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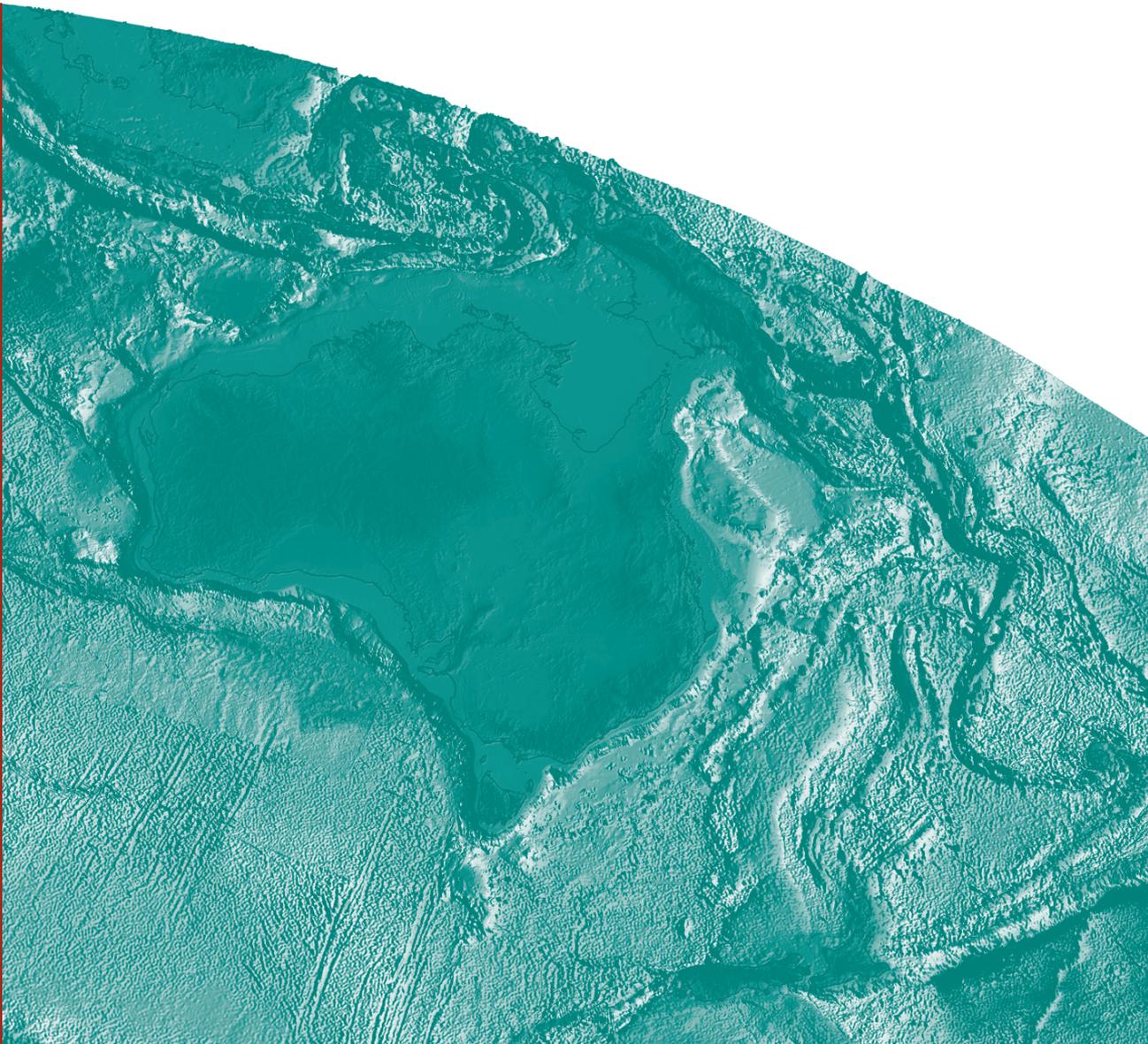
National Geochemical Survey of Australia: Comparison of Geochemical and Airborne Radiometric Data

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John Wilford, Patrice de Caritat, Brian Minty & Michelle Cooper

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GEOSCIENCE AUSTRALIA
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by

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Australian Government
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National Geochemical Survey of Australia: Comparison of Geochemical and Airborne Radiometric Data

Department of Resources, Energy and Tourism

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Geoscience Australia

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Executive Summary

The National Geochemical Survey of Australia (NGSA) provides the first national coverage of multi-element chemistry at a continental scale. The NGSA data is an important complement to other national-scale geological and geophysical datasets, particularly the Radiometric Map of Australia. The Radiometric Map of Australia shows potassium (K) measured directly from gamma-rays emitted when ^{40}K decays to argon (^{40}Ar), whereas thorium (Th) and uranium (U) do not emit gamma-rays. Instead, their abundances are inferred indirectly by measuring gamma-ray emissions associated with parent radionuclides (thallium-208 for Th, and bismuth-214 for U) within their radioactive decay chains. Airborne-derived grids provide a continuous prediction of these radioelements across the Australian landscape. In contrast, the NGSA data provide a series of precise single point geochemical measurements of surface (0-10 cm) and near-surface (~60-80 cm depth) unconsolidated catchment outlet sediments.

This report compares the measurements of K, Th and U from the surface (0-10 cm depth) sediment samples collected as part of the NGSA project with the inferred distribution of these elements from the national Radiometric Map of Australia. Specifically, the report explores the potential to develop improved estimates of airborne-derived K, Th and U concentrations based on calibration with surface geochemistry, and the degree to which the geochemical sample is representative of its associated catchment.

Three different methods of comparing the geochemical and radiometric data were considered: (1) point comparisons where sample locations were intersected with the corresponding grid cell (pixel) in the airborne dataset, (2) airborne averages based on 300 m and 600 m radius circles centred on each sampling point and (3) airborne averages of the corresponding sample point catchment. The 300 and 600 m averages were used to ensure that some of the values in the radiometric grid corresponded to values along the flightline and not just interpolated values between the flightlines (which typically are ~400 m apart). Geochemical samples that had a high proportion of coarse material (>2 mm) were excluded from the analysis because they are considered less likely to represent the bulk surface geochemistry measured by the airborne method.

The highest correlation between airborne-measured and ground-measured K, Th and U values was with the 300 m radius average with the samples with excessive coarse material removed. The coefficients of determination (r^2) derived between ground- and airborne-measured K, Th and U are 0.73, 0.73 and 0.33, respectively. All correlations are statistically significant at the $p < 0.01$ level given the sample size ($N \approx 900$). The airborne-measured concentrations under-estimate ground-measured concentrations for K and U, whereas Th is slightly over estimated compared with soil values. Calibration coefficients were developed to adjust the airborne values so as to more closely reflect the ground geochemical values. These coefficients should be considered when investigating quantitative relationships and concentrations of these elements in bedrock and regolith materials at the continental scale.

A series of catchment maps were generated showing (1) the difference between ground values and airborne-derived catchment means, and (2) the variability of airborne K, Th and U within each catchment. Both catchment classifications assist in understanding the degree to which the geochemical value is representative of its catchment. The concentration and variability of these elements largely reflects the geochemistry and mineralogy of the bedrock and regolith materials within the catchment and flow-linked associated catchments. Differential weathering of the catchment outlet sediments is a significant factor when comparing ground values to catchment averages. This is particularly relevant for soluble elements such as K that can readily be leached during weathering or pedogenesis.

Introduction

A 5-year Onshore Energy Security Program (OESP) was announced in 2006 to enable Geoscience Australia to deliver high quality pre-competitive geoscience information relating to onshore energy prospectivity (Johnson, 2006). The National Geochemical Survey of Australia (NGSA), which is part of this programme (Baldwin, 2007), collected transported regolith (sediment) samples from across the Australian continent and determined their inorganic chemical composition. The NGSA provides the only nation-wide, internally consistent geochemical dataset with state-of-the-art detection limits. It aimed to:

- Help calibrate and ground-truth the airborne radiometrics coverage of Australia (including addressing mother-daughter disequilibrium in the uranium-decay chain);
- Fill gaps in the existing airborne radiometric and geochemical coverages of Australia with quality data;
- Permit multi-element characterisation and ranking of radiometric anomalies (e.g., differentiation of uranium signatures from 'hot' granites, black shales or palaeochannels); and
- Provide fundamental data to enable first-order characterisation of geothermal hot-spots.

As such, the NGSA project (<http://www.ga.gov.au/energy/projects/national-geochemical-survey.html>) supports and adds value to a number of other OESP projects, particularly the Australia-Wide Airborne Geophysical Survey project (<http://www.ga.gov.au/energy/projects/awags.html>; Minty *et al.*, 2009a) and the Geothermal Energy project (<http://www.ga.gov.au/energy/projects/geothermal-energy.html>). Additionally, the NGSA results will have wider applications in mineral exploration for other commodities and natural resource management.

At its completion in June 2011, the NGSA delivered:

- A geochemical dataset that is national in scope, internally consistent and acquired through the application of state-of-the-art methods and instrumentation;
- A web-delivered geochemical atlas of Australia for some 68 elements showing for the first time what the concentrations of these elements are in surface materials and how they vary spatially; and
- Reports and papers documenting the project in detail and drawing implications on energy resource prospectivity and other applications.

Samples were collected from 1186 catchments (or 1315 sites, including duplicates), which together cover over 6.174 million km² or ~81% of Australia at the average sample density of 1 site per 5200 km² (statistics based on equal area projection). Approximately 200 catchments in South Australia and Western Australia could not be sampled during this project due to access limitations. Collaboration with State and Northern Territory geoscience agencies was critical for the completion of the project, particularly regarding the sampling phase.

Sampling procedures were reported in Lech *et al.* (2007) and field data was compiled in Cooper *et al.* (2010). Sample preparation protocols were documented in Caritat *et al.* (2009) and sample analysis methods were detailed in Caritat *et al.* (2010). A data quality assessment is presented in Caritat & Cooper (2011a), and all geochemical maps derived from the NGSA are collated in Caritat & Cooper (2011b). In the present report, we undertake a preliminary comparison between the NGSA data and airborne radiometric data relating to the concentrations and distributions of the radioelements K, Th and U. We show how the relationships between measured K, Th and U and airborne gamma-ray spectrometry can be used to calibrate the radiometric grids to provide improved mapping of these elements across the Australian landscape.

Background to the Project

The NGSa project aimed to provide pre-competitive data and knowledge to support exploration for energy resources in Australia. In particular, it improved existing knowledge of the concentrations and distributions of energy-related elements such as uranium (U) and thorium (Th) at the national scale.

The project was underpinned by a series of pilot geochemical surveys carried out in the previous four years by Geoscience Australia and the Cooperative Research Centre for Landscape Environments and Mineral Exploration (CRC LEME) to test robust and cost-effective protocols for sample collection, preparation and analysis. Examples of these are the Riverina (Caritat *et al.*, 2005; Caritat *et al.*, 2007), the Gawler (Caritat *et al.*, 2008a) and the Thomson (Caritat & Lech, 2007; Lech and Caritat, 2007) pilot geochemical surveys. Selected results from these pilot projects were summarised in Caritat *et al.* (2008b).

The current national project, briefly described below, was conducted in collaboration with all the State and the Northern Territory geoscience agencies.

RATIONALE

The national geochemical survey was initiated because of the absence of a complete geochemical coverage for Australia and because such a data layer is an important complement to national-scale geological and geophysical datasets (Caritat *et al.*, 2008c).

The distribution of geochemical data available at the commencement of the survey through the national repository (OZCHEM database) is shown in Figure 1. The map shows that there are vast areas of the country (>60 %) that lack geochemical information. Where geochemical data are available in the public domain, they may often not be internally comparable as a result of:

- Inconsistent sampling material (e.g., rocks of various types and/or degree of alteration, mineralisation or weathering);
- Inconsistent sample preparation methods (e.g., total analyses *versus* partial digests with weak acids);
- Differences in instrumentation used and between laboratories, leading to variable lower limits of detection (LLD) between datasets (e.g., older *versus* state-of-the-art instruments);
- Lack of metadata on data quality (e.g., instrument calibration, bias, precision, sample type description, replicates, etc.); and
- Variable suite of elements analysed (e.g., sometimes a very limited suite such as gold (Au) only or Au + copper (Cu)).

Although a significantly improved nation-wide, levelled coverage of airborne gamma-ray spectrometric (radiometric) data (Minty *et al.*, 2009a,b) became available (Figure 2) during the course of the NGSa project, field calibration of radiogenic elements potassium (K), uranium (U) or thorium (Th) is perhaps not as systematic as is desirable and the question of disequilibrium in the radiogenic decay chain is poorly constrained as a result. It is hoped the NGSa data, in combination with the new radiometric coverage, will yield insights into this problem.

Some regional geochemical surveys have been carried out in parts of Australia (e.g., Morris *et al.*, 1998; Cornelius *et al.*, 2008; the pilot geochemical surveys mentioned above), but until this year no national coverage existed. The modern concept of regional geochemical surveys was first developed in the 1960s, and it has since proven to be a reliable tool for mineral exploration and other applications at various scales (Garrett *et al.*, 2008; Smith & Reimann, 2008).

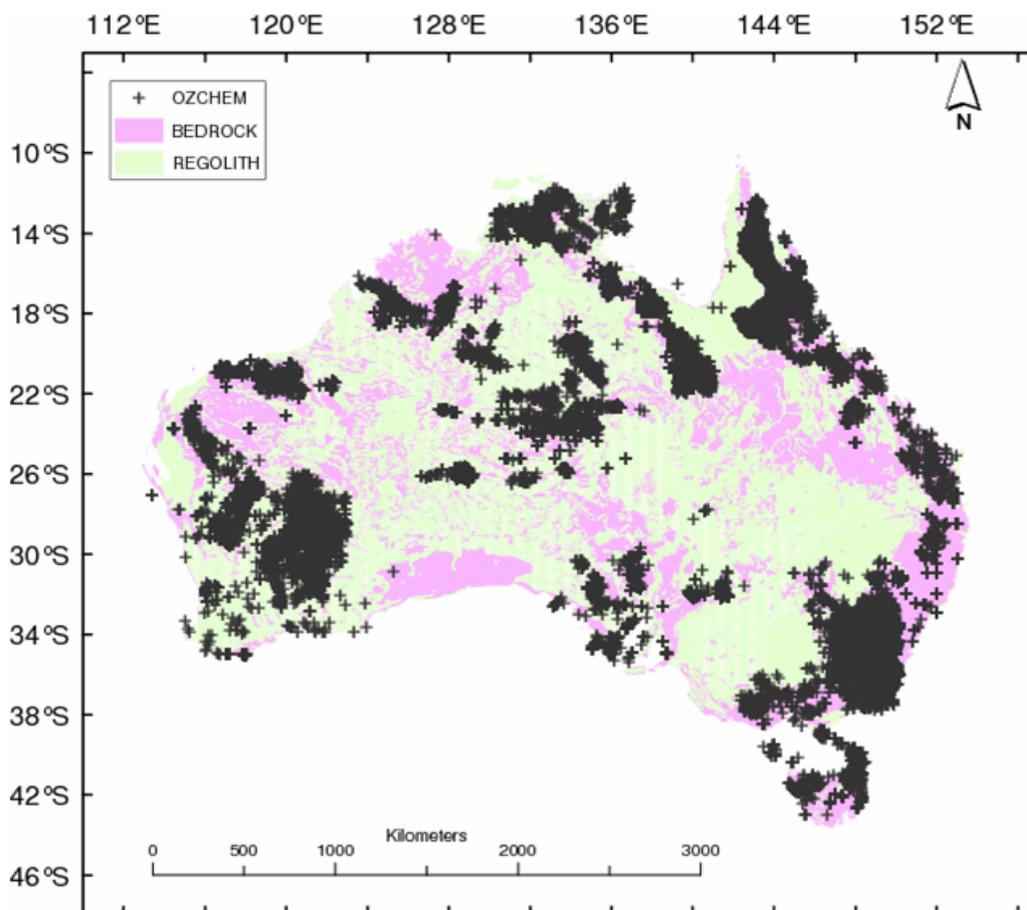


Figure 1. Distribution of whole rock geochemical data in Australia (plus signs) extracted from the OZCHEM national database as at June 2006, overlain on bedrock and regolith coverage.

