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Assessment of naturally occurring radionuclides in England and Wales

Science Report: SC030283/SR

SCHO0906BLBK-E-P

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This report is the result of research commissioned and funded by the Environment Agency's Science Programme.

Published by:
Environment Agency, Rio House, Waterside Drive, Aztec West, Almondsbury, Bristol, BS32 4UD
Tel: 01454 624400 Fax: 01454 624409
www.environment-agency.gov.uk

ISBN: 1844325679

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Dissemination Status:
Publicly available

Keywords:
naturally occurring radionuclides; uranium isotopes; thorium isotopes; potassium-40; radium-226; polonium-210; lead-210; non-human species;

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Science Project Number:
SC030283

Product Code:
SCHO0906BLBK-E-P

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Steve Killeen Head of Science

Executive Summary

The Environment Agency is developing methods to determine the risk to plants and animals from ionising radiation in the environment. An interim method to assess the impact of ionising radiation on wildlife is now being used by the Environment Agency to assess Natura 2000 sites across England and Wales. The contribution that naturally occurring radionuclides make to the radiation dose received by non-human species in England and Wales needs to be determined, to complement the impact assessment methods used by regulators and industry.

The overall aim of this work was to determine the activity concentration ranges of naturally occurring radionuclides in non-human species and to estimate the background radiation dose rates in terrestrial and freshwater ecosystems across England and Wales. The specific aims were to:

- review and collate data from existing literature to establish the activity concentrations of naturally occurring radionuclides from the ^{238}U and ^{232}Th series and ^{40}K in environmental samples including soils, sediments, waters, and particularly non-human species (wild animals and plants) for terrestrial (including coastline) and freshwater ecosystems.
- identify gaps in the collated data, particularly for non-human species, and to design and carry out a sampling and analytical campaign to partially fill these data gaps.

Activity concentrations of naturally occurring radionuclides in non-human species in the United Kingdom were compiled from the literature and existing in-house datasets. Particular emphasis was placed on finding data suitable for the International Commission for Radiological Protection's (ICRP) proposed reference animals and plants (RAPs) to enable the results of this work to feed into the further development of RAPs by the ICRP. The results of the review were used to design a sampling strategy to collect additional data.

The novel and compiled data were combined into a database available in Microsoft Excel and ArcView data files. The most abundant data are for ^{40}K followed by ^{232}Th, ^{234}U and ^{238}U respectively; there are relatively few data for ^{210}Pb, ^{210}Po, ^{226}Ra and ^{230}Th activity concentrations in some RAPs. Whilst not the focus of this work, the database also contains some data for natural radionuclides in marine organisms in UK waters.

Estimates of activity concentrations of ^{40}K, ^{238}U and ^{232}Th series radionuclides in environmental media (soil, stream sediments and stream waters) were derived from total K, U and Th concentrations derived mainly from the ongoing geochemical survey of the United Kingdom (G-BASE), conducted by the British Geological Survey. The geochemical survey data are currently incomplete for England and Wales, but almost complete coverage was obtained for K in stream sediments by using the Wolfson Atlas data for southern England.

For U and Th in sediments and K, U and Th in soils, more complete coverage was achieved by geological extrapolation (using relationships between soils/sediments and bedrock/superficial geology). For soils and sediments, datasets are provided for both: (i) geometric mean concentrations from measured samples on a 5 x 5 km square basis where data are available; and (ii) extrapolated surfaces covering all of England and Wales. It is recommended that where data are available for an area being assessed, these should be used rather than the extrapolated values. For waters, only geometric mean concentrations are provided, where data are available. The relationship between radioelements in waters and geology was not considered sufficiently strong to justify extrapolation in this case.

