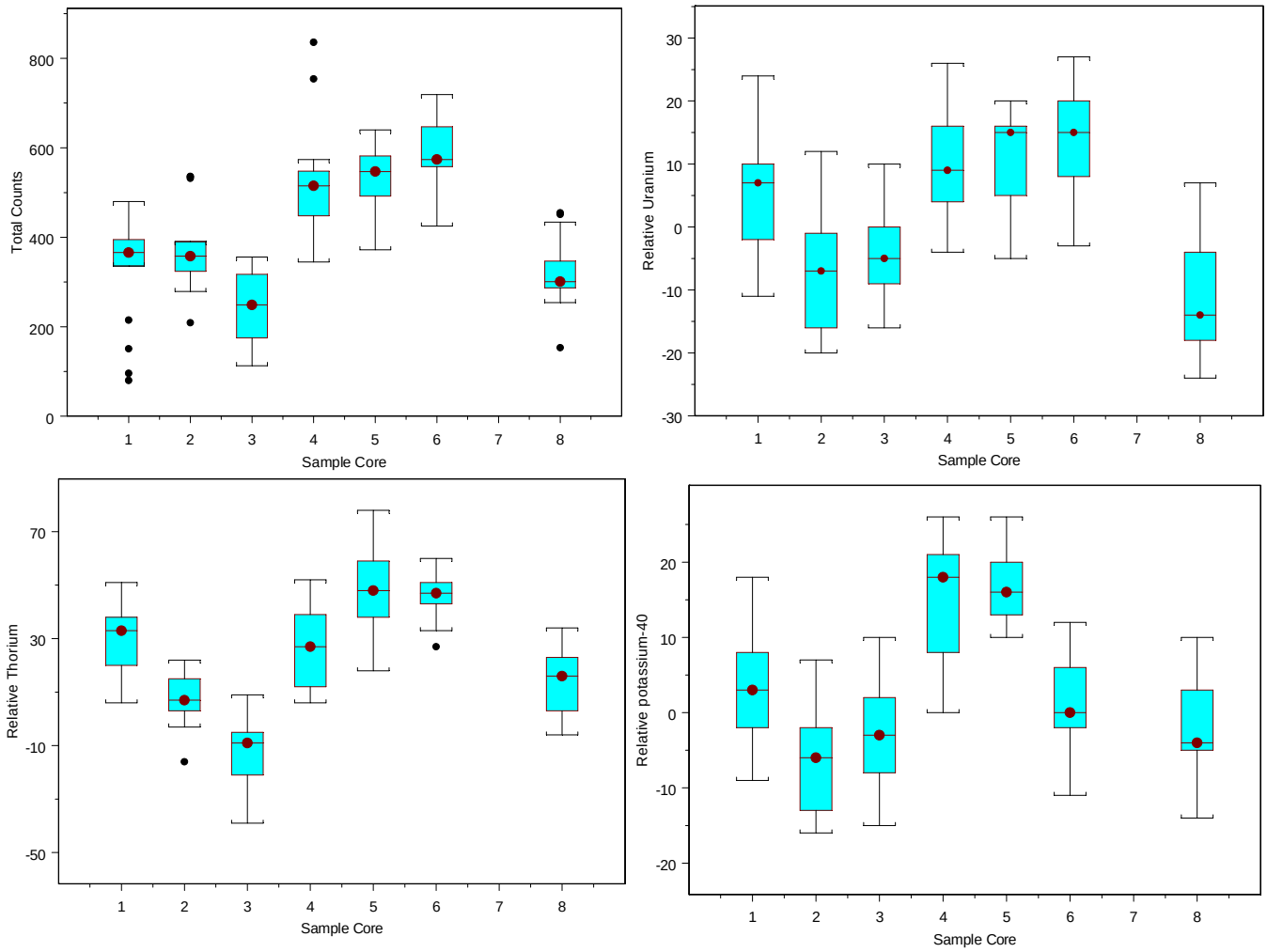


Figure 30- Box and whisker plots displaying total count, relative uranium, relative thorium and relative potassium data from gamma spectrometry analysis of 7 core samples from Warsop Vale (see Figure 26 for site locations).

A similar pattern is observed for the relative concentrations of potassium, uranium and thorium for the cores with cores 2, 3 and 8 showing a generally low radioelement content, core 1 showing moderate radioelement values and cores 4, 5 and 6 a showing higher radioelement content with the exception of K in core 6. When this is related to the ground-based continuous data, exemplified with the potassium data in Figure 31, core 2 would have an approximate concentration of 0.7 % K; core 3, 1.1 % K; core 4 1.8 % K; core 5, 1.0 % K; core 6, 1.2 % K and core 8, 0.8 % K.

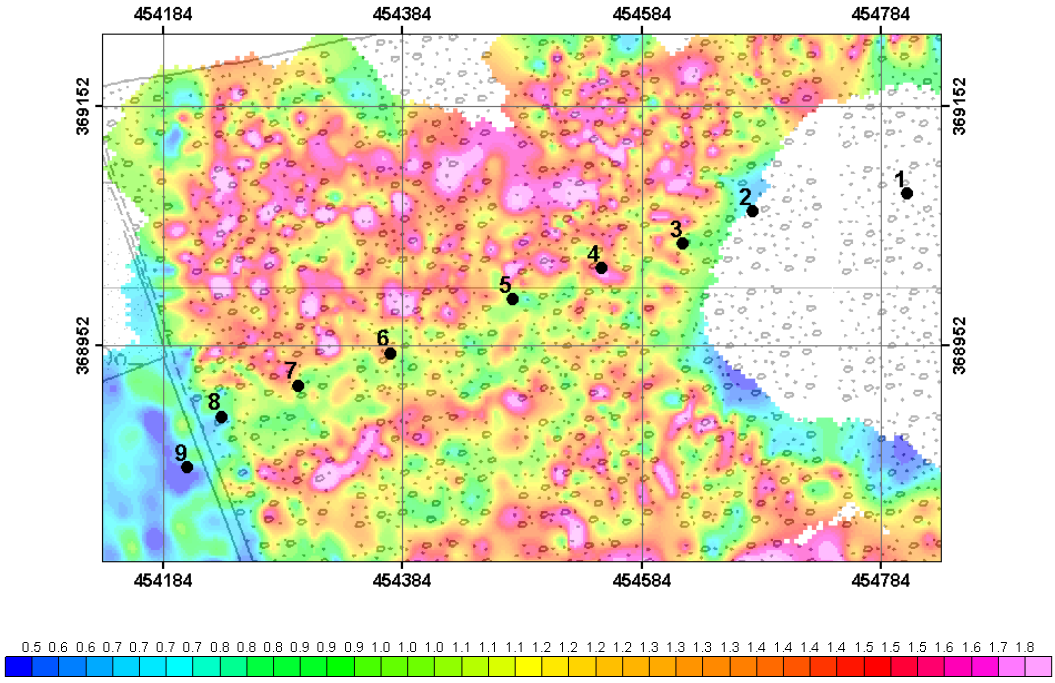


Figure 31- Core site locations (sites 1- 6 and 8) with continuous ground-based gamma spectrometry K (%) data.