OBJECTIVES

The objectives of the NGSa project were to:

- Collect transported regolith samples at the outlet of large catchments covering >90 % of Australia using an ultra low sampling density approach;
- Prepare and analyse the samples to extract the maximum amount of geochemical information (60+ elements/parameters) using internally consistent, state-of-the-art techniques;
- Populate the national geochemical database with the resulting new data; and
- Compile an atlas of geochemical maps for use by the mineral exploration industry to identify areas of interest in terms of energy-related resources and other mineral commodities, which can then be the focus of targeted exploration efforts.

STRATEGY

The sampling method was adapted to Australian landscape and climate conditions (e.g., importance of aeolian landforms in some areas, climate ranging from tropical to arid). It was fine-tuned and field-tested during the Riverina, Gawler and Thomson pilot projects (e.g., see Caritat *et al.*, 2008b). The cost of a national survey was minimised by applying an ultra low sampling density approach (generally accepted to mean between 1 site/1000 km² and 1 site/10,000 km²).

The strategy adopted for the national geochemical survey is briefly described below.



Figure 2. Ternary image (K-red, eU-blue, eTh-green) of Australia derived from the new levelled National Radioelement Database (Minty *et al.*, 2009b)

Sampling medium

Catchment outlet sediments (similar to floodplain sediments in most cases) were sampled at two depths (0-10 cm below the surface as well as at a depth of between ~60 and 80 cm on average). The term 'catchment outlet sediment' is deliberately chosen because it is more general than 'floodplain sediment' to allow for those cases where aeolian influence is important in the regolith formation process.

Sampling sites

Initially 1385 catchments covering 91% (or about 7 million km²) of Australia across all States and Territories were targeted for sampling (Figure 3). Five catchments were assigned a second sampling site (i.e., target site) due to their size and topography so in total, it was expected that 1529 samples (1385 catchments + 10 % of which are sampled in duplicate + 5 large catchments sampled twice) would be collected. Most catchments were sampled near their outlet, while those exhibiting internal or poorly defined drainage were sampled at, or as close as possible to, their lowest point. Lech *et al.* (2007) give details of the method for determining sampling sites. Catchments smaller than 1000 km² (mostly coastal) and small islands were not included in the survey. The resulting distribution of catchment outlet sites targeted for sampling is shown in Figure 4 and translates to an average sampling density of around 1 site/5500 km².

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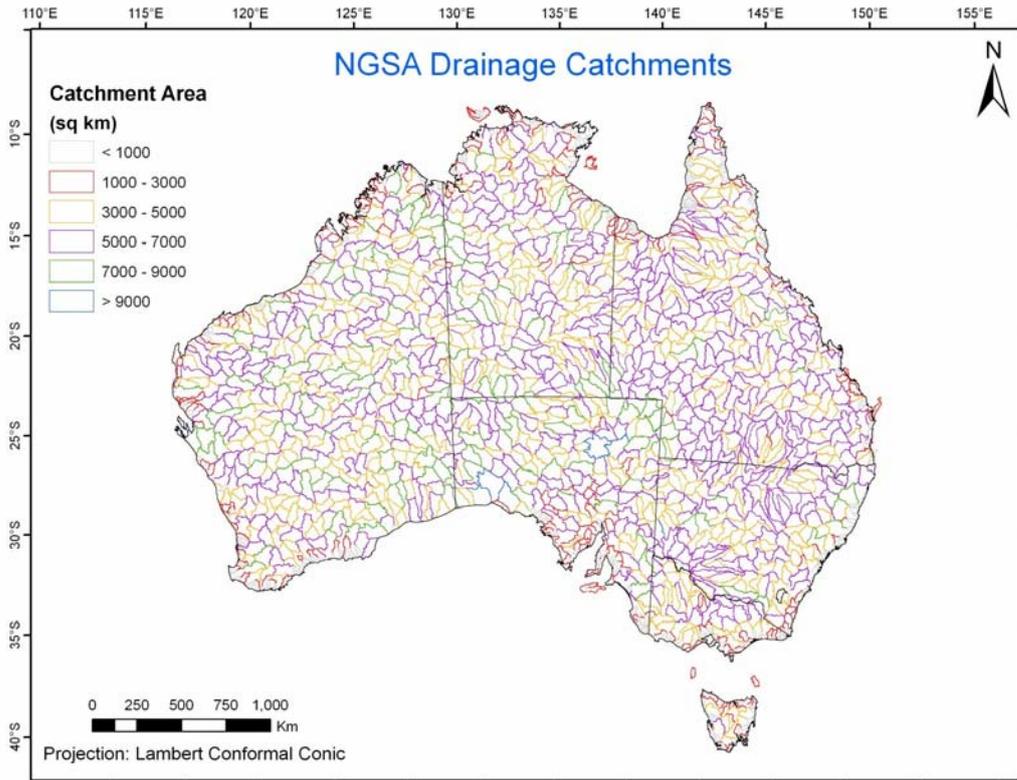


Figure 3. Distribution of catchments for the National Geochemical Survey of Australia.

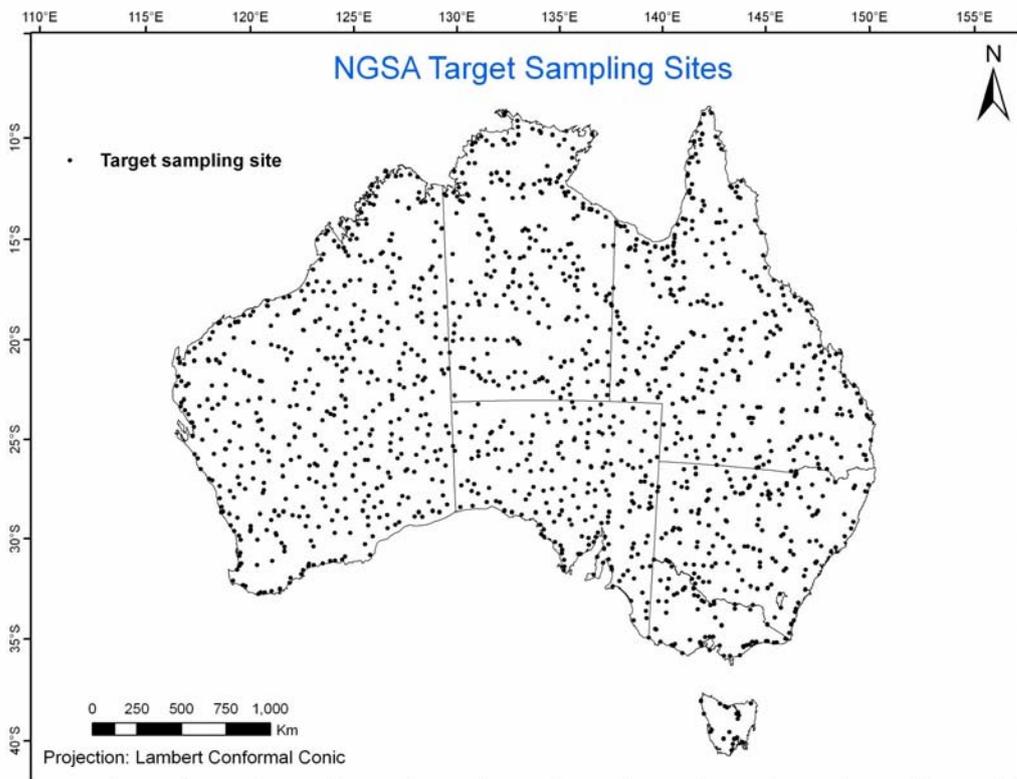


Figure 4. Distribution of initial target sampling sites for the National Geochemical Survey of Australia.

Sample collection

A detailed Field Manual was compiled (Lech *et al.*, 2007) and all sampling equipment and consumables were centrally provided. Despite best efforts to access all target sites, only 1186

catchments could be sampled before the deadline for field work was reached. Of these 1186 catchments, 123 were sampled in duplicate (on average ~200 m away from the original site) for quality control purposes, and six of the largest catchments were sampled at two widely separated locations (on average 69 km apart), yielding a total of 1315 sampled sites (Figure 5). At each site, a Top Outlet Sediment (TOS) sample was collected from 0-10 cm (below the root zone, if applicable), and a Bottom Outlet Sediment (BOS) sample was collected from a depth of (on average) between 60 and 80 cm. To reduce natural soil heterogeneity, every sample collected was a composite either from a shallow soil pit (TOS) or from at least three auger holes or, rarely, a pit (BOS) at a given site (see Lech *et al.*, 2007, for more information). An average of 9 kg of sediment was collected per sample.

Sample collection was carried out by the State and Northern Territory geoscience agencies following a hands-on, in-field training period with the Geoscience Australia NGS team. At each locality a detailed site description, field pH, and dry (if possible) and moist Munsell® soil colours were recorded and several digital photographs were taken (Cooper *et al.*, 2010). All information was recorded digitally to facilitate subsequent uploading into databases.

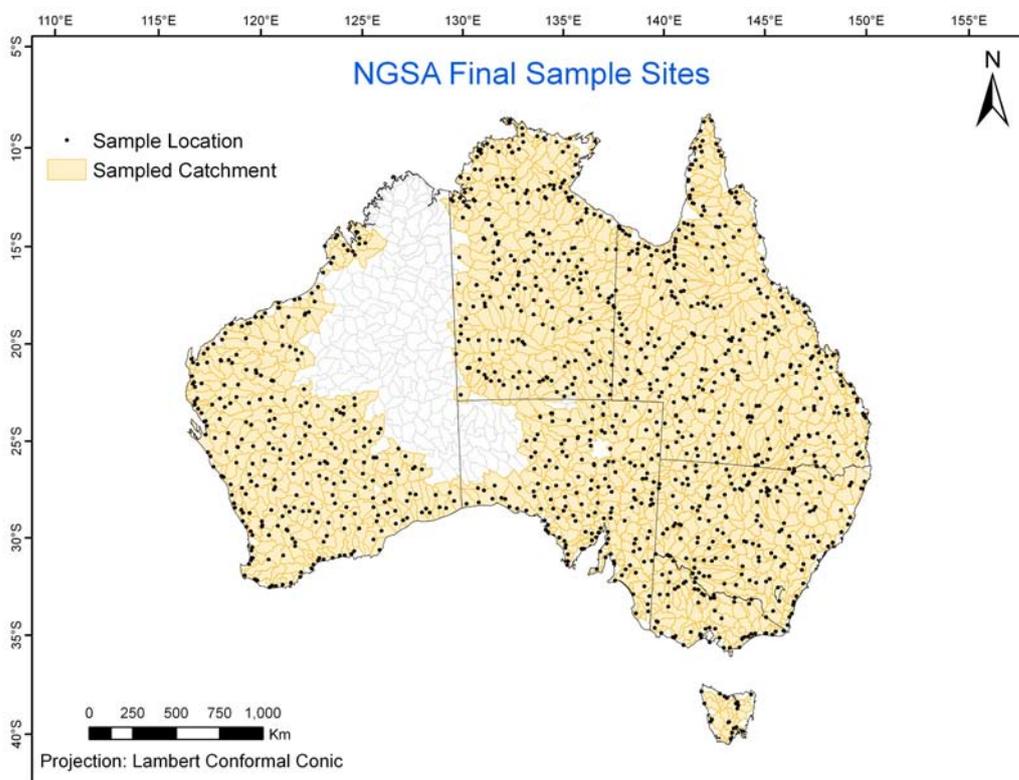


Figure 5. Final distribution of catchments sampled for the National Geochemical Survey of Australia (1186 catchments, or 86 %, completed).

Sample preparation

All samples were sent to Geoscience Australia for processing, where they were air-dried at 40 °C and homogenised. A bulk split (~50 %) of each sample was archived for future investigations. The remainder was riffle split and dry sieved to <2 mm and <75 µm fractions. The <2 mm fraction was mechanically ground for some analyses, while the finer fraction was not. The sample preparation protocols were discussed in detail in the NGS Sample Preparation Manual (Caritat *et al.*, 2009).

Sample analysis

The analysis philosophy from the outset was to apply a multi-element approach on two grain-size fractions prepared by at least two digestion methods, in order to maximise the amount of

geochemical information extracted from the samples. Sample analysis was carried out for 68 elements (Total element content) using X-ray fluorescence (XRF) and (reaction cell) inductively coupled plasma-mass spectrometry (ICP-MS) at Geoscience Australia. The ICP-MS analyses were carried out on a total digest (HF + HNO₃) of fragments of the XRF beads (Pyke, 2000). Additional parameters recorded at Geoscience Australia were pH 1:5 (soil:water), electrical conductivity (EC) 1:5 (soil:water), and laser particle size analysis (LPSA). Analyses for selected elements not available at Geoscience Australia (e.g., Au, fluorine (F), selenium (Se), and platinum group elements (PGEs)) were undertaken externally. Additional digests/analyses (e.g., after Aqua Regia digestion, ligand-based extractions, near visible-infrared spectroscopy) were also carried out externally. The Mobile Metal Ion analysis (MMI™) was made possible through collaboration with SGS Minerals Services. Details of the sample analysis protocols are discussed in the NGSA Analytical Methods Manual (Caritat *et al.*, 2010).

Quality assessment/quality control

Sample numbers were randomised to minimise regional bias, help separate false from true anomalies and obtain meaningful estimates of the variance of duplicates. Field duplicates, analytical duplicates, internal standards and certified reference materials were introduced at regular intervals in the analytical streams. Care was also taken throughout the project to minimise contamination, cross-contamination and mislabelling risks. Details are discussed in the NGSA Data Quality Assessment report (Caritat & Cooper, 2011a).

Data analysis

National-scale geochemical maps were produced and released as a Geochemical Atlas (Caritat & Cooper, 2011b) and a web-based map collection (<http://www.ga.gov.au/ngsa>). Reports providing preliminary interpretations and including graphical and statistical analysis will be prepared separately and released/announced via the above Geoscience Australia website.