Unweighted absorbed internal doses were estimated from the collated biota whole-body activity concentration data. For all organisms, unweighted absorbed doses from ^{40}K predominated (of the order of 10^{-2} µGy h^{-1}). Absorbed doses from most other radionuclides for which there were data rarely exceeded 10 per cent of the ^{40}K value. Not enough data were available to estimate internal absorbed doses due to ^{228}Th, ^{234}Th and ^{228}Ra for any RAPs, nor ^{210}Pb, ^{210}Po or ^{226}Ra for some RAPs.

External absorbed dose rates were estimated for the RAP geometries using the derived media concentrations. Whilst ^{40}K was the single largest contributor (10^{-2} µGy h^{-1}) to the total dose rates estimated for terrestrial RAP geometries, dose rates estimated for both ^{228}Th and ^{228}Ra were generally within approximately a factor of two of the ^{40}K contribution. Absorbed dose rates for all other radionuclides were at least two orders of magnitude lower than those estimated for ^{40}K. For the rat, exposure whilst underground (assumed to be 50 per cent of the time) dominated the total external absorbed dose rates. External dose rates due to radionuclides below ^{234}U in the ^{238}U series were not estimated.

The datasets of naturally occurring radionuclide concentrations in biota and the environment will help to significantly improve assessments of the background exposure of wild animals and plants within England and Wales. The media concentration datasets, together with concentration ratios, could be used to estimate some of these missing biota activity concentration values. More complete media concentration datasets derived from actual data, rather than geological extrapolation, could be generated once the G-BASE coverage of England and Wales is complete (anticipated to be by 2021).

Contents

1 Introduction

The Environment Agency is developing methods to determine the risk to plants and animals from ionising radiation in the environment, through their past involvement in the EURATOM FP5 project FASSET (Framework for assessment of environmental impact; Larsson *et al.,* 2004) and now as a partner in the FP6 ERICA project (http://www.erica-project.org). The aim of ERICA (Environmental risk from ionising contaminants: assessment and management) is to provide an integrated approach to scientific, managerial and social issues concerned with the environmental effects of contaminants which emit ionising radiation, with emphasis on non-human species and ecosystems.

The Environment Agency has also funded work to develop an interim method for assessing the impact of ionising radiation on wildlife (for example, Copplestone *et al.,* 2001) and is now carrying out assessments of Natura 2000 sites across England and Wales (Copplestone *et al.,* 2005). The contribution that naturally occurring radionuclides make to the radiation dose received by non-human species in England and Wales needs to be determined, to complement the impact assessment methods used by industry and regulators. Pentreath (1999, 2002) suggests that the potential impacts of artificially released radionuclides should be compared to that of the natural background radiation dose rate normally experienced by animals and plants, and this concept is being considered further by the International Commission on Radiation Protection (ICRP, 2005).

1.1 Objectives

The overall aim of this work was to determine the activity concentration ranges of naturally occurring radionuclides in animal and plant species and to estimate the background radiation dose rates in terrestrial and freshwater ecosystems across England and Wales. The specific aims were to:

- review and collate data from existing literature to establish the activity concentrations of naturally occurring radionuclides from the uranium and thorium series (see Table 1.1) and ^{40}K (α, β, γ-emitters) in environmental samples including soils, sediments, waters, and particularly non-human species (wild animals and plants) for terrestrial (including the coastline) and freshwater ecosystems.
- identify gaps in the collated data, particularly for non-human species, and to design and carry out a sampling and analytical campaign to partially fill these data gaps.