As spoil is not formed in situ, it was unlikely to have any well-formed depth profile of radionuclide distribution such as is observed in cores of intertidal sediments or soils formed in situ (Hutchinson and Prandle, 1994; Jones *et al*., 1999; Tyler, 1999; Fox *et al*., 1999; Brown *et al*., 1999). Figure 32 demonstrates that the depth profile of two cores, one chosen from an area of relatively high radionuclide concentration and one from a lower area, is highly variable and shows no discernible pattern in the depth distribution of the radionuclides.

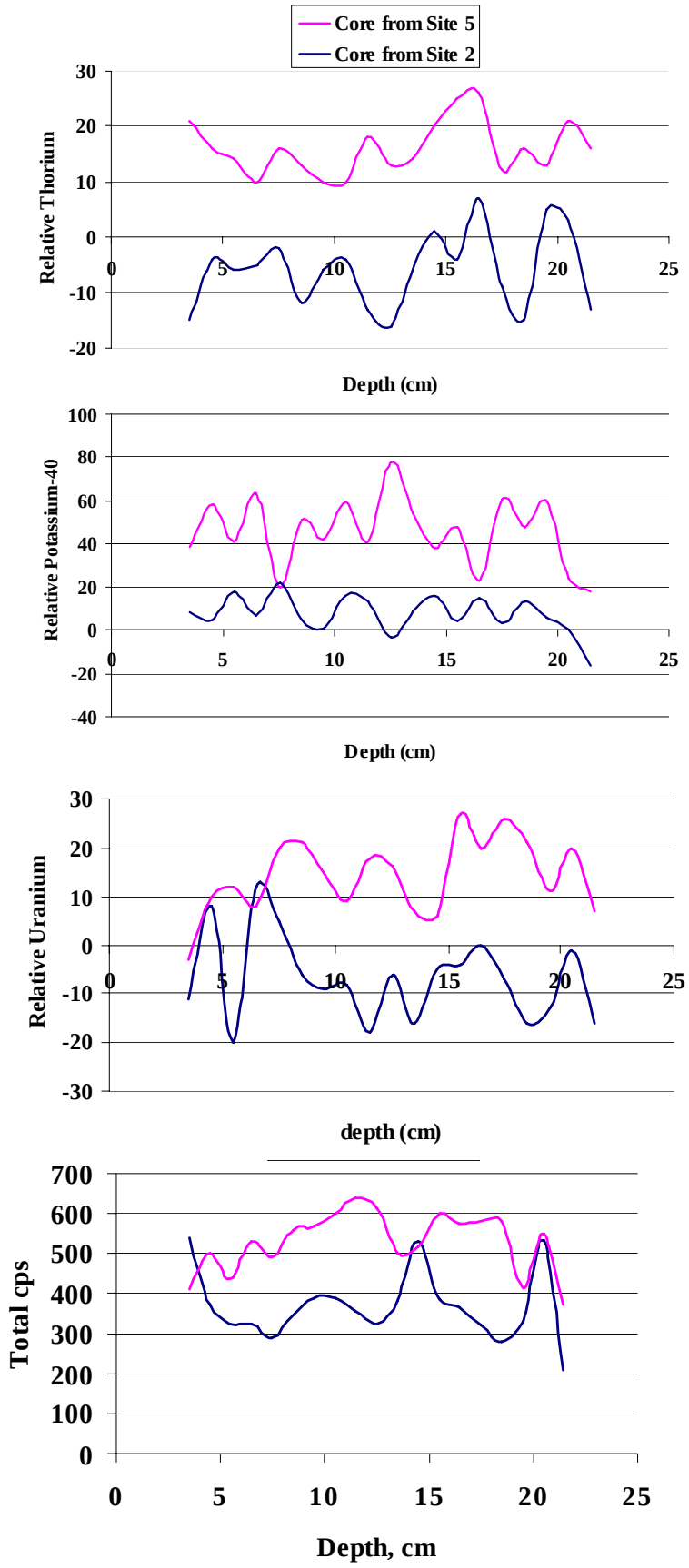


Figure 32- Depth profile of cores 2 and 5 showing relative thorium, relative potassium, relative uranium and total counts.

3.4 RELATIONSHIPS BETWEEN DATA SETS

3.4.1 Statistical overview and comparison between data sets

To give an overview of data ranges, outlying data points and the median for each technique, potassium, uranium and thorium data are presented as box and whisker plots (Figures 33- 38). All data is taken from within the British National Grid boundaries:

Shirebrook: Easting 453200- 455400, Northing 366200- 367600 (Figures 33- 35).

Warsop Vale: Easting 454000 to 455200, Northing 368200 to 369200 (Figures 36- 38).

3.4.2 Shirebrook

The ground-based continuous gamma spectrometry showed the widest variation in potassium, uranium and thorium, as would be expected from the smaller sampling footprint and greater number of observations. The GTK airborne survey results showed a wider range of values than the HiRES-1 data for the same reasons. The GTK survey median values for potassium, uranium and thorium were higher than that of the HiRES-1 and the continuous ground-based survey, which both agreed well. As discussed earlier, this is probably due to the very different geology of the Finnish calibration range. The other sets of data all had inter-related calibrations. The *in situ* ground-based gamma spectrometry results agreed well with the XRF uranium values, but the thorium values determined by XRF were slightly higher. This is likely to result from calibration differences.