Timeline

Following planning in the first half of 2007, fieldwork, including initial training, began in mid-2007 and concluded in late 2009. Figure 5 shows the final distribution of catchments actually sampled for NGSA at the end of fieldwork (December 2009). Sample preparation started in early 2008 and concluded in December 2009. Sample analysis started late 2008 and finished in late 2010. Data analysis and reporting took place in 2010 and early 2011. The project concluded in late June 2011, with a dataset and product release at the Association of Mining and Exploration Companies (AMEC) convention in Perth, 28-30 June 2011.

Comparison of geochemical and airborne radiometric data

BACKGROUND

Data collected as part of the National Geochemical Survey of Australia (NGSA) and airborne radiometric data are complementary. The geochemical survey collected and analysed a surface sample (0-10 cm depth) and a subsurface sample (on average 60-80 cm depth) at each site. Locations of these sample sites and associated catchments are shown in Figure 5. Major, minor and trace elements (total) concentrations, including those of K, Th and U, were quantitatively determined using X-ray fluorescence (XRF) and inductively coupled plasma-mass spectrometry (ICP-MS) (see Caritat *et al.*, 2010).

Airborne gamma-ray spectrometry measures the surface distribution of K, Th and U to a depth of approximately 35 cm. Airborne measurements are acquired at a much higher density (every ~60 m along the flightlines, which typically are ~400 m apart) than the geochemical data and are gridded to generate raster images showing variations of these elements across the landscape (Figure 2). K abundance is measured directly as gamma-rays emitted when ^{40}K decays to ^{40}Ar . Th and U abundances, however, are derived indirectly by measuring gamma-ray emissions associated with parent radionuclides ^{208}Tl (thallium) and ^{214}Bi (bismuth), respectively (Dickson & Scott, 1997). Accurate predictions of Th and U assume that the measured parent radionuclides are in secular equilibrium within their decay series, including Th and U themselves. Disequilibrium is not common for Th, but is a problem when estimating U (Minty, 1997). U disequilibrium occurs where soluble parent radionuclides occurring above the measured ^{214}Bi in the decay chain, particularly ^{226}Ra , are either enriched or removed, thereby giving an over- or under-estimate of U, respectively (Minty, 1997). Th and U concentrations derived from gamma-ray spectrometry are normally expressed in units of 'equivalent' parts per million (eTh ppm and eU ppm) because the concentrations are based on this assumption of equilibrium in their respective decay series.

Geochemical data from the NGSA project have the potential to be used for assessing the accuracy of the airborne radioelement grids of K, eTh and eU. Conversely, the distribution of radioelements from airborne data enables assessment of the degree to which the site measurements are representative of the broader landscape or catchment. The aims of the present comparison of both methods include to:

1. Assess the relationships between surface and near-surface geochemistry with airborne measured K, eTh and eU,
2. Develop regression coefficients, where appropriate, to improve the estimation of airborne measured K, eTh and eU for geochemical mapping and modelling, and
3. Assess the degree to which the catchment outlet sediment sample is representative of the whole catchment.

METHODS

The NGSA collected compositional data for a Top Outlet Sediment (TOS) sample collected from 0 to 10 cm depth, and a Bottom Outlet Sediment (BOS) sample collected from between 60 and 80 cm on average, at 1315 sites covering over 80% of Australia. Only the TOS samples were analysed in this study because they correspond to the material measured by airborne spectrometry. The radiometric data are based on the radiometric map of Australia, which represents levelled and merged composite K, eTh and eU grids over Australia at 100 m resolution (Minty *et al.*, 2009b). Three different methods of comparing the geochemical and radiometric data were considered in this

study. First, the NGSa data was compared with the radiometric data for the pixel from the airborne dataset at the same location. Second, NGSa measurements were compared to airborne averages based on 300 m and 600 m radius circles centred on each NGSa sampling point. Third, NGSa sample measurements were compared to the airborne averages for K, Th and U for the whole catchment.

As part of the sample preparation for the NGSa project the TOS and BOS samples were sieved to <2 mm and <75 µm fractions. Only the coarse (<2 mm) grain-size fraction subsample was used in this investigation because it is likely to reflect more closely the bulk surface soil composition that is measured by airborne spectrometry. Sample sites (with field duplicate samples removed) totalled 1192; of these sites not all had radiometric coverage (because of data gaps in the national radiometric coverage) leaving a total of 1046 samples for statistical analysis. A second culled subset of 933 sites was generated by removing samples with more than 25% by weight of material that was greater than 2 mm in size. The rationale behind this culling is that if there is so much coarse material, which is not analysed in the geochemical samples, then there is little scope that the geochemical analysis and the radiometric measurement are comparable. Both the large (N = 1046) and smaller (N = 933) sample subsets were analysed.

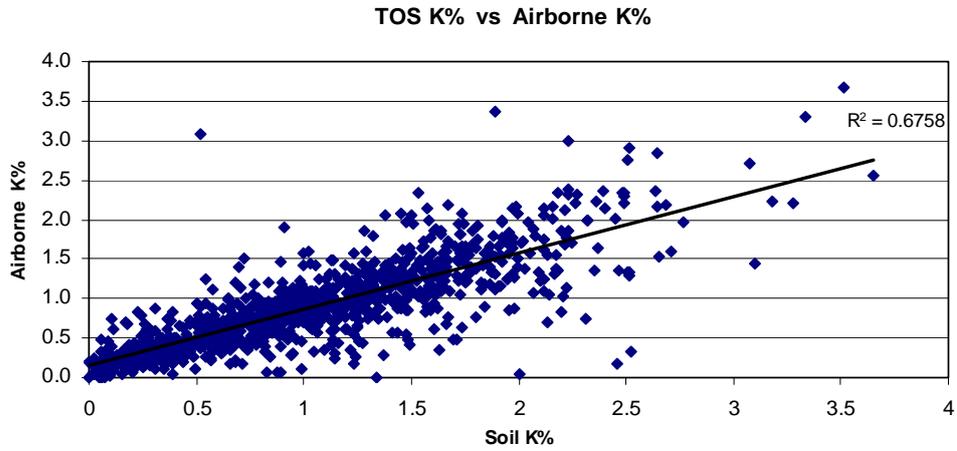
Scatter plots and associated correlation regression lines were generated for all the above combinations. Point radius averages and catchment statistics were determined using the ZONALSTATISTICS command in ArcInfo GIS.

RESULTS

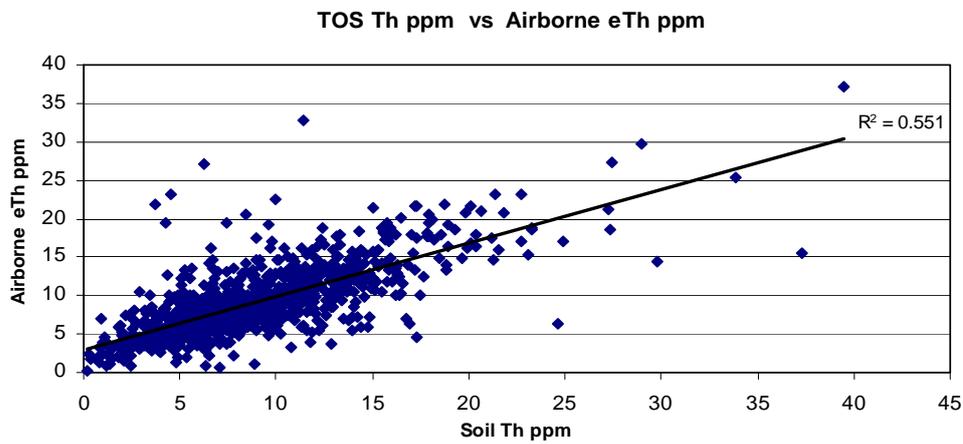
Point and area correlations between geochemistry and airborne data

Figures 6 to 11 show scatter plots comparing the airborne-derived and soil-measured concentrations of K, Th and U for single pixel, and 300 m and 600 m radius averages of the radiometric data, first with all samples, then with samples with excessive coarse material removed (see above).

a)



b)



c)

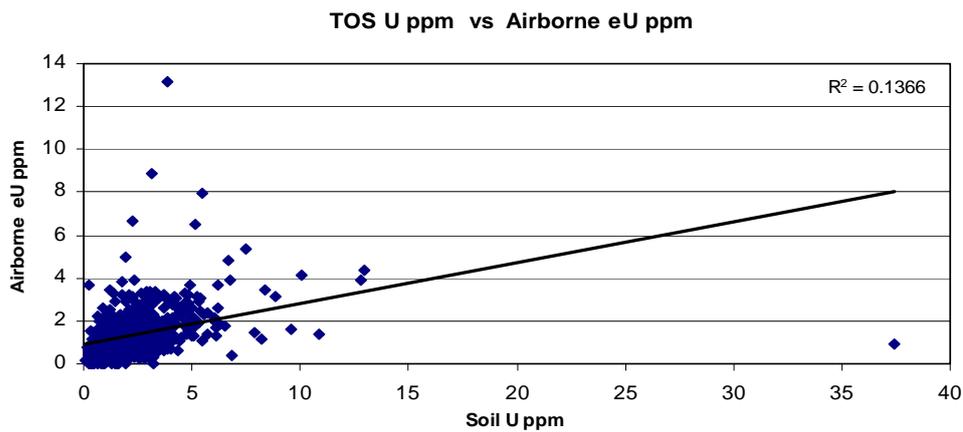
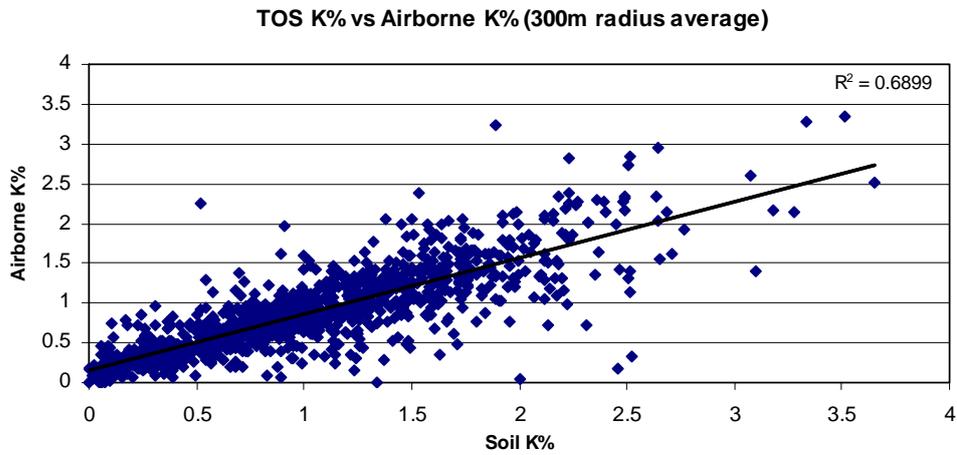
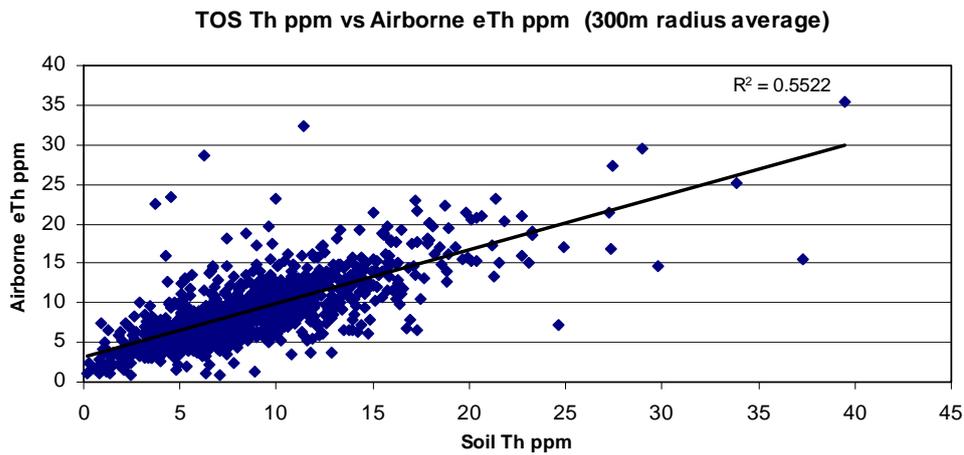


Figure 6. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on single pixel values.

a)



b)



c)

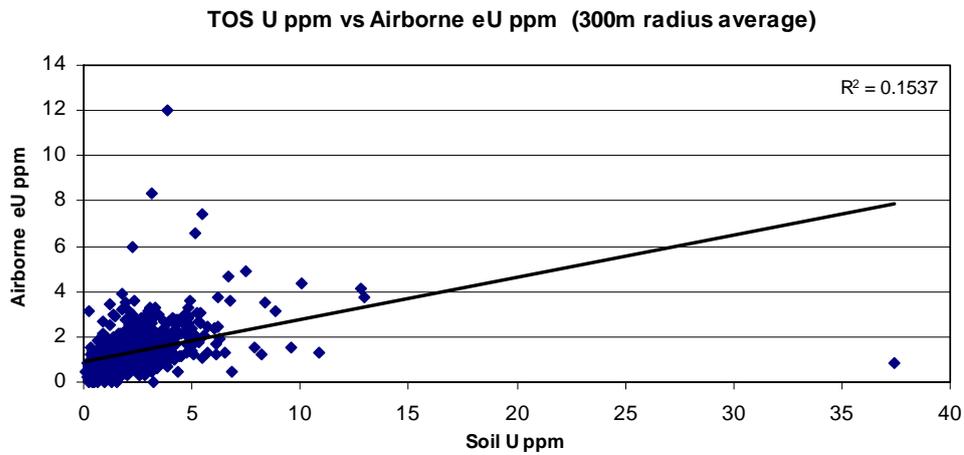
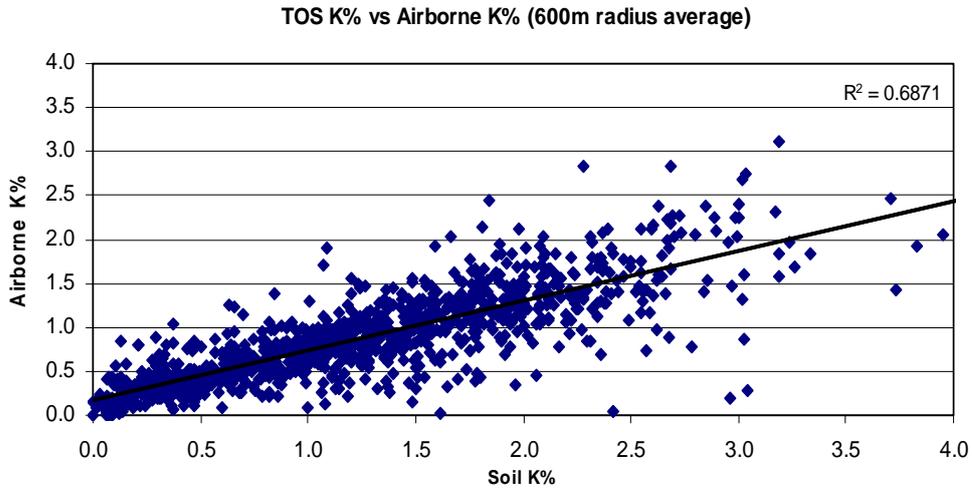
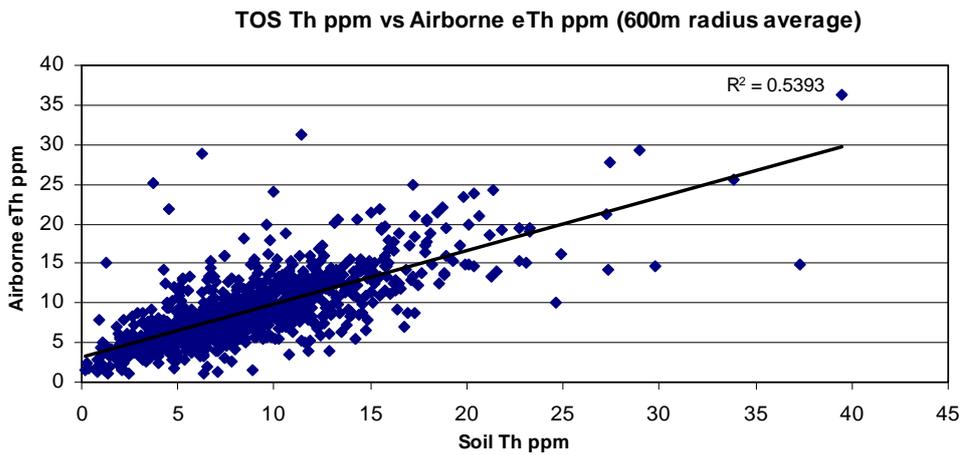