Table 1.1: Primary decay schemes of ^{238}U and ^{232}Th (adapted from Whicker and Schultz, 1982)

Uranium-238			Thorium-232		
Radionuclide	**Half-life**	**Radiation**	**Radionuclide**	**Half-life**	**Radiation**
^{238}U	$4.5x10^{9}$ y	α,γ	^{232}Th	$1.4x10^{10}$ y	α,γ
^{234}Th	24 d	β,γ	^{228}Ra	6.7 y	β,γ
^{234}Pa	1.2 min	β,γ	^{228}Ac	6.1 h	β,γ
^{234}U	$2.5x10^{5}$ y	α,γ	^{228}Th	1.9 y	α,γ
^{230}Th	$8.0x10^{4}$ y	α,γ	^{224}Ra	3.6 d	α,γ
^{226}Ra	$1.6x10^{3}$ y	α,γ	^{220}Rn	55 s	α,γ
^{222}Rn	3.8 d	α,γ	^{216}Po	0.16 s	α,β
$^{218}Po^{*}$	3.1 min	α,β	^{212}Pb	11 h	β,γ
^{214}Pb	27 min	β,γ	$^{212}Bi^{*}$	61 min	α,β,γ
$^{214}Bi^{*}$	20 min	α,β,γ	^{212}Po	$3.0x10^{-7}$ s	α
^{214}Po	$1.6x10^{-4}$ s	α	^{208}Pb	Stable	None
^{210}Pb	19 y	β,γ			
^{210}Bi	5.0 d	α,β,γ			
^{210}Po	138 d	α,γ			
^{206}Pb	Stable	None			

Notes: *Alternate, less frequent branching decays not shown

2 Biota

2.1 Proposed ICRP reference animal and plants

The ICRP has recently suggested a set of reference animals and plants (RAPs) as points of reference for assessing radiation effects in non-human species (ICRP, 2005). The proposed list of reference animals and plants comprises: deer; rat; duck; frog; trout; flatfish; bee; crab; earthworm; pine tree; wild grass; and brown seaweed. One aim of the work described in this report was to categorise available data for naturally occuring radionuclides in England and Wales on the basis of the reference animals and plants proposed by the ICRP.

Dimensions for RAPs have been proposed for phantom dose modelling, assuming solid ellipsoids (ICRP, 2005). Those for the adult forms of RAPs relevent to terrestrial and freshwater ecosystems are presented in Table 2.1; brown seaweed is included in the table as this review also considered the coastline.

In the preliminary stages of the work, it was recognised that much of the available data would not be specific to the proposed ICRP RAPs. Therefore, available data were attributed to appropriate RAPs as follows:

- Duck – any species of wild bird.
- Frog – any species of amphibian.
- Trout - any species of freshwater fish.
- Bee – any species of flying insect.
- Earthworm – any species of earthworm.
- Pine tree – any species of tree.
- Wild grass – any species of wild grass or herb, and any data recorded as ‘grass’.
- Brown seaweed – any species of seaweed.

Given that the ICRP recommends two mammalian RAPs, data were collated for ‘any wild mammalian species’ and separately for deer (data for any species of wild deer) or rat. The original species is listed in the database along with the ICRP RAPs that it represents.

Whilst this categorisation has been used to attribute data to RAPs, the availability of data specifically for the RAPs as defined by ICRP is also briefly discussed.

Table 2.1: Suggested dimensions for adult stage of proposed reference animals and plants (taken from ICRP, 2005)

Reference animal or plant (RAPs)	Major axis of ellipsoid (cm)	Minor axis of ellipsoid (cm)	Second minor axis of ellipsoid (cm)
Deer (adult)	130	60	60
Rat	20	6	5
Duck	30	10	8
Frog	8	3	2.5
Trout	50	8	6
Bee	2	0.75	0.75
Earthworm (elongated)	10	1	1
Pine tree (trunk)	3,500	150	150
Grass (spike)	5	1	1
Brown seaweed	50	0.5	0.5

2.2 Availability of data for terrestrial biota

2.2.1 Data review

Whilst the focus of this review was England and Wales, data for all of the United Kingdom were collated because of lack of data for England and Wales alone. Available sources of data for natural radionuclide concentrations in biota included: (i) 'grey' literature; (ii) refereed papers; and (iii) in-house databases.