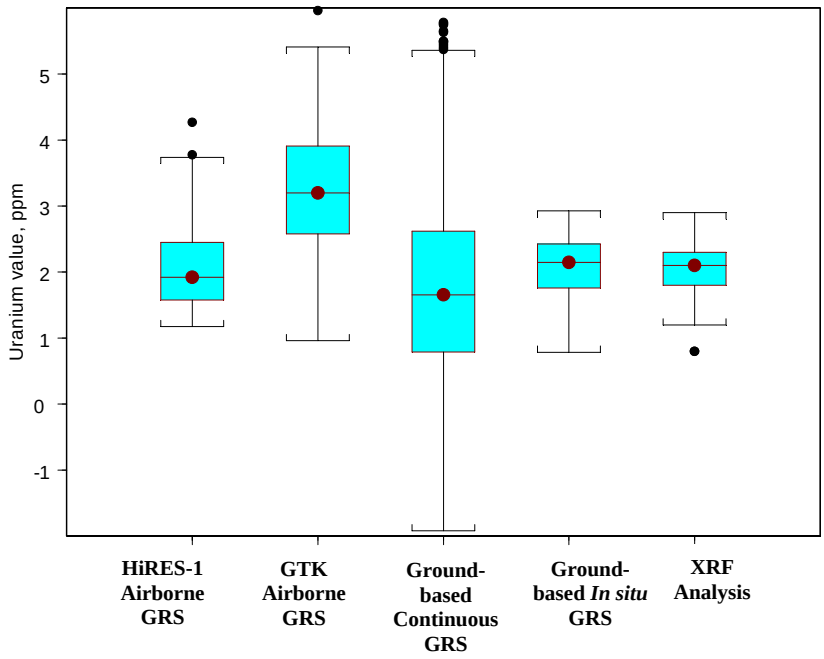


Figure 33- Box and Whisker plot summarising uranium values at Shirebrook by the five different survey methods.

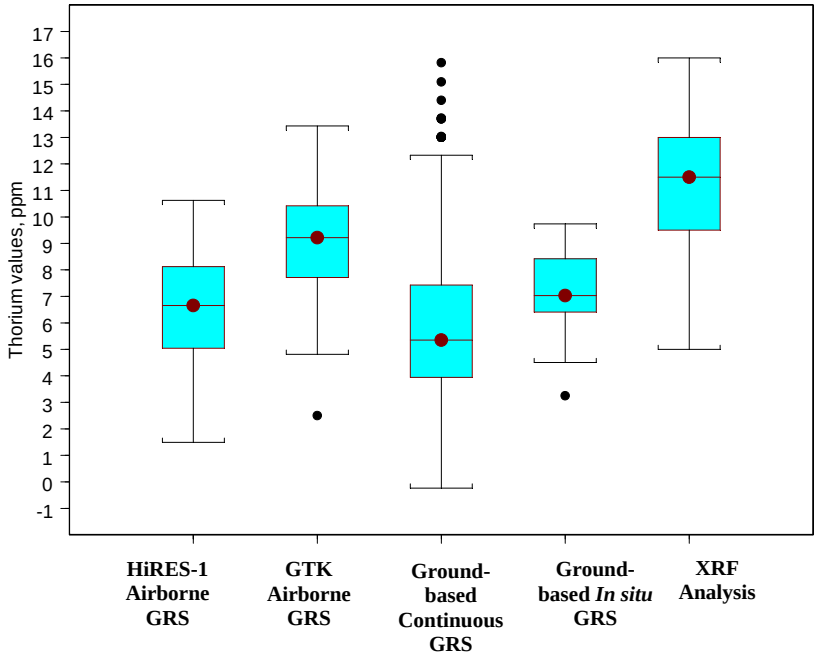


Figure 34- Box and Whisker plot summarising thorium values at Shirebrook by the five different survey methods.

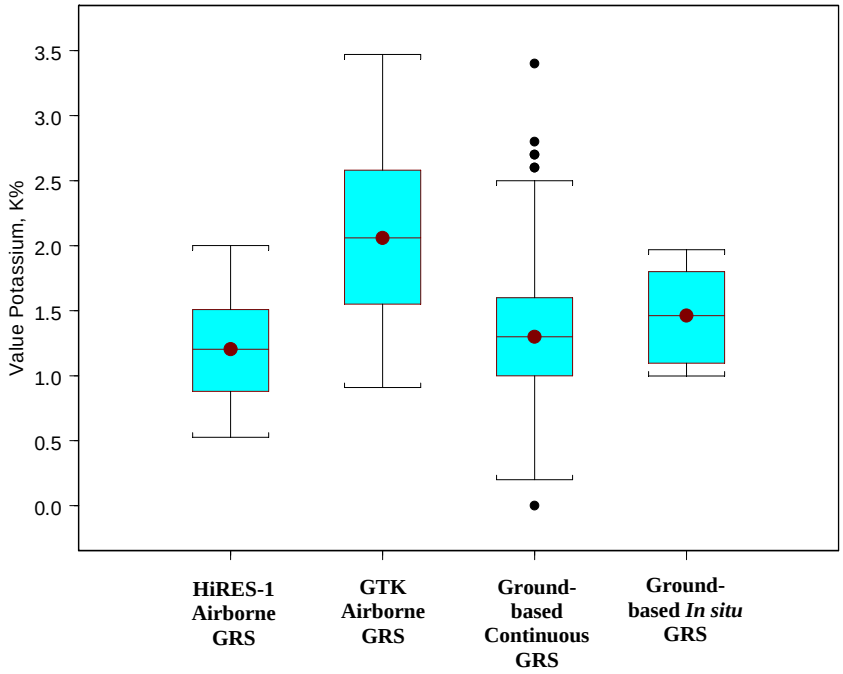


Figure 35- Box and Whisker plot summarising radiometric potassium values at Shirebrook by four different survey methods.

3.4.3 Warsop Vale

Again, as expected, the ground-based gamma spectrometry showed a larger degree of variation than the GTK survey with the smallest range for the HiRES-1 data. The GTK survey median was again slightly higher than the other survey methods except that, once again, the XRF analysis provided higher thorium values. The HiRES-1 survey results agreed well with the ground-based values. *In situ* ground-based gamma spectrometry results showed a slightly higher median for uranium and thorium probably reflecting that the majority of the static sites were chosen on the spoil rather than the surrounding land.