Figure 7. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on 300 m radius averages.

a)



b)



c)

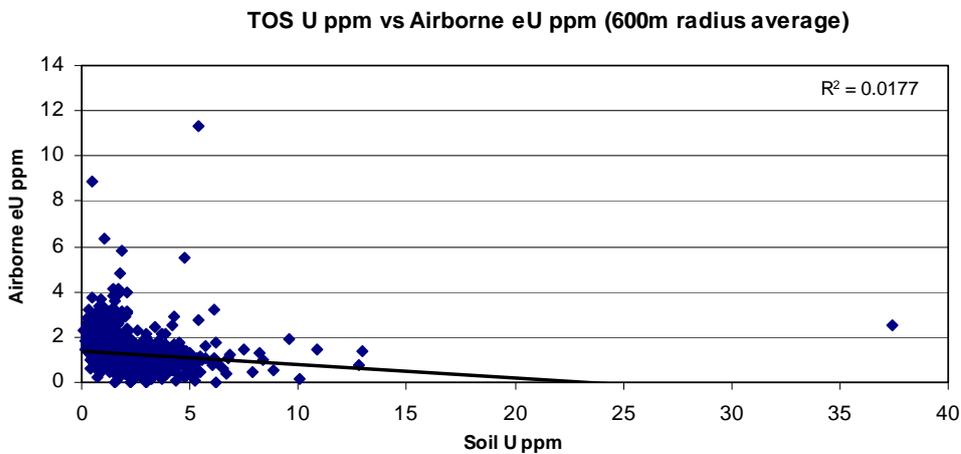
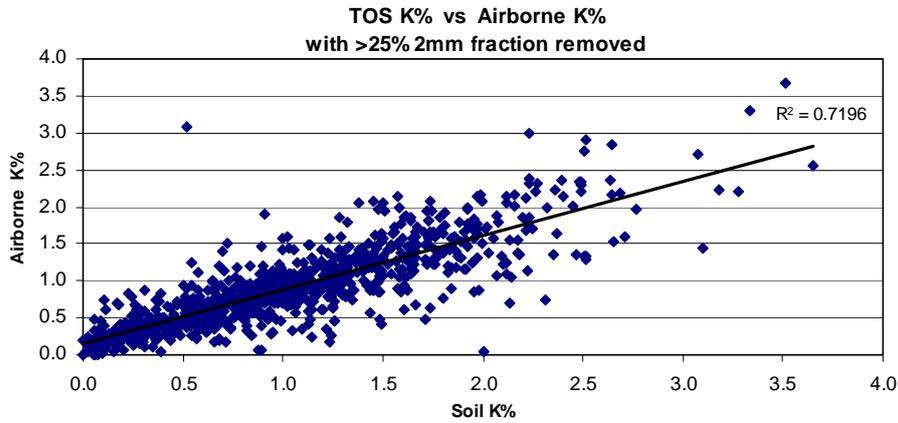
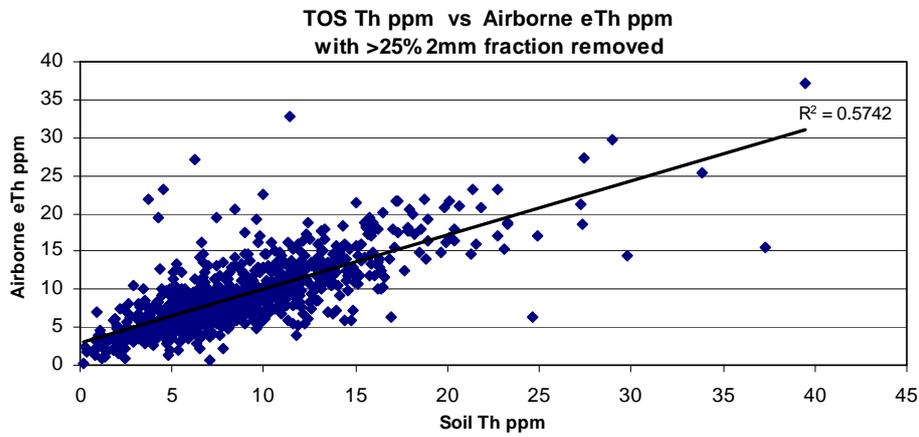


Figure 8. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on 600 m radius averages.

a)



b)



c)

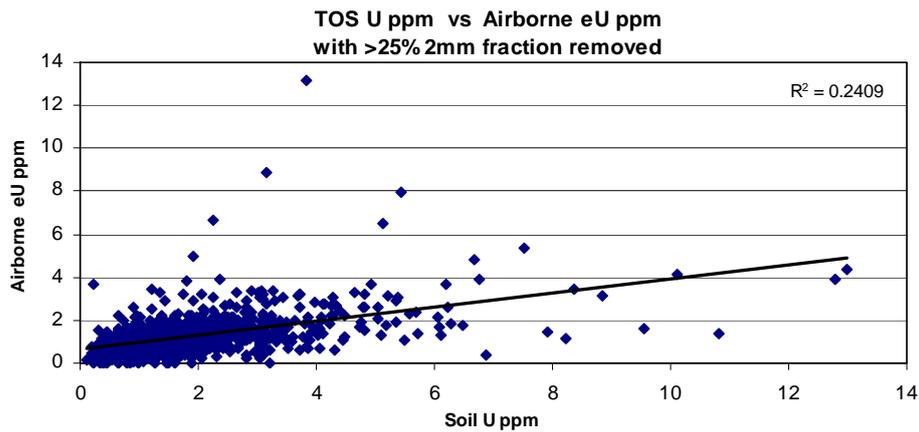
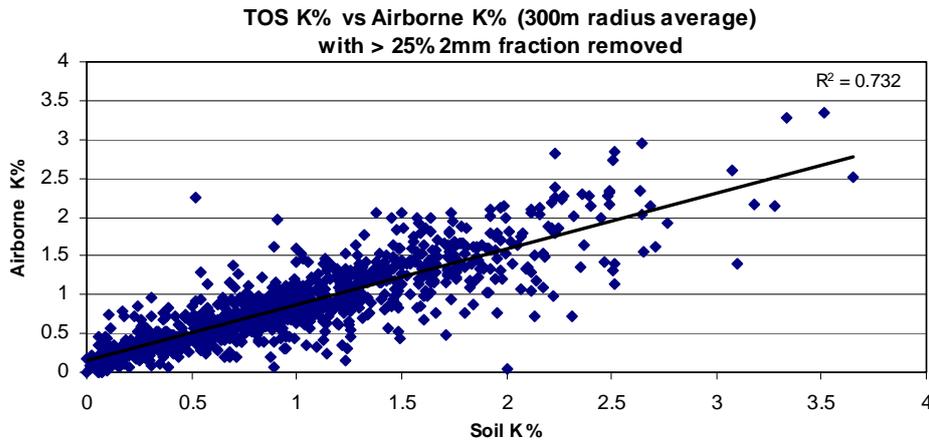
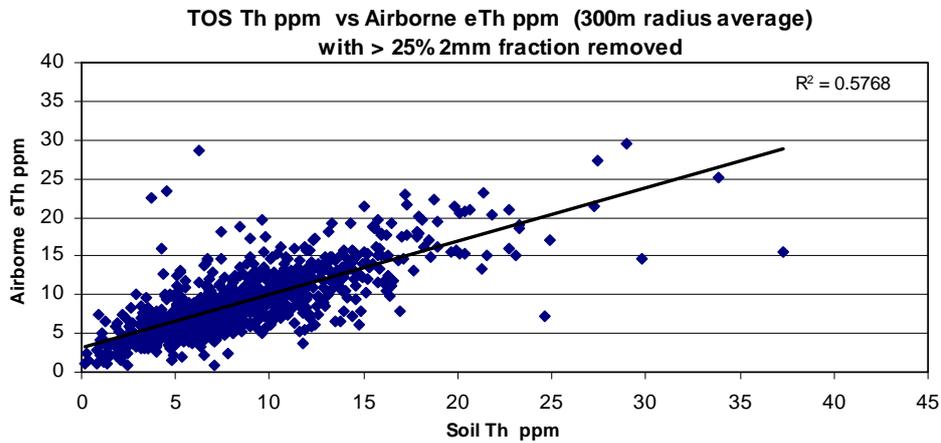


Figure 9. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on single pixel values, after the removal of samples that had more than 25% of the total sample weight above 2 mm size fraction.

a)



b)



c)

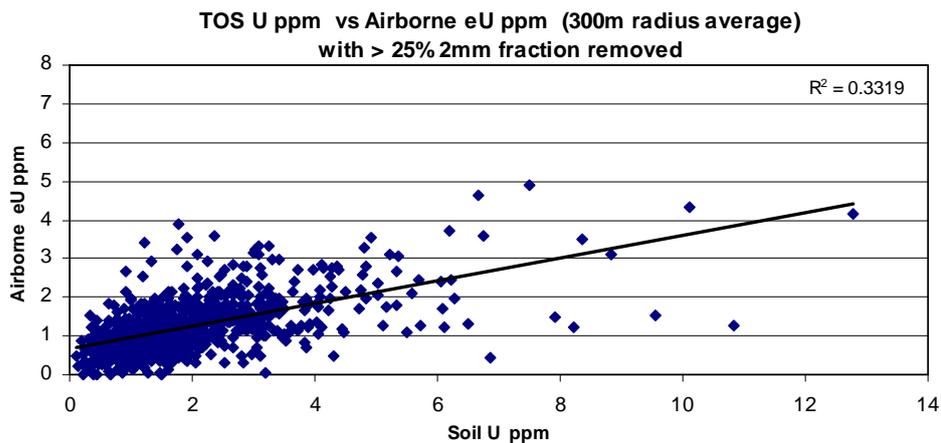
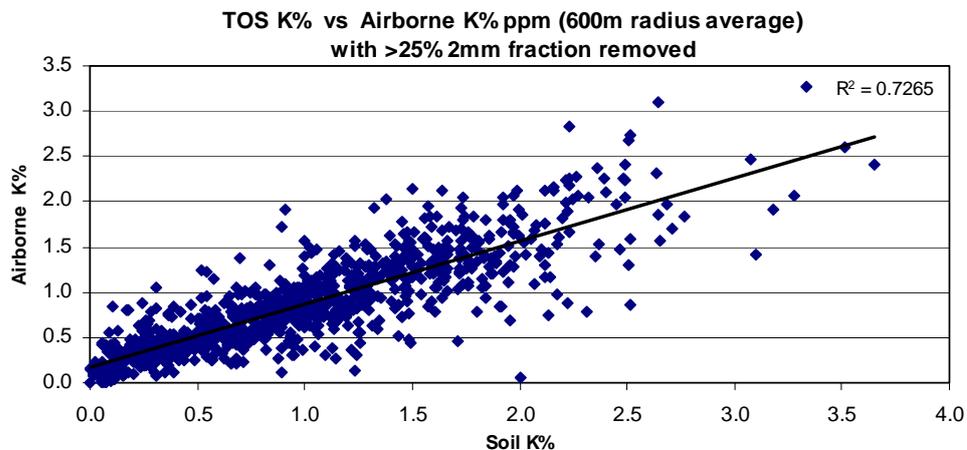
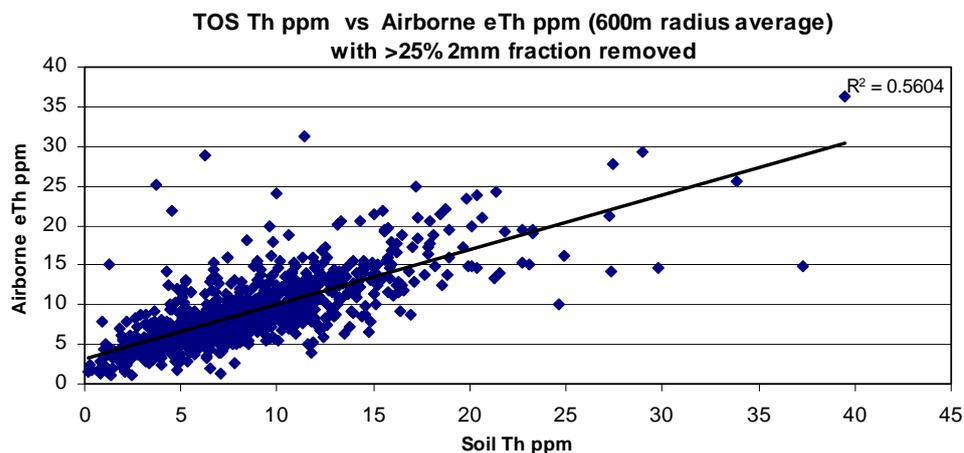


Figure 10. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on 300 m radius averages, after the removal of samples that had more than 25% of the total sample weight above 2 mm size fraction.

a)



b)



c)

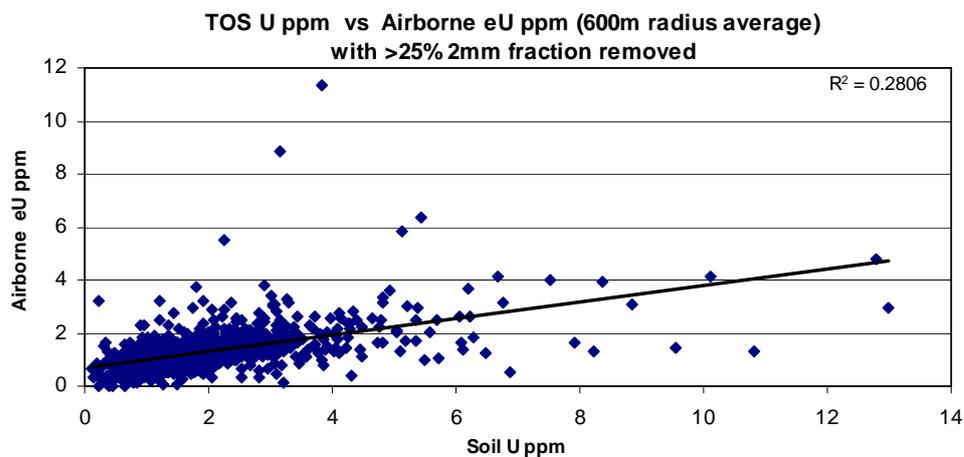


Figure 11. Scatter plots and coefficients of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for K (a), eTh (b) and eU (c) values based on 600 m radius averages, after the removal of samples that had more than 25% of the total sample weight above 2 mm size fraction.