The RIFE (*Radioactivity in Food and the Environment*) reports are published annually by UK agencies and report radionuclide activity concentrations throughout the United Kingdom (predominantly close to sites discharging radionuclides to the environment). These reports provide activity concentrations for some naturally occurring radionuclides in biota such as game and plant species. With the exception of grass, sampled biota are the edible components of species eaten by man. Data were compiled from RIFE reports presenting the results of sampling conducted between 1995 and 2004 (RIFE, 1996 - 2005). Sampling coordinates (as four figure OS grid references) for RIFE sampling sites were provided by the Environment Agency.

In addition to the RIFE reports, a number of other grey literature publications were reviewed and data from some of these used (a full list of grey literature reviewed is provided in Appendix A.1).

To review the refereed literature, 88 variations of keyword searches were performed using *Web of Knowledge*. The Centre for Ecology and Hydrology (CEH) radioecological bibliographical database (>15,000 references) was also searched. Whilst these searches identified a large number of references (>7,000 references) to screen, the number of potentially useful references was less than 50 (reference details and abstracts, where available, can be found in Appendix A.2) of which only five yielded usable data.

2.2.2 In-house data

The CEH has previously conducted many analyses of biota for anthropogenic radionuclides. These samples were obtained predominantly from studies on the

deposition and behaviour of radiocaesium following the 1986 Chernobyl accident, and assessments of the transfer of radioactivity discharged from the Sellafield reprocessing plant. Whilst previously not reported, gamma analysis of these samples had yielded results for a number of naturally occurring radionuclides relevant to this review. These data were collated from existing databases, or from the original analytical printouts, and contributed the majority of the available ^{40}K data.

As the determination of natural series radionuclides was not the primary aim of these studies, samples were not sealed and stored to ensure isotopic equilibrium prior to analyses. However, most ^{238}U and ^{232}Th series radionuclides were below detection limits, which are comparatively high when analysed by gamma spectroscopy.

2.2.3 Availability of data

Table 2.2 summarises the availability of data by RAPs and radionuclide. Figure 2.1 shows the location of available data where a georeference was available. The most abundant data were available for ^{40}K. Many of the data for mammals and birds were from Scotland. No data were available for species considered within the review for frog, bee or earthworm, nor specifically for the proposed reference rat. All of the data allocated to pine tree were predominantly for edible portions of deciduous trees. Overall, there were few data for the ^{232}Th and ^{238}U series radionuclides and little data originating from Wales.

Table 2.2: Numbers of samples for which ^{40}K, ^{232}Th series and ^{238}U series radionuclide activity concentrations in UK biota were compiled from the literature/in-house sources. Available total U data are also indicated. Note the numbers include measurements reported as below detection limits.

RAP	^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{228}Th	^{230}Th	^{232}Th	^{234}U	^{235}U	^{238}U	Total U
Duck	27										
Frog											
Bee											
Earthworm											
Pine tree	2	5	5	5		5	5	9	8	9	
Wild grass	218	117	147	123		4	151	160	157	160	163
Brown seaweed	13	92	35		10	10	23	19	15	58	
Trout	44										
Total mammal*	141	28	31			2	32	4	4	3	3
Deer	72	1									
Rat											

*Total mammal includes available deer and rat data in addition to that for other mammalian species.

The lack of available data to assess exposure of non-human species to the naturally occurring radionuclides identified here is in agreement with other recent assessments for aquatic and terrestrial biota conducted on a European scale (Brown *et al.,* 2004; Gómez-Ros *et al.,* 2004). In their review, Brown *et al.* (2004) state that no references on naturally occurring radionuclides in European freshwater biota were identified. No data for wild terrestrial species are quoted by Gómez-Ros *et al.* (2004) in an

assessment of the exposure of terrestrial organisms to naturally occurring radionuclides.