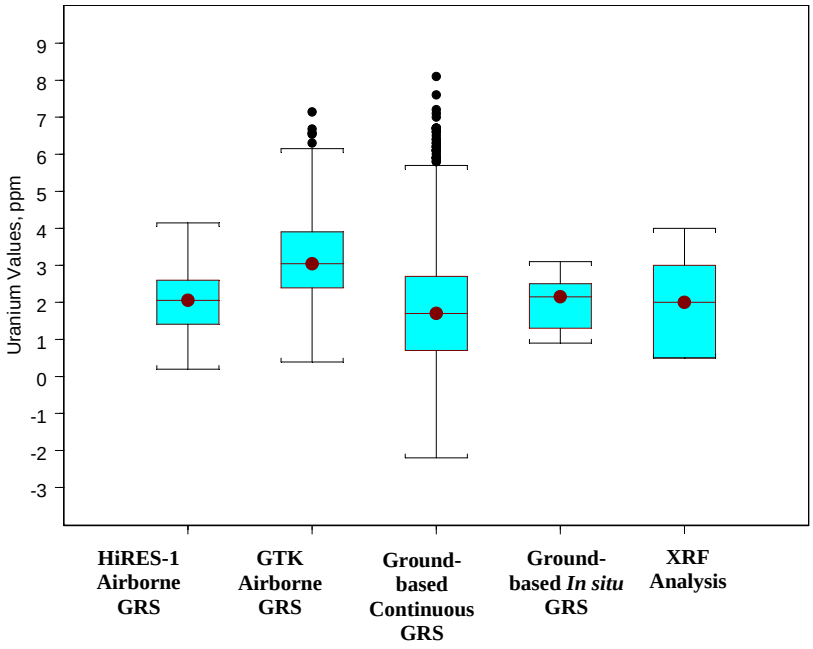


Figure 36- Box and Whisker plot summarising uranium values at Warsop Vale by the five different survey methods.

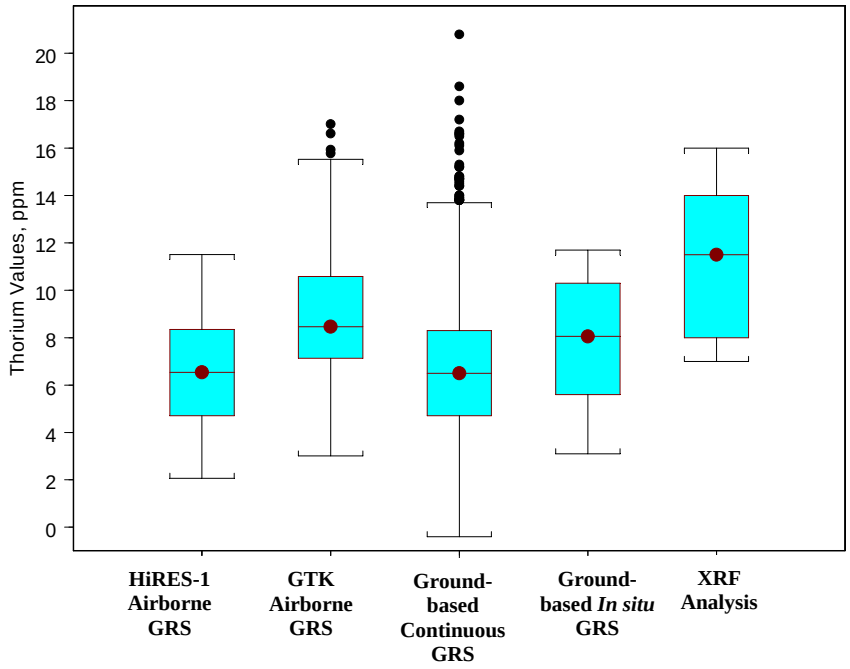


Figure 37- Box and Whisker plot summarising thorium values at Warsop Vale by the five different survey methods.

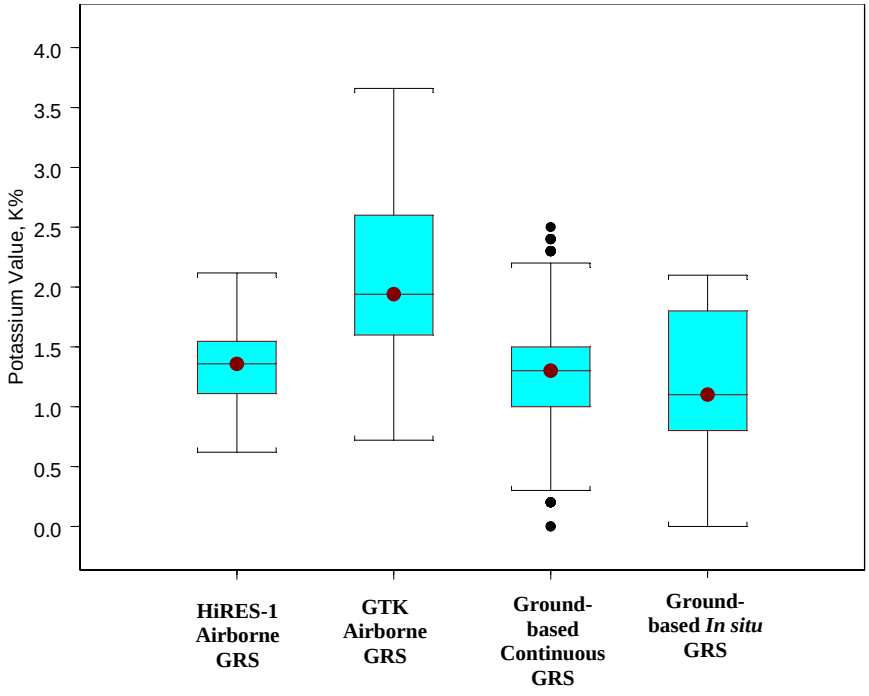


Figure 38- Box and Whisker plot summarising potassium values at Warsop Vale by the four different survey methods

3.4.4 Uranium and thorium ratios across the five survey techniques

To further compare the two colliery spoil sites and the five different survey methods, U/Th ratios were calculated (Table 4). The XRF analysis U/Th ratios were low for each site reflecting the higher measured thorium values. The ground-based continuous and static gamma spectrometry techniques had very similar U/Th ratios at the two sites. This is to be expected as the two methods are measuring the same gamma-ray energy peaks and over the same area. The GTK survey had the highest U/Th ratios reflecting a higher influence of uranium in the measurements.

Typically, an average U/Th ratio in coal is 0.40 (NCRP, 1987). In this study the U/Th ratio ranges from 0.18 to 0.37. Of course, the majority of the colliery spoil is made up of shale and siltstones, with other rock types which occur above, below and within coal seams and which are extracted with the coal during mechanised mining. The dominance of ^{232}Th in the spoil may be a result of radon and ^{238}U daughter nuclide losses from the spoil, and hence disruption to secular equilibrium, due to enhanced mobility in this unconsolidated material.