CATCHMENT BASED ANALYSIS

Relationships between soil geochemistry and airborne derived catchment means for K, eTh and eU.

Figures 12 to 14 highlight the differences between the geochemical concentrations of soil-measured K, Th and U to their corresponding catchment means of airborne measured K, eTh and eU. In addition to the catchment means for the three radioelements, Th point differences are also shown (Figure 15) to illustrate that the spatial patterns observed in the catchment average are also observed in the point comparisons. Negative numbers indicate that geochemical values are higher than mean catchment values derived from the radiometric data. Positive values indicate lower mean values compared to the geochemical measurement. Colours are scaled into 8 quantiles with the 4 central quantiles (25th to 75th percentiles) coloured yellow. Differences are expressed in wt% (weight percent) for K and in ppm (parts per million) for Th and U. The uncoloured catchments (white regions) have not been sampled and those catchments in black have insufficient radiometric coverage to determine a representative catchment mean value for the element.

The figure below shows the difference between airborne catchment mean potassium and geochemical value (wt%).

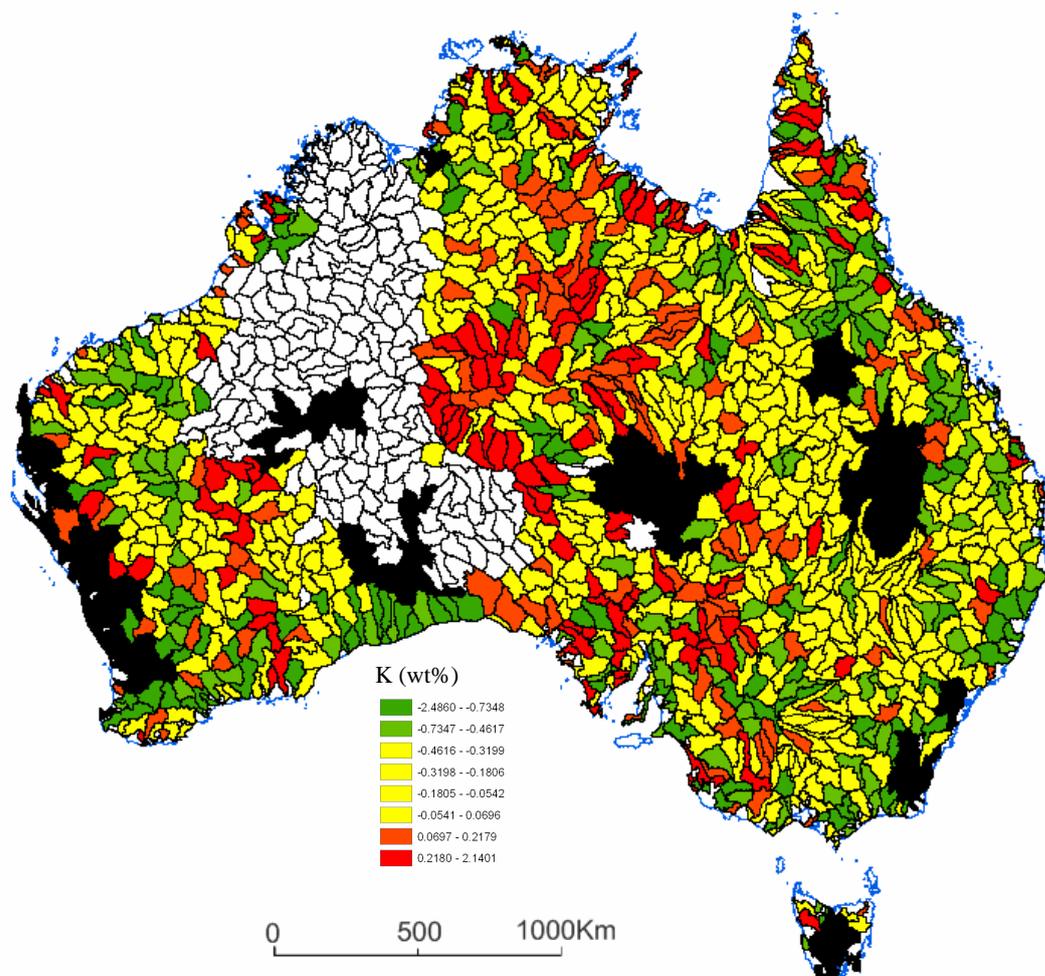


Figure 12. Difference between radiometric catchment mean and K soil geochemistry. Greens and reds indicate geochemical values are considerably higher or lower than the catchment radiometric mean, respectively. Yellows indicate broadly similar values between the two measurements.

The figure below shows the difference between airborne catchment mean eTh and geochemical value.

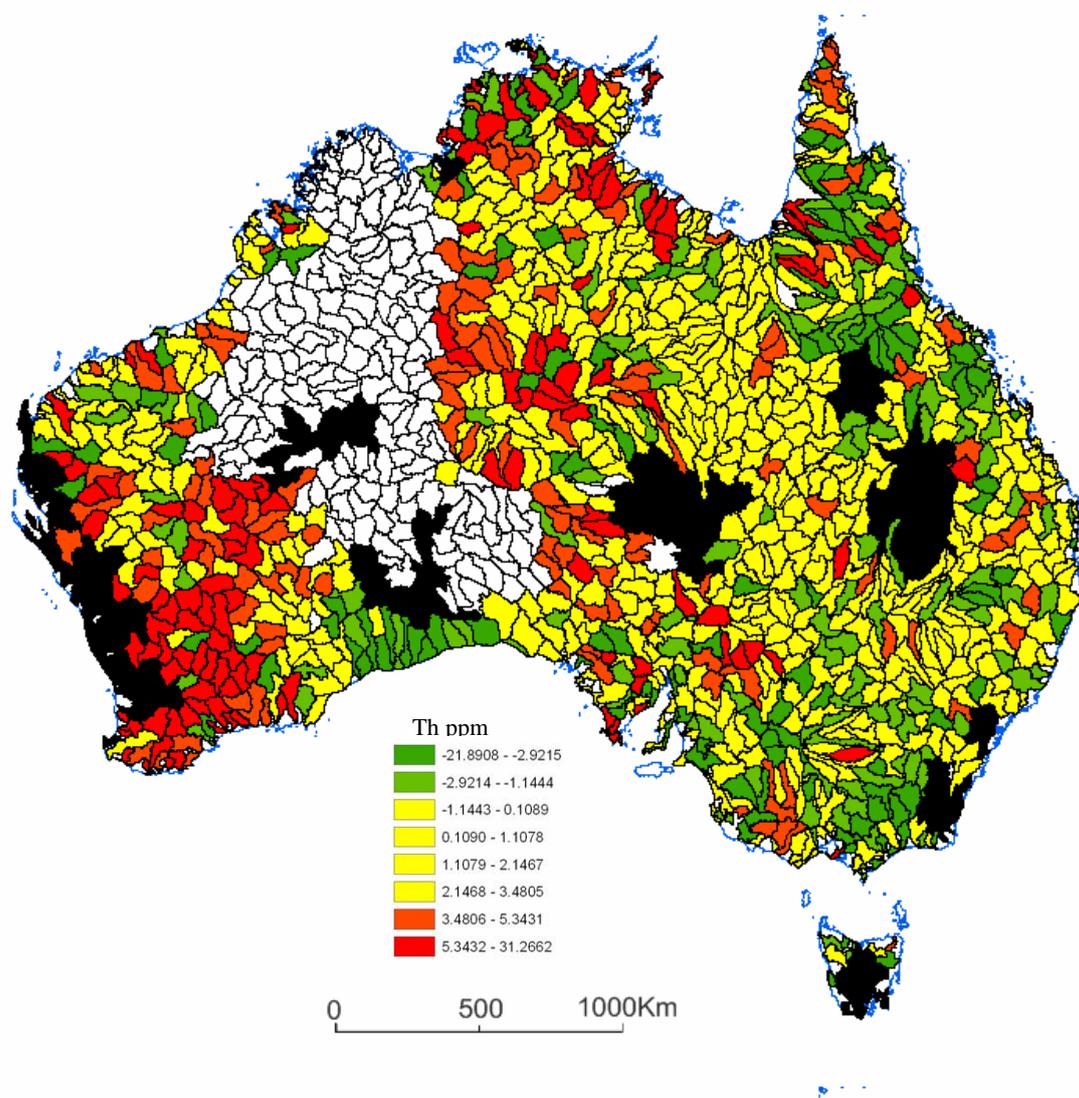


Figure 13. Difference between catchment airborne eTh and Th soil geochemistry. Greens and reds indicate geochemical values are considerably higher or lower than the catchment radiometric means, respectively. Yellows indicate broadly similar values between the two measurements.

The figure below shows the difference between airborne catchment mean eU and geochemical value.

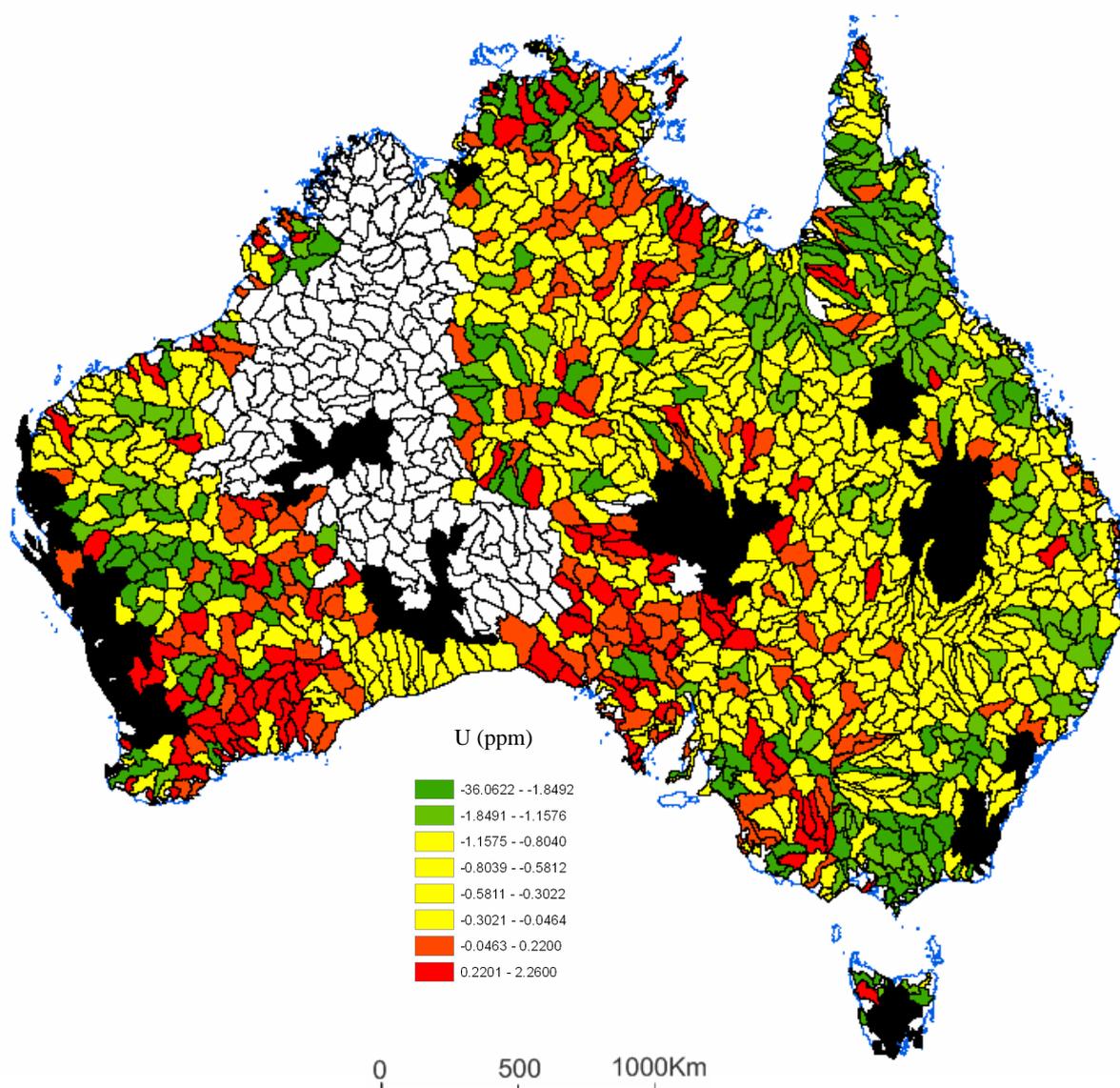


Figure 14. Difference between catchment airborne mean eU and U geochemistry. Greens and reds indicate geochemical values are considerably higher or lower than the catchment radiometric means, respectively. Yellows indicate broadly similar values between the two measurements.

The figure below shows the difference between airborne 300 m radius average eTh ppm values and geochemical value.

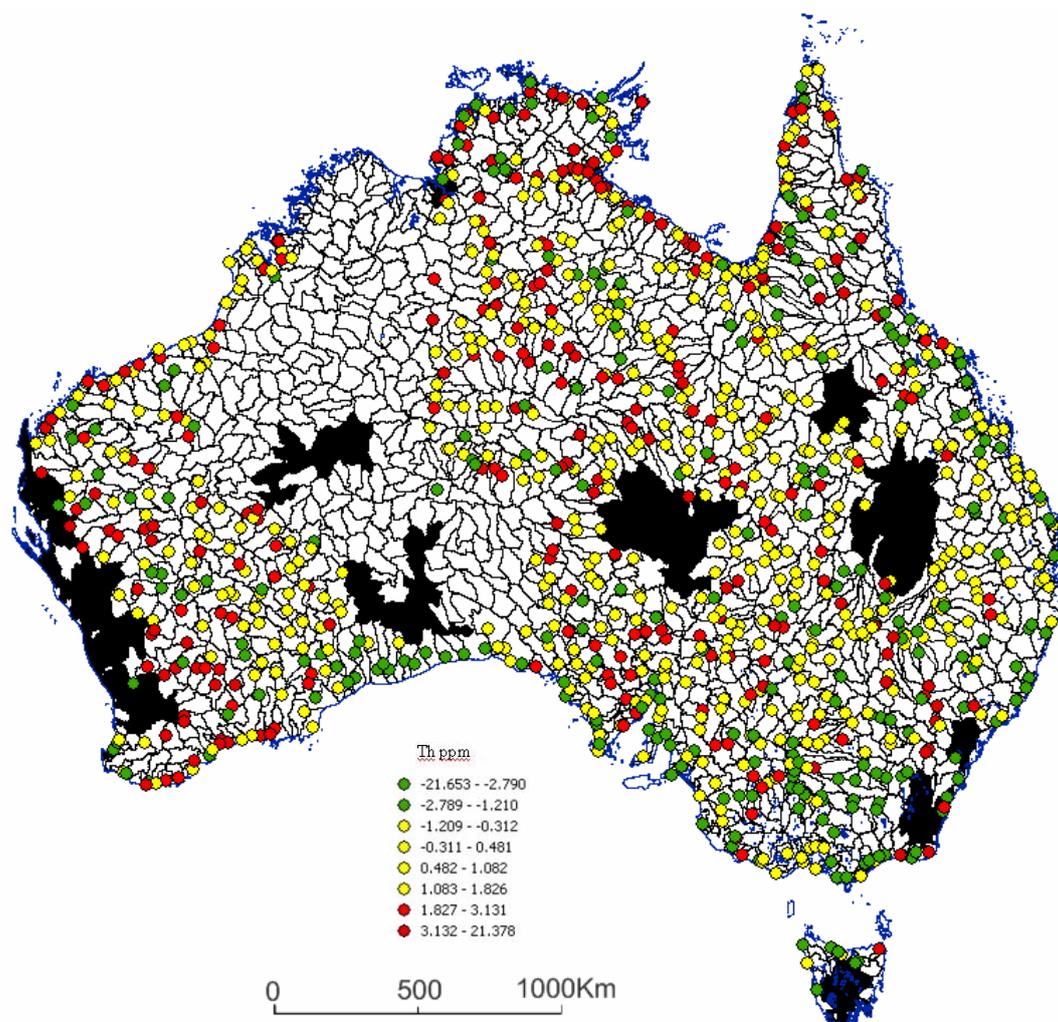


Figure 15. Difference between airborne eTh and Th soil geochemistry. Greens and reds indicate geochemical values are considerably higher or lower than the catchment radiometric means, respectively. Yellows indicate broadly similar values between the two measurements.

Catchment variability analysis

Figures 16 to 18 highlight the relative degree of radioelement variation within each catchment for K, eTh and eU. The analysis is sensitive to one decimal point for each of the elements (e.g., 1.2 wt% K or 2.3 ppm Th) and records the total number of unique airborne radiometric pixel values of K, eTh and eU for each catchment. These values are then normalised for catchment size to remove any bias in comparing small and large catchments. The catchments are scaled from low variability (green) to high variability (red). The patterns shown by the variability maps (particularly K) are broadly similar to the catchment relief map shown in Figure 19. Areas of relative high variability tend to correspond to outcrop areas and higher relief.

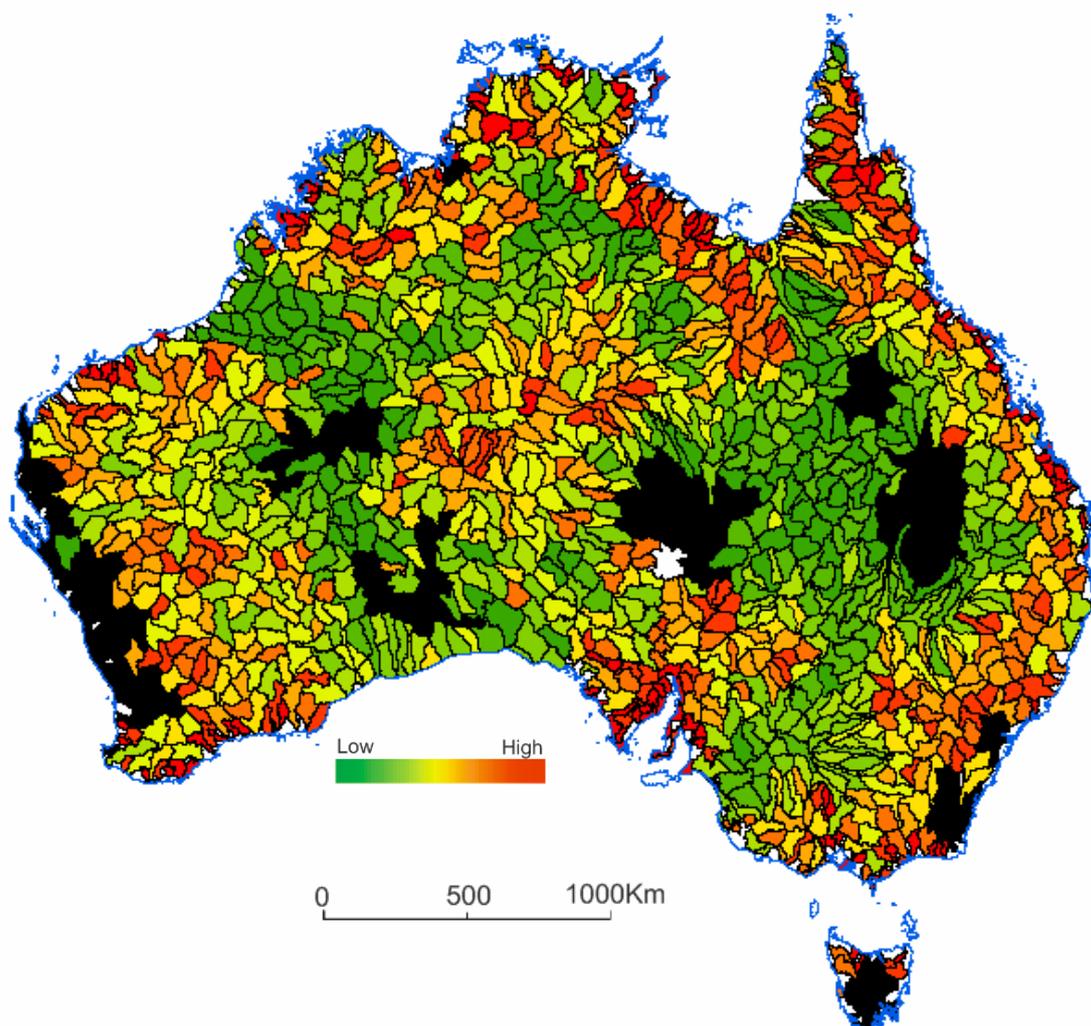


Figure 16. Variability of radiometric K values within each catchment (measures the number of unique pixel values in each catchment). Green = low variability; red = high variability.

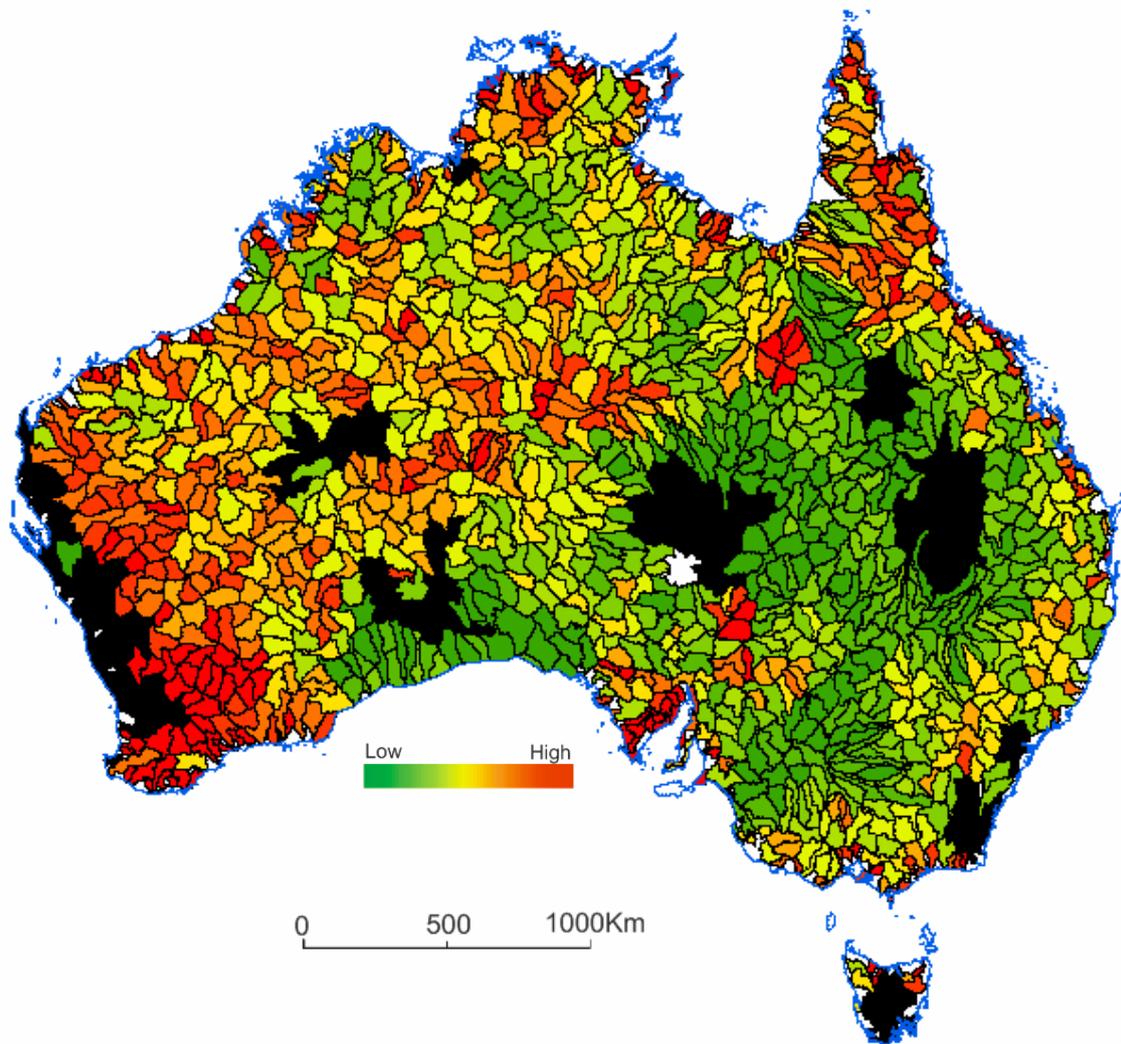


Figure 17. Variability of radiometric eTh values within each catchment (measures the number of unique pixel values in each catchment). Green = low variability; red = high variability.

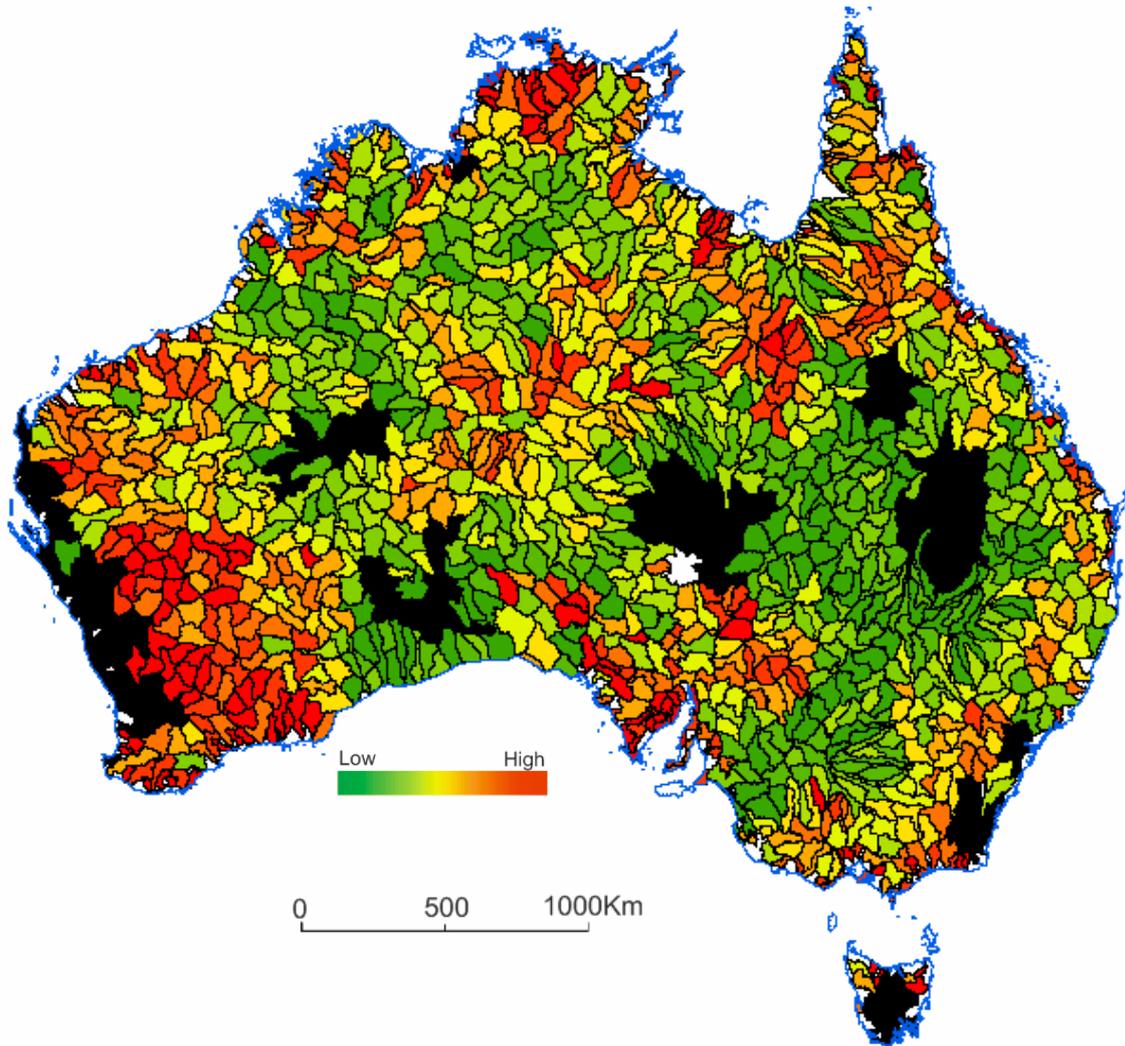


Figure 18. Variability of radiometric eU values within each catchment (measures the number of unique pixel values in each catchment). Green = low variability; red = high variability.