Table 4- Uranium and Thorium ratios over 5 survey methods

Uranium / Thorium Ratios	Shirebrook	Warsop Vale
HiRES-1 Airborne Survey	0.18	0.30
GTK Airborne Survey	0.37	0.36
Continuous Ground Gamma Spectrometry	0.30	0.27
Static Ground Gamma Spectrometry	0.30	0.25
XRF	0.18	0.18

3.4.5 Comparison of the two airborne surveys: GTK and HiRES-1

HiRES-1 was flown between May and September 1998 and the GTK survey one year later in June 1999. Data from the two surveys were compared, in the study area, by linking the nearest point in the GTK survey to each HiRES data point and then using only those points where the distance was less than 50 m. This should mean that the points compared should have significant overlap in their ground footprint.

The radiometric data from the two surveys correlate at the 99.95 % level for uranium (Figure 39, $r = 0.4$, $p = 0.0005$), thorium (Figure 40, $r = 0.60$, $p = 0.0005$) and potassium (Figure 41, $r = 0.68$, $p = 0.0005$). The GTK values are slightly higher however, and this is probably due to the different calibration range used, as discussed earlier. Reclamation may have occurred at the two sites in-between the surveys, which does provide an added complication. Even post-reclamation spoil was not completely covered by topsoil, particularly at Warsop Vale, and values would be expected to be lower post-reclamation, not higher as seen in the GTK survey data.

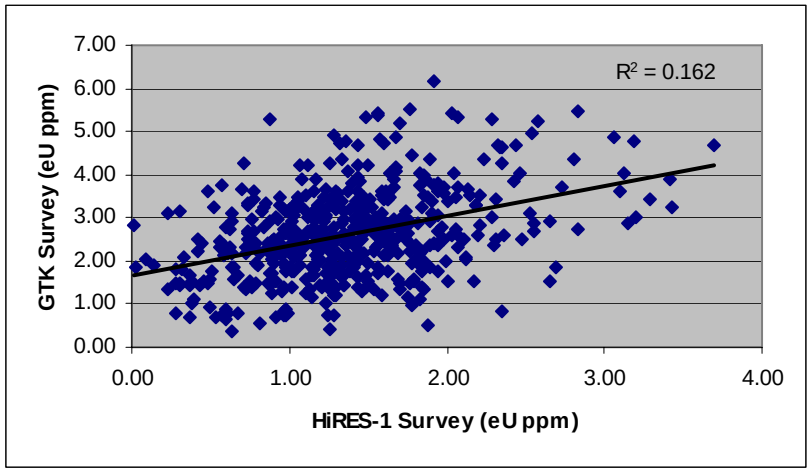


Figure 39- Correlation of HiRES-1 and GTK airborne survey uranium data for the area surrounding Shirebrook and Warsop Vale former collieries.

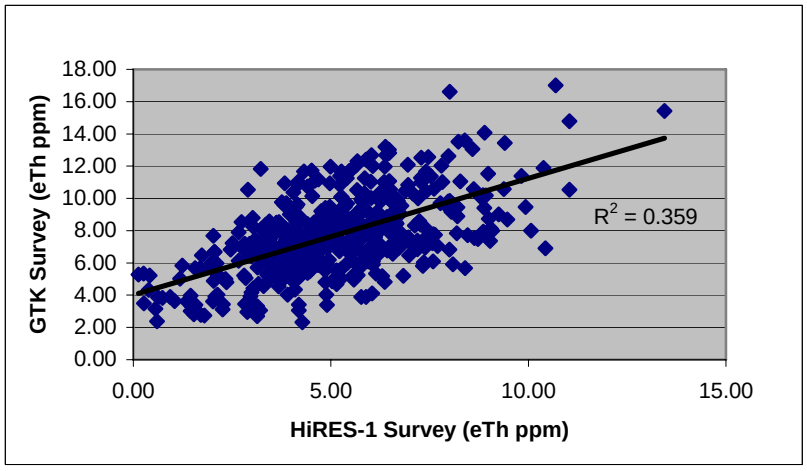


Figure 40- Correlation of HiRES-1 and GTK airborne survey thorium data for the area surrounding Shirebrook and Warsop Vale former collieries.

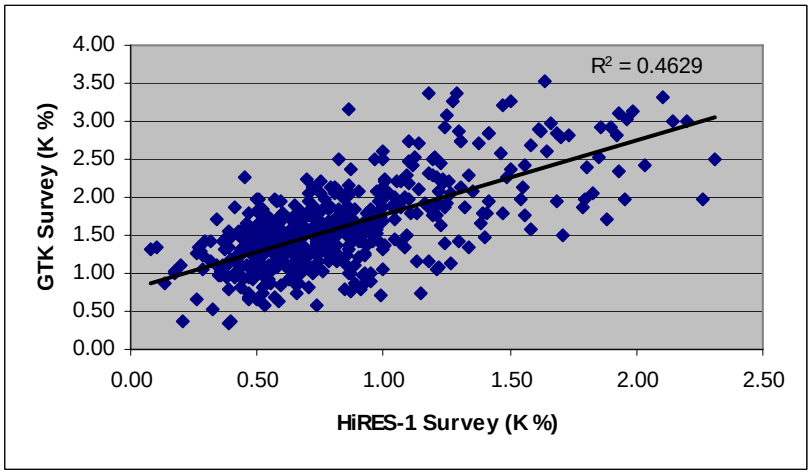


Figure 41- Correlation of HiRES-1 and GTK airborne survey potassium data for the area surrounding Shirebrook and Warsop Vale former collieries.

3.4.6 Comparison of XRF and ground-based gamma spectrometry analysis for both colliery spoil sites

A correlation was observed between the XRF and static ground-based gamma spectrometry analysis which was significant at the 99.95 % level for uranium (Figure 42, r =0.82, p =0.0005, n =18), thorium (Figure 43, r =0.87, p =0.0005, n =27) and potassium (Figure 44, r =0.84, p =0.0005, n =27.

The results obtained by gamma spectrometry analysis are generally slightly lower than that of the XRF measurements, as the gamma spectrometry method averages the gamma radiation from an area with an approximate radius of 10 m, where the XRF analysis is direct on a smaller sample volume. In addition, the gamma spectrometry method determines an equivalent uranium (eU) value from the ^{214}Bi gamma peak and an equivalent thorium (eTh) value from the ^{208}Tl gamma peak and equilibrium with the parent nuclide (^{238}U and ^{232}Th, respectively) is assumed. However, disequilibrium in the natural radioactive decay series is a likely cause for differing results between gamma spectrometry and XRF analysis. The spoils sites have been worked in recent decades potentially altering any states of secular equilibrium. Accelerated radon gas escape, changes to the geochemical environment and hence leaching of members of the decay chain would all contribute to disequilibrium.

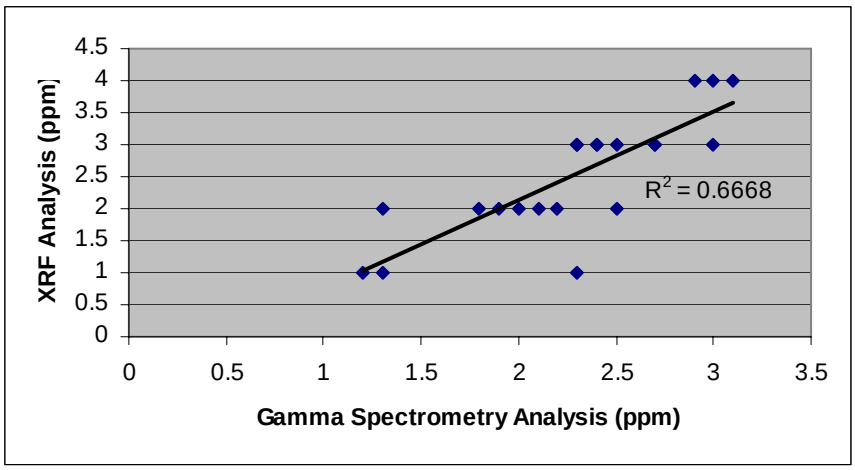


Figure 42- Comparison of uranium data by XRF and static ground gamma spectrometry analysis from Shirebrook and Warsop Vale

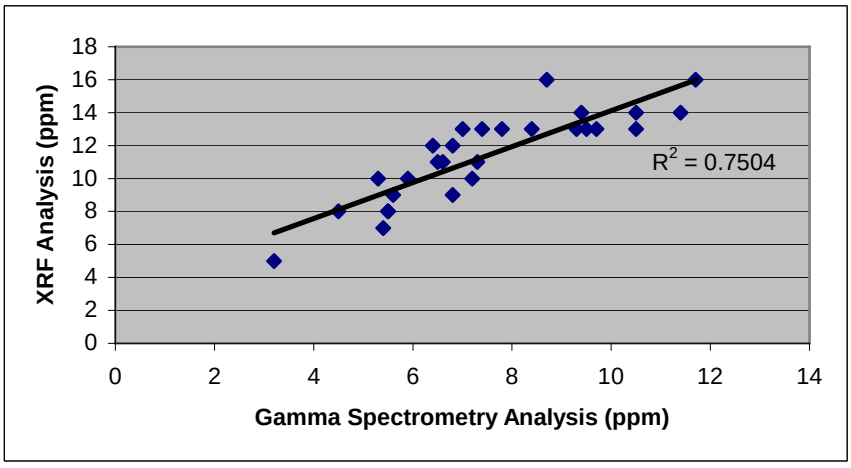


Figure 43- Comparison of thorium data by XRF and static ground gamma spectrometry analysis from Shirebrook and Warsop Vale

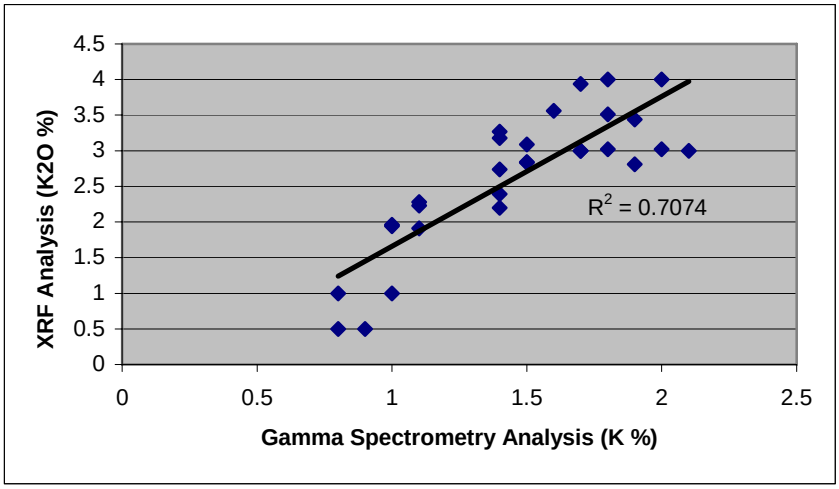


Figure 44 - Comparison of potassium oxide analysis by XRF and potassium by static ground gamma spectrometry analysis from Shirebrook and Warsop Vale.

3.4.7 Detailed comparison of airborne and ground-based gamma spectrometry

The HiRES-1 airborne data and the continuous ground-based gamma spectrometry display good agreement for U, Th and K (Figures 33-38). As was previously noted, the GTK survey data shows comparable, but generally higher values. Ideally for each airborne measurement point, the ground-based continuous measurements in the elliptical shaped 'footprint' of that airborne point would have been averaged to give an overall ground value for an area equivalent to the airborne measurement. A similar approach to this, taking ground measurements on an expanding hexagonal pattern beneath the centre of an airborne measurement, produced a good correlation between ground and airborne measurements (Sanderson *et al*., 2002). Further processing of the existing continuous ground-based data could be a possibility for future work in this direction.

The airborne and ground based gamma spectrometry gridded maps do show similar patterns of surface activity. This is well demonstrated at Warsop Vale (Figures 45- 47). Each survey highlighted the marked difference between the radiometric signature of the spoil heap and neighbouring agricultural land to the west and the increased level of detail in the ground-based data highlighted that the spoil itself is not a uniform mass. Different stages of extraction in the collieries history probably contributed several different rock types to the spoil.