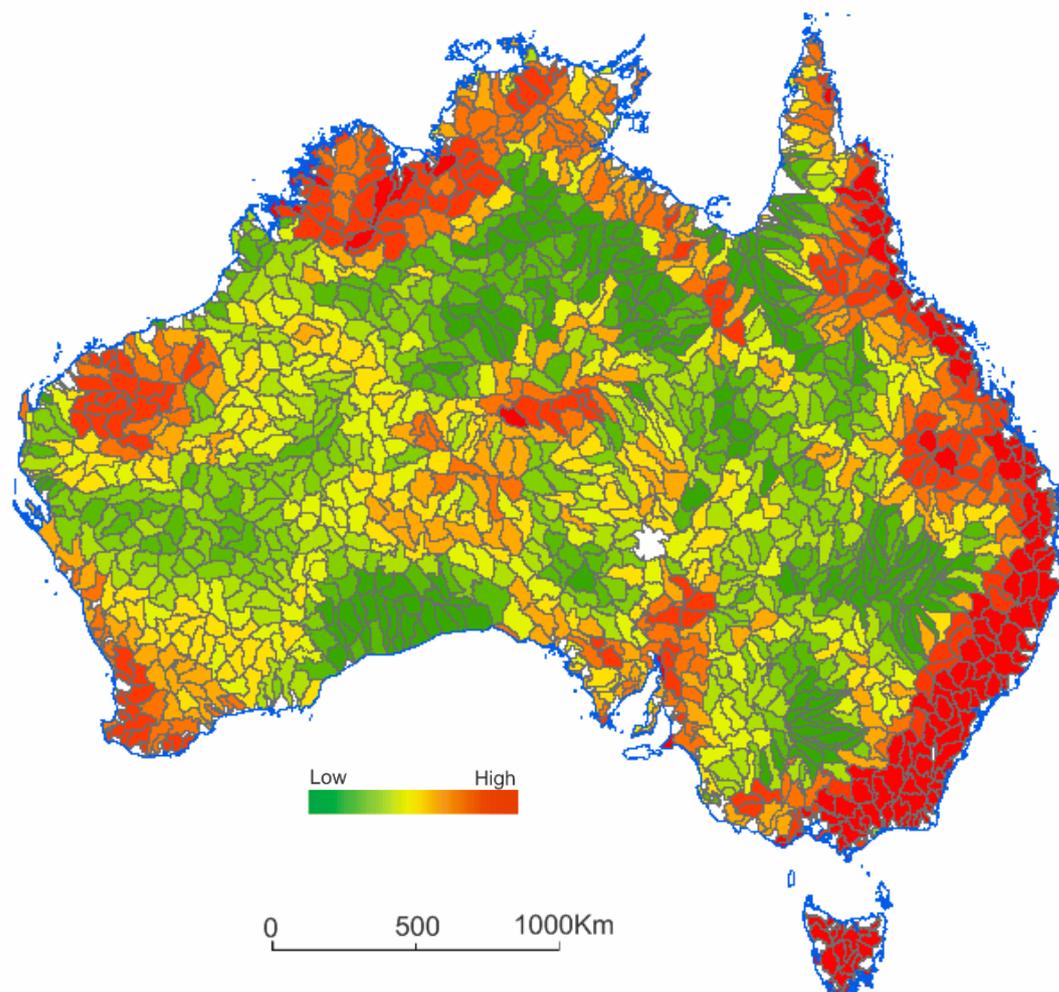


Figure 19. Mean catchment relief based on the 90 m digital elevation model (Shuttle Radar Topography Mission (SRTM) elevation data).

DISCUSSION

Potassium

Potassium is the most significantly correlated radioelement between the geochemical and airborne measurements. Correlations with TOS samples improved slightly from the single point measurements to the 300 m and 600 m radius averages ($r^2 = 0.67, 0.69, 0.69$, respectively). Removal of samples with a relatively high proportion of coarse material (i.e., where more than 25 % by weight of the sample is represented by material with a grain size exceeding 2 mm in diameter) improved the correlation. The highest correlation ($r^2 = 0.732$) corresponded to the 300 m radius average with coarse samples removed. Overall there is a slight to moderate under-estimation of airborne measured K compared to the ground measurements (Table 1).

Thorium

As with K, Th showed a significant correlation between geochemical data and airborne inferred eTh distribution. The single pixel and radius average comparisons gave a correlations between $r^2 = 0.54 - 0.55$. These correlations improved to $r^2 \approx 0.58$ with the removal of samples with a high proportion of coarse material (i.e. material which was not analysed by XRF). Unlike K and U, airborne Th is slightly over estimated compared with soil geochemistry.

Uranium

Uranium is the least abundant of the three radioelements in the Earth's continental crust with an upper continental crust concentration estimated to be in the range of 1-4 ppm (Wedepohl, 1969; Reimann & Caritat, 1998; Rudnick & Gao, 2003). As a consequence, the signal-to-noise ratio is lower for U compared with K and Th. This partly explains the weaker correlations observed between geochemical and airborne values. The other factors to consider are disequilibrium processes that can lead to errors in estimating eU. The highest correlation ($r^2 = 0.33$) was observed with the 300 m radius average with coarse material removed (Table 1). This correlation is still significant at the $p < 0.01$ level given the population size of $N \approx 900$. The correlations indicate a significant under-estimation of airborne predicted eU values. Other studies (Taylor *et al.*, 2002; Wilford, 2009) have shown much higher correlations between surface soil geochemistry and airborne U values. These investigations, however, were based on higher resolution airborne gamma-ray survey datasets (≤ 100 m flightline spacing) and smaller survey areas. In contrast, the national radiometric map is compiled mostly from medium resolution surveys (typically ~ 400 m flightline spacing).

Table 1. Sample and data treatments with corresponding coefficients of determination (r^2). All coefficients of determination shown are significant at the $p < 0.01$ level (highest in bold).

Sample and data type	K	Th	U	N
TOS vs pixel	0.676	0.551	0.137	1046
TOS vs 300 m radius	0.690	0.552	0.154	1046
TOS vs 600 m radius	0.687	0.539	0.018	1046
TOS vs point (coarse removed)	0.720	0.574	0.241	933
TOS vs 300 m radius (coarse removed)	0.732	0.577	0.332	933
TOS vs 600 m radius (coarse removed)	0.727	0.560	0.281	933

Calibrating airborne grids using surface geochemistry

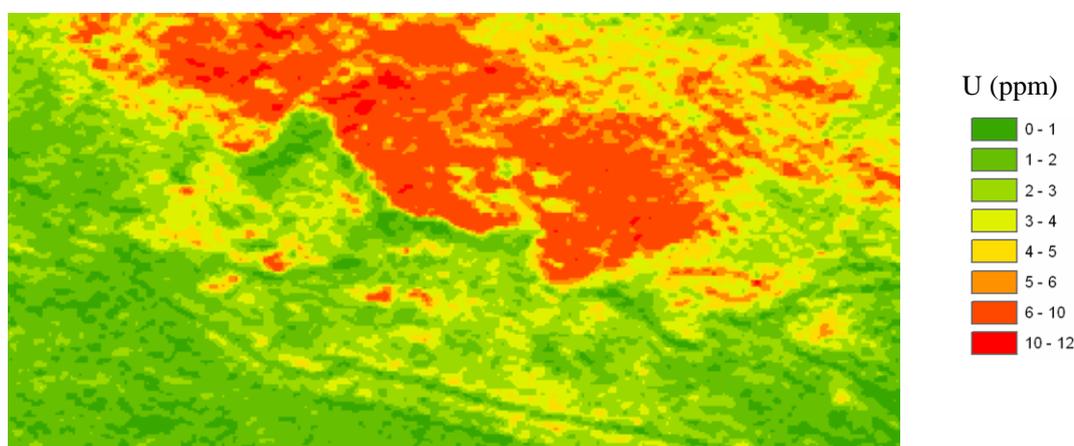
The relationships between site geochemical values and airborne radiometric values do not form a one-to-one correlation. A linear shift and offset based on the least square regression equations for K, eTh and eU can be used to calibrate the airborne grids so that they more accurately reflect surface geochemistry. Correction equations proposed from the present comparison of radiometric data with NGS outlet sediment geochemistry for K, Th and U are shown in Table 2. The U channel data is

the most significantly changed of the three radioelements and addresses the observed underestimation of airborne values compared to measured geochemical concentrations. Changes in the concentration of eU before and after the correction is applied are illustrated in Figure 20. These correction equations should be considered when investigating quantitative relationships between the distribution of these of elements in bedrock and regolith materials at the national scale. Corrections to eU distribution are also likely to be locally significant in U mineral system studies and prospectivity analysis.

Table 2. Proposed radiometric correction equations for K, eTh and eU.

Element	Linear regression corrections (forced through zero)
K	$1.33 \times \text{radiometric K}$
Th	$0.92 \times \text{radiometric eTh}$
U	$1.46 \times \text{radiometric eU}$

a)



b)

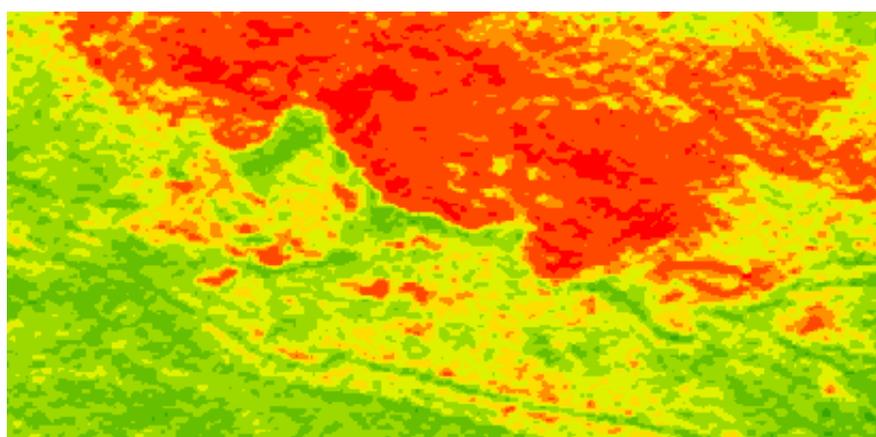


Figure 20. Original airborne eU (ppm) image (a) compared to corrected eU (ppm) image using the correction equation based on NGS data (b).

The application of these equations to the national radiometric map data assumes that the correction factors are uniform across the country. However we do see systematic, spatially-correlated differences between soil-measured and airborne-measured K, Th and U. For example Figure 13 shows differences between mean catchment eTh values and soil Th. Instead of seeing a random distribution of values, we see clusters of catchments with similar values. This clustering is also apparent when comparing sites (i.e., not catchment means) values (Figure 14). These differences can not be explained by disequilibrium because disequilibrium only affects U decay series. Assuming that the soil geochemical measurements are accurate then the patterns might be reflecting differences in the vertical distribution of K, Th and U in the soil/regolith profile or possibly shielding of gamma-

rays by vegetation. Another factor to consider is moisture content of the soil at the time of the survey. Increasing soil moisture decreases gamma emission with correspondingly lower estimations of K, Th and U. More research is needed to better explain these regional differences.

Regional differences in ground and airborne measured geochemistry may also have important implications in previous assumptions and calibration procedures in compiling the Radiometric Map of Australia. The Radiometric Map of Australia was calibrated to a standard global radioelement datum using the Australia Wide Airborne Geophysical Survey (AWAGS) (Minty *et al.*, 2009a) consistent with international reference standards of the International Atomic Energy Agency (IAEA, 2003). This process involved back-calibration of the AWAGS survey to the IAEA global radioelement datum using a portable gamma-ray spectrometer. The sensitivities of portable gamma-ray spectrometers are established by measuring the response of the instrument over concrete calibration pads doped with known concentrations of the radioelements. Ground measurements using the calibrated spectrometry were used to adjust the AWAGS data, which in turn was used to level the many individual gamma-ray surveys that together make up the national Radiometric Map of Australia. However many of the back-calibrations field sites were located in the Riverina area of south central New South Wales, which corresponds to an area identified in this study as having relatively low airborne values compared to soil measured concentrations (Figure 15). The most likely reason for the low eU values in this region is a loss of radon from the top layers of soil (see discussion on U disequilibrium). Consider the following scenario: let us assume that the portable spectrometer used for back-calibrating the AWAGS survey was calibrated over pads that exhibit no loss of radon. Subsequent measurements over a soil with 30% emanation radon, and where most of the emanation radon in the top-most 35 cm has been lost to the atmosphere, will result in an under-estimate of U concentration by almost 30%. So, depending on the emanation power and the permeability of the source, accurate estimation of uranium concentrations using *in situ* gamma-ray spectrometry may not be possible. This is the likely explanation for the overall under-estimation of airborne eU compared to the surface soil measurements. Back-calibration using carefully selected soil samples with total concentration of K, Th and U determined geochemically, as done here, is likely to be a more accurate approach than using one geophysical method to correct another.

U Disequilibrium

U disequilibrium occurs where one or more of the parent isotopes in the U decay series is preferentially added or removed. For example, release of ^{222}Rn (radon) gas through the soil or joints in bedrock and into the atmosphere can have a marked effect on the activity of ^{214}Bi , the element used to infer eU abundance. Early morning still air conditions can lead to a build up of ^{222}Rn above the soil in low-lying areas of the landscape. These conditions can result in an over-estimation of eU. To avoid this problem, most surveys commence flying later in the morning when surface heating and convection mix and dilute the ^{222}Rn gas within the air column. Rain can also deposit radioactive daughter products of atmospheric radon onto the ground, resulting in a significant increase in apparent U concentration. Fluctuations of the soluble element Ra (radium) can also lead to over- or under-estimations of U. ^{226}Ra is soluble and can be leached or concentrated through weathering and groundwater processes. Large U anomalies have been linked with ^{226}Ra exsolved from groundwater (Giblin & Dickson, 1984; Dickson, 1985). Within the soil, Ra can be adsorbed onto the surfaces of clays and precipitated with Fe-oxyhydroxides. U is also mobile and can be leached from soluble minerals under oxidising conditions and precipitate when conditions become more reducing. Therefore, U can be leached relative to Ra, which would lead to over-estimation of airborne value compared to the soil-measured value. All these factors may partly explain the relatively high degree of scatter between the soil and airborne U values in the scatter-plots. Some of the outliers in the scatter plots between soil and airborne eU are associated with palaeochannel and lake systems or areas of high rainfall (northern tropics) where precipitation and leaching processes are potentially most active (Figure 21). However, overall airborne measured eU is significantly lower than measured soil U. The most likely reason for the low eU estimates in these measurements is because of a loss of radon from the top layers of soil. It is well known that the mobility of radon is a serious

problem for the accurate, *in situ* estimation of the U concentrations of rocks and soils using gamma-ray spectrometry (Minty, 1997; Grasty, 1997). Minty and Wilford (2004) identified significant differences in ground measured eU due to changes in near-surface radon concentrations. Their study showed that radon concentration reaches a maximum at about one hour after sunrise, before slowly dispersing over a period of 2-3 hours.