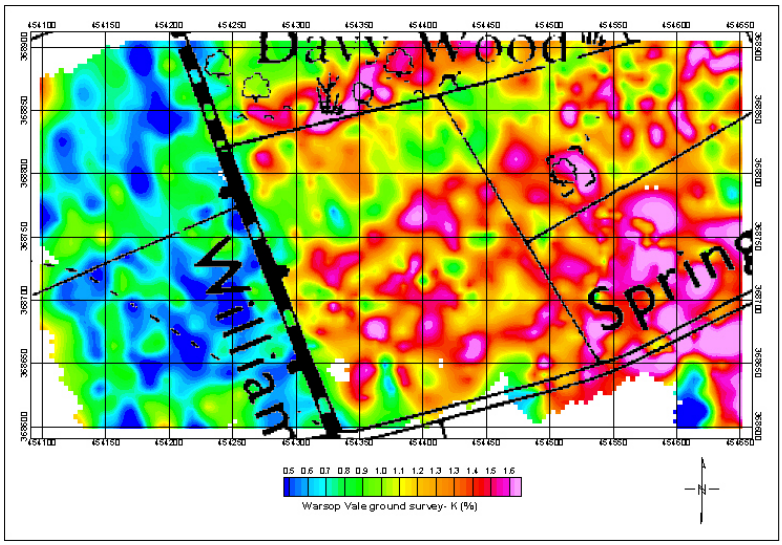


Figure 45- Close-up of continuous ground-based gamma spectrometry K (%) data at Warsop Vale.

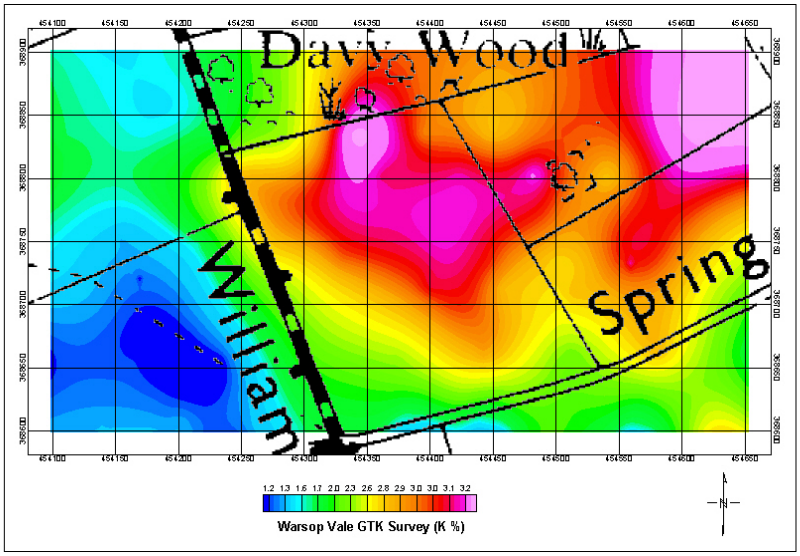


Figure 46- Close-up of GTK survey K (%) data at Warsop Vale.

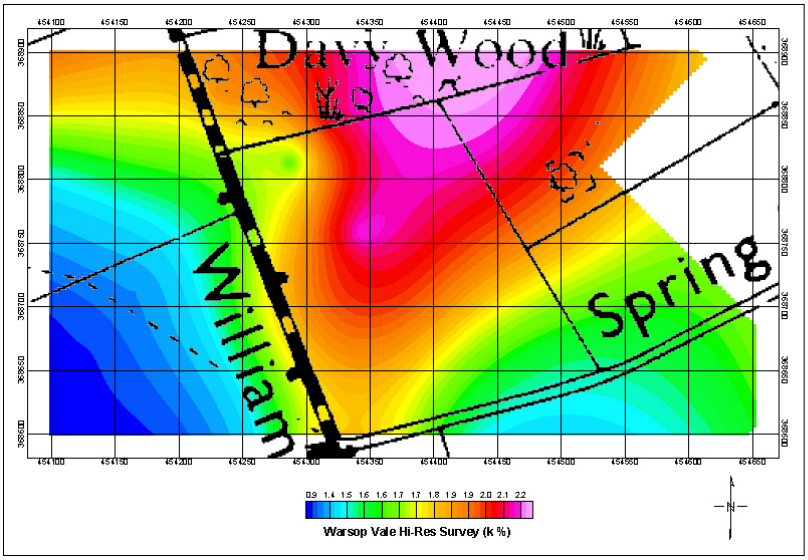


Figure 47- Close-up of HiRES-1 survey K (%) data at Warsop Vale.

3.5 NATURAL RADIOACTIVITY OF COLLIERY SPOIL AND COAL

Fossil fuels, like most other materials found in nature, contain trace quantities of the naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K (Beck, 1989). Through man's activities, technological enhancement of naturally occurring radioactive material occurs through mining, phosphate processing, fossil-fuel burning, oil and gas extraction and ore and heavy mineral processing.

Colliery spoil is a by-product of the mining and processing of coal. It consists mainly of shales and siltstones, with some sandstone and other rock types, which occur above, below and within coal seams and which are extracted with the coal during mechanised mining. This material is largely separated from the coal based on its higher density. The mostly non-coal material is tipped onto spoil heaps or disposed of in lagoons. Some older spoil heaps consist of partly burnt spoil (red shale), a product of the internal combustion of coal and other materials in the tip (Highley *et al*., 1997). This dominance of shale, and the presence of marine bands enriched in natural radioactivity, contribute significantly to the radiometric signature of colliery spoil.

Typical shales have an average uranium concentration around 3.7 ppm (Dale and Fardy, 1986; Krauskopf, 1979) but concentrations can reach more than 200 ppm (Wignall, 1994). Thorium concentrations in shale are typically around 12 ppm (Dale and Fardy, 1986; Krauskopf, 1979), but in black shales have been recorded up to 100 ppm.

In addition to the shales, siltstones and sandstones making up the colliery spoil, coal left over from the sorting process also contributes to the radiometric signature of the spoil. Although coal formations are usually amongst the least radioactive of the common sedimentary strata, considerable concentrations of uranium can occur in some coals. Typically these radioactive coals are thin and they usually form the topmost seam of a sequence overlain by strata from which uranium may be leached by downward-percolating waters. It is believed that coal adsorbs uranium (and other heavy elements) from these waters either as a result of an ion-exchange mechanism, or in the form of organo-metallic compounds. The radioactivity of coal seams is directly proportional to the ash content of the coal, the organic matter being virtually devoid of radioactive elements (Davidson and Ponsford, 1954). As the organic fraction of the coal does not contribute significantly to the radionuclide content, after combustion of the coal radionuclides can become concentrated in the fly-ash (Beck, 1998).

Average estimates of typical ^{238}U, ^{232}Th and ^{40}K ranges in bulk coal supplies are given in Table 5. Concentrations of these naturally occurring radionuclides more than an order of magnitude higher than those stated in Table 5 have however been recorded.

Table 5 – Typical radionuclide activity concentrations in bulk coal supplies (Beck 1998)

	^{238}U	**^{232}Th**	**^{40}K**
Average (Bq kg^{-1})	20	20	50
Typical range (Bq kg^{-1})	10-600	10-200	30-100

Most studies have found that all the members of the ^{238}U and ^{232}Th decay chains are usually in secular equilibrium in coal. There have been a few reports of disequilibrium, particularly between ^{210}Pb, ^{210}Po and ^{226}Ra in the ^{238}U series, presumably as a result of radon migration either into or out of the coal seam. These appear to represent atypical situations.

3.6 DOSE IMPLICATIONS OF ENHANCED RADIOACTIVITY OF COLLIERY SPOIL

Since the radioactivity of the spoil is significantly enhanced relative to the surrounding land it is worth considering briefly the dose implications to members of the public. This is particularly true since the former colliery sites are now accessible for recreational activities. The National Radiological Protection Board (NRPB), now the Radiation Protection Division of the Health Protection Agency, has defined Generalised Derived Limits (GDLs) as convenient levels against which the results of environmental monitoring can be compared (e.g. NRPB, 2000). They provide a conservative estimate of the level of a radionuclide in the soil that would give rise to a dose limit of 1 mSv per year. Monitored levels should not exceed 10 % of the GDL. If they do then a more detailed investigation should be carried out. The GDL for U is set at 20 kBq kg^{-1}.

The highest value for U, recorded in our studies at the Shirebrook and Warsop Vale sites, are equivalent to about 100 Bq kg^{-1} and are therefore well below the GDL. Taking the highest values encountered for K, eU and eTh (2.5%, 8 ppm and 20 ppm respectively) gives rise to an external dose of approximately 130 nGy h^{-1}. If a person spent the whole year at this hypothetical spot their annual dose would only amount to about 50 μSv. Admittedly this does not allow for other external and internal dose pathways, but suggests that the spoil does not present a significant radiological hazard unless there are significant hot spots that our surveys have missed.

4 Conclusions

1. Two high-resolution airborne geophysical surveys were conducted over parts of the UK in 1998 and 1999. Four radiometric anomalies in the East Shirebrook region were identified. These areas of elevated K, U and Th, relative to the surroundings, were identified as four colliery spoil tips: Shirebrook, Thoresby, Warsop Vale and Welbeck.
2. The radiometric data from the two airborne surveys correlate at the 99.95 % level for uranium ($r = 0.4$, $p = 0.0005$), thorium ($r = 0.60$, $p = 0.0005$) and potassium ($r = 0.68$, $p = 0.0005$). The GTK values are slightly higher, however, and this is probably the result of using the Finnish calibration range with very different rock types to those found near Shirebrook. The two surveys showed very similar patterns of radioactivity. They both distinguish well between areas of spoil and adjacent agricultural land. The higher spatial resolution (closer line spacing) of the GTK survey delineated the anomalies with a higher degree of detail. The GTK survey flying height was also lower than that of the HiRES-1 survey, thus reducing the sampling area, or 'footprint' from which the gamma radiation was averaged. This too increased the level of detail visible in the GTK survey results.
3. Ground follow-up work was undertaken at Shirebrook and Warsop Vale, the two disused collieries from the four radiometric anomalies identified by the airborne surveys in the East Shirebrook area. The ground survey entailed ground-based continuous gamma spectrometry traverses over accessible parts of each site, ground-based static gamma spectrometry measurements over transects of the sites, surface soil sampling at these transect points for laboratory XRF analysis and short core sampling at Warsop Vale for gamma spectrometry analysis on a core logger. The ground survey confirmed, and more accurately delineated the anomalies observed by the airborne surveys.
4. The ground-based continuous gamma spectrometry highlighted the marked difference between the radiometric signature of the spoil heap and neighbouring agricultural land particularly at Warsop Vale. This was particularly obvious in the potassium and total count data sets. A high degree of spatial variation was apparent in the ground-based gamma spectrometry data and highlighted that the spoil itself is not a uniform mass.
5. The extent of the spoil tips, as indicated by the radiometric data, does not always coincide exactly with the mapped boundaries of the spoil on Ordnance Survey 1: 50 000 or 1: 25 000 maps. This suggests that, in the absence of up-to-date maps, or where spoil extends beyond the mapped boundary, the radiometric data may be used to accurately define the current extent of the spoil. This may usefully be linked to EM data that show the extent of conductive (and potentially contaminated) groundwater (e.g. Beamish, 2002b, Klinck *et al*., 2004).
6. The results obtained by gamma spectrometry and XRF analysis correlate significantly. The results obtained by gamma spectrometry are generally slightly lower than the XRF measurements, as the gamma spectrometry method averages the gamma radiation from an area with an approximate radius of 10 m, whereas the XRF analysis is from a smaller sample volume. Disequilibrium in the natural radioactive decay series may also contribute to these differences, particularly for U, whilst for Th it could be that the XRF calibration is a factor.
7. U/Th ratios range from 0.18 to 0.37. The majority of the colliery spoil is made up of shale and siltstones, with other rock types which occur above, below and within coal seams and which are extracted with the coal during mechanised mining. The dominance of ^{232}Th in the spoil may, in part, be a result of differential U and radon daughter losses from the surface spoil layers, due to enhanced mobility in this unconsolidated material, and hence disruption to the secular equilibrium of the ^{238}U decay series. This could be

examined by comparing U/Th ratios for typical unweathered Coal Measures and overlying strata.

8. Uranium values from regional G-BASE soil sampling near to the sites were comparable to the XRF analysis carried out on soil samples obtained on the spoil tips. Thorium values, however were elevated from an average value of 7.1 ppm in the surrounding area to 11.2 ppm on the spoil. This agrees with Kestell's findings (Kestell, 2000), that thorium concentrations were most elevated above background levels.
9. Depth profiles of radionuclides in the cores were highly variable. Typical cores, one chosen from an area of relatively high radionuclide concentration and one from a lower area showed no discernible pattern in the depth distribution of the radionuclides.
10. Although reclamation of the two former collieries has been successful in terms of the appearance of the site and in the return of wildlife to the areas, it appears that the radiometric anomaly still exists post-reclamation. However, the U concentrations encountered are well below Generalised Derived Limits and the maximum external annual gamma dose is less than 50 μSv. There does not appear to be a significant radiological hazard to members of the public from an external dose perspective.

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