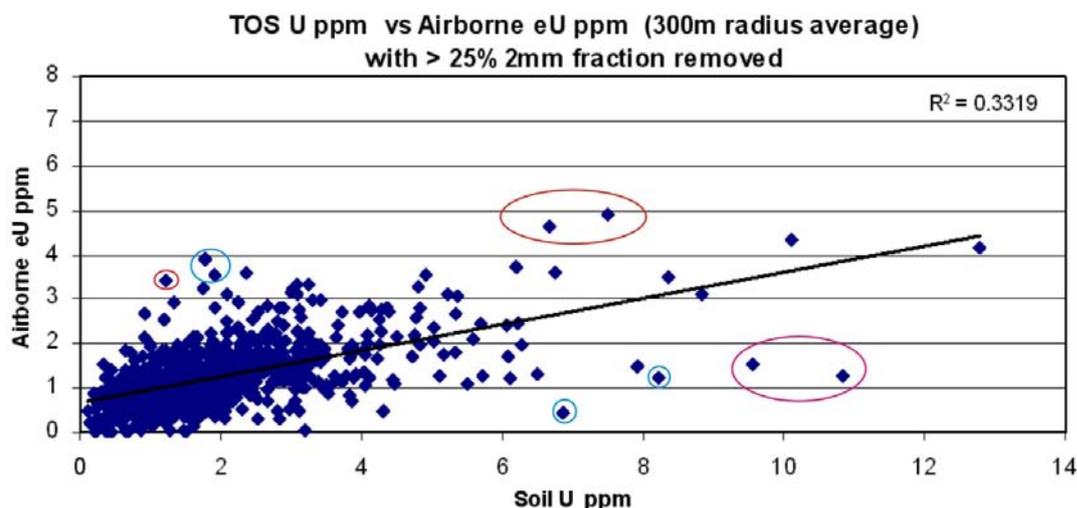


Figure 21. Scatter plot and coefficient of determination (r^2) between TOS (<2 mm) and gamma-ray spectrometry for eU values based on 300 m radius averages, after the removal of samples that had more than 25% of the total sample weight above 2 mm size fraction. Some of the significant outliers occur in palaeo-channel and lake systems (red circles) and in areas of high rainfall in the northern tropics of Australia (blue circles).

Inferring U using Th as a proxy

The abundance of U and Th increases during the fractionation of igneous rocks so that felsic igneous and volcanic rocks, for example, have higher U averages than mafic and ultramafic rocks. Th has an average crustal abundance approximately 4 times that of U (e.g., Rudnick & Gao, 2003) and is generally less mobile than U. Weathering and alteration associated with hydrothermal systems can preferentially concentrate U compared to Th. Th is therefore more stable within the weathering environment than U, and based on the relative proportion of these elements within the crust we can infer a possible original U content (U_{mod} ; D. Champion and S. van der Wielen, pers. comm., 2011) by dividing the Th concentration by 4. Dividing airborne eU by modelled U ($Th/4$) and plotting this ratio against the soil U values highlights potential sites where disequilibrium might be causing either over- or under-estimations of U (Figures 22 and 23).

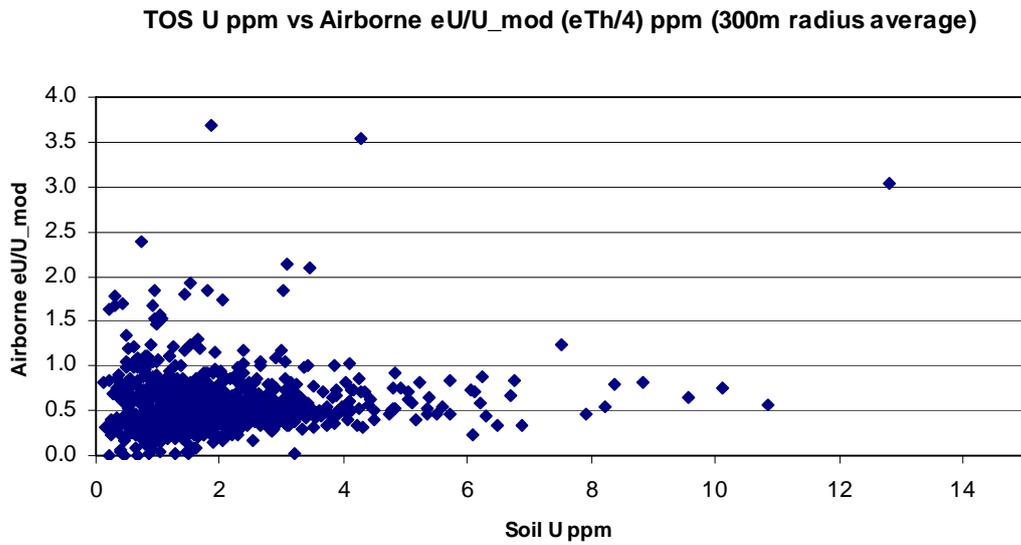


Figure 22. Scatter plot of the ratio between airborne eU and modelled (U_{mod}), based on dividing Th concentrations by 4, on the Y-axis against soil U values on the X-axis. Correlations are based on TOS samples with removal of sites having >25% of the total sample weight above 2 mm size fraction. Departure from the ratio value of 1 indicates potential sites of U disequilibrium. Most of the samples fall below 1 indicating an under-estimation of eU to that predicted by the concentration of Th.

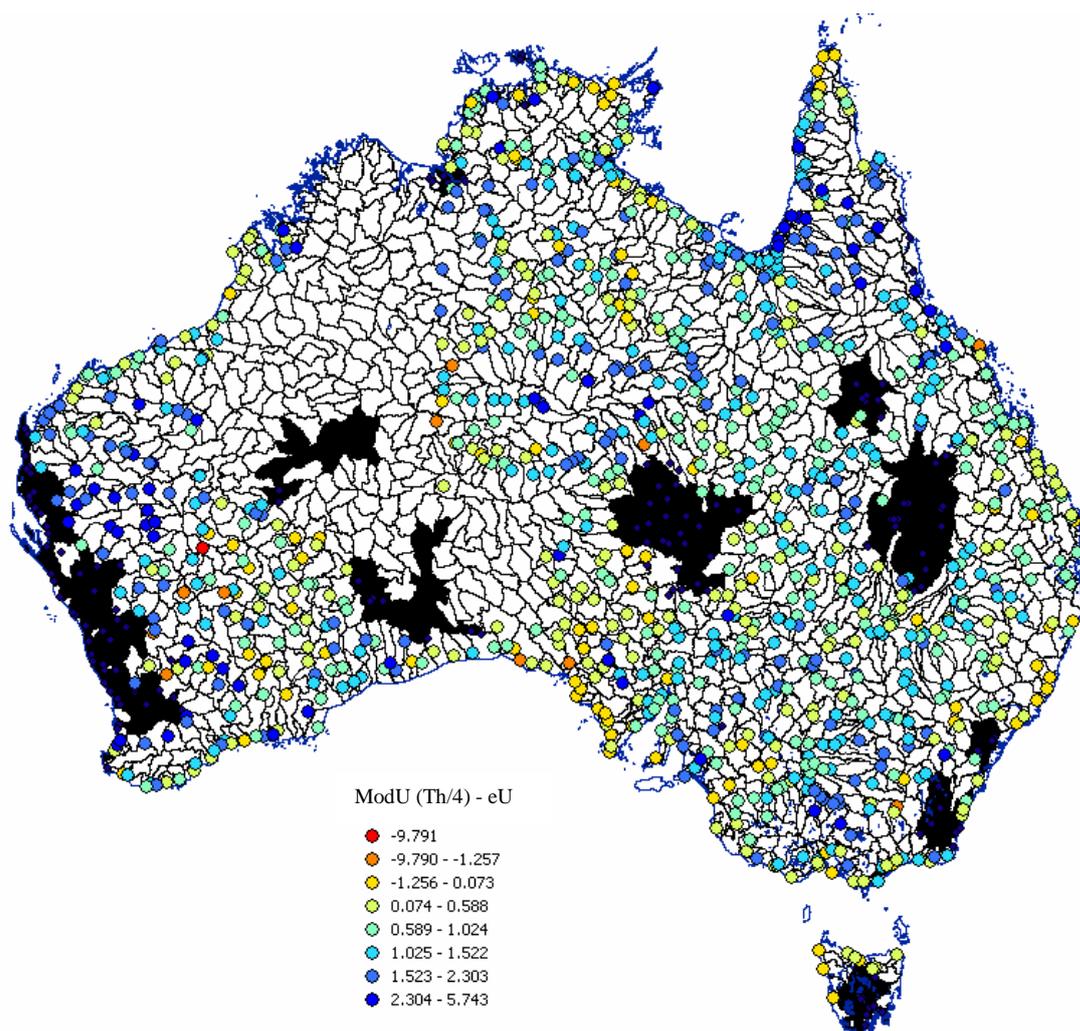


Figure 23 Map highlighting the difference between modelled U (Th/4) for each soil site and airborne measured eU. Positive numbers (cool colours) indicate higher modelled values compared to airborne measured eU (therefore areas of potential under estimation of airborne U). Negative numbers (warm colours) indicate lower modelled values compared to airborne prediction eU.

Relationships between site geochemistry and catchments

Sampling channel and overbank sediments in geochemical surveys is widely practised because of the assumption that these materials broadly reflect the mixture of materials from within their catchments. The geochemical ‘footprint’ of a sediment sample will invariably be much greater than samples from locally derived residual (*in situ*) materials. Maps showing the difference between site values and catchment airborne averages for K, eTh and eU (Figures 12, 13 and 15) provide a framework for better understanding the degree to which the sampling site value is representative of its catchment. The analysis indicates that 50% of the mean catchment values are within the range of -0.46 – 0.07 wt% for K, -1.14 – 3.48 ppm for Th and -1.16 – -0.05 ppm for U. Catchments more significantly above or below the measured value (lower and upper quartiles of the distributions) are flagged by this approach. Values significantly above or below the catchment means can relate to variations in the relative rates of depositional and weathering processes (Figure 24). Active areas of deposition will tend to reflect the geochemistry of the bedrock from which the sediments have been derived. Conversely, the geochemical composition of sediments in less active depositional settings is likely to reflect modifications due to weathering processes and pedogenesis (e.g., leaching of soluble elements and retention of more stable elements). The degree to which these processes are occurring and the area they cover in the catchment will influence the degree to which the sample points from the NGSa project are representative of the catchment (Figure 24).

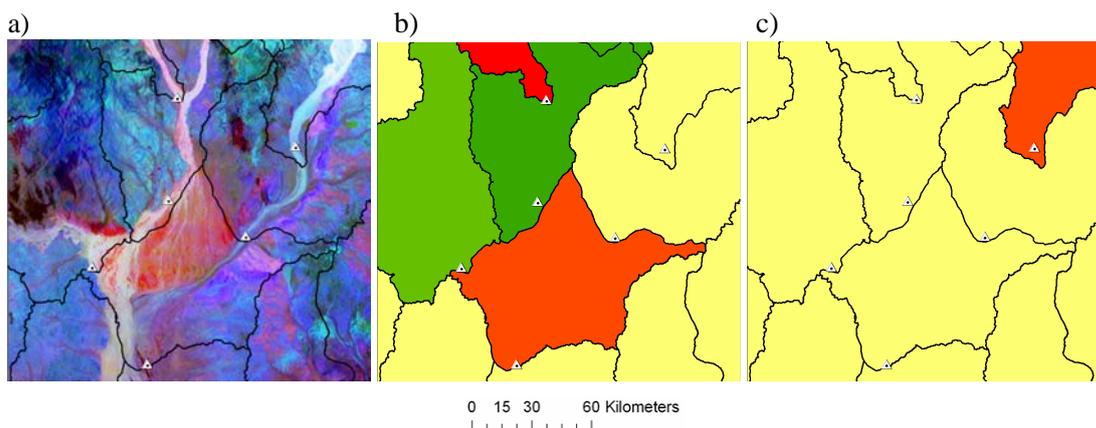


Figure 24. An example of the relationships between geomorphic activity and weathering on sample geochemistry and the degree to which the site samples are representative of their catchments from central west Queensland. On the ternary radiometric image (a) red and white hues relate to active areas of erosion and deposition, green and blue hues correspond to less active depositional and erosional parts of the landscape where weathering has modified the surface geochemistry. On the corresponding map showing the relative difference between site-measured K and airborne K (b), red, green and yellow colours relate to site values that are lower, higher or similar to the catchment means. Sediment outlet samples for the green and orange catchments occur in floodplain sediments that are not representative of the total catchment. On the corresponding Th map (c), differences in Th are not evident compared with K, which is likely to reflect higher mobility of K compared to Th during weathering (e.g., Th is not changing significantly despite differences in weathering between the catchments).

Variations in K, eTh and eU values within each catchment will reflect the composition and distribution of bedrock and regolith materials (Figures 16, 17, 18). K variability increases in areas of high overall relief suggesting that changes in K concentration are largely reflecting variations in bedrock geochemistry. Conversely, in lower relief areas variations in K are also lower. Areas of lower relief are likely to be more weathered; under these conditions leaching of K would reduce its concentration and range, resulting in lower overall variability. In comparison, eTh and eU are less well correlated with relief and show a relatively high degree of variability over a range of relief classes (Figures 19). Th and U are commonly concentrated in weathered materials where they are associated with Fe-oxyhydroxides and clays in the regolith. Changes in eTh and eU concentrations therefore appear to reflect variations in weathering intensity and regolith composition as well as bedrock geochemistry. The combination of the catchment mean differencing and the variability classification approaches provide a framework to better assess the degree to which the site values are representative of the surrounding catchment.

CONCLUSIONS

Airborne concentrations of K and Th show a high degree of correlation to ground-based measurements. However, U concentrations derived from the radiometric data are significantly underestimated compared with ground geochemistry. Correction factors can now be applied to the airborne data to more accurately reflect surface geochemistry. These correction equations should be considered when investigating quantitative relationships between the distribution of these elements in bedrock and regolith materials at the national scale. Corrections to eU distribution are also likely to be locally significant in U mineral system studies and prospectivity analysis. However, systematic and spatially-correlated differences between soil-measured and airborne values across the Australian continent suggest that several local correction factors may be a more effective than one single equation for smaller scale investigations. It also highlights the limitations of the local area back-calibration procedure for correcting gamma-ray survey datasets of this size. In this context the correction factors developed in this paper should be seen as a general adjustment to match airborne values with surface soil geochemistry for very large (up to continental-scale) areas. For more local scale investigations local correction factors should be developed and applied to the airborne data.

The degree to which the ground geochemical value is representative of its catchment has been assessed by showing the difference between the site value and the airborne catchment mean, as well as classifying catchments based on the number of unique K, Th and U values normalised by the catchment size. Results from this investigation are preliminary and further work is required to better understand the relationships between ground and airborne values, in particular the effects of U-disequilibrium in estimating U. More detail analysis is also required to better understand the influence of parent materials, geomorphic and weathering processes over different spatial and temporal scales on the distribution of these elements within the catchment and more generally with the landscape.

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