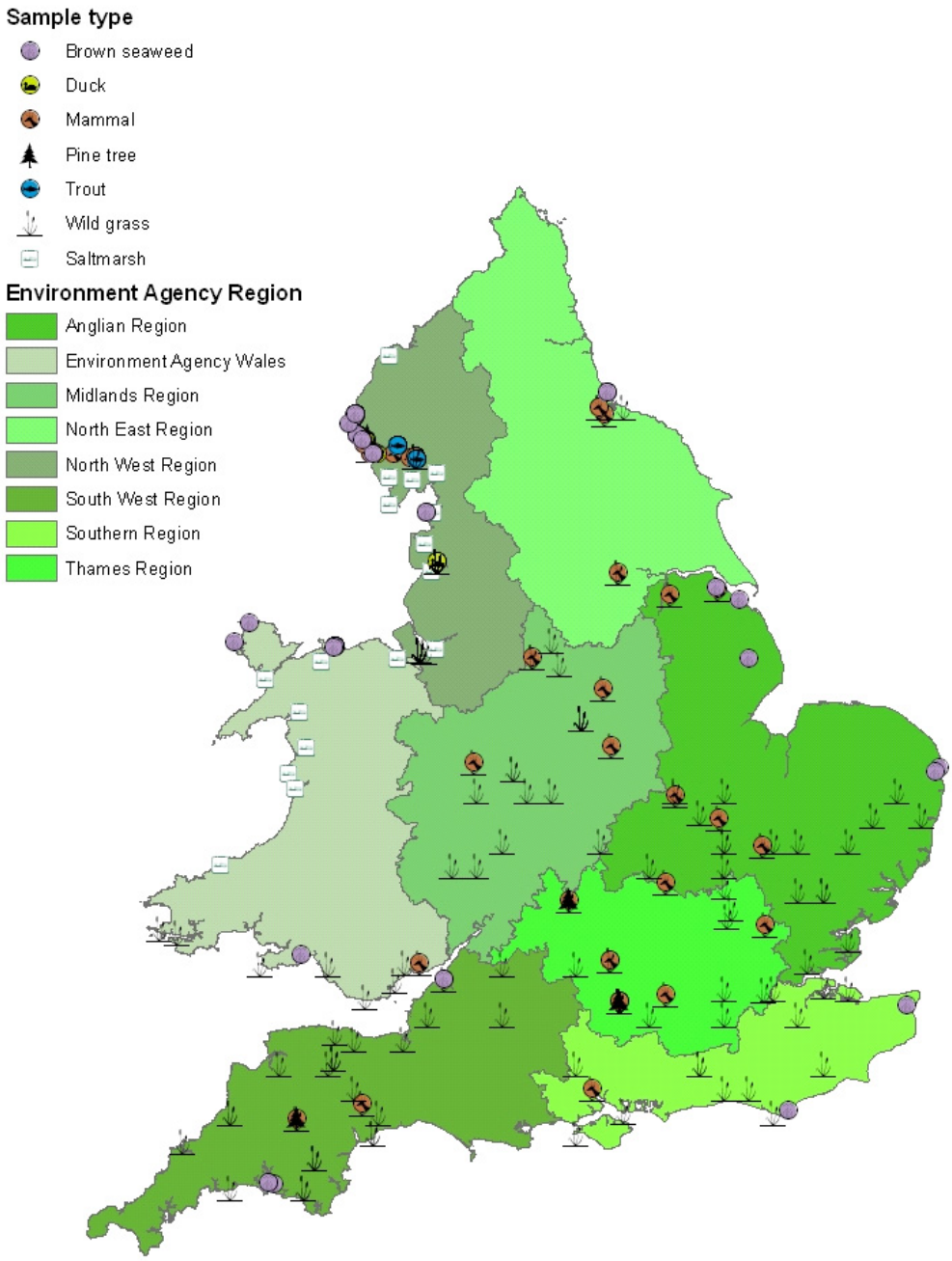

Figure 2.1: Sampling locations for natural radionuclide data identified from previous publications and in-house datasets. Saltmarsh sampling points are discussed in Section 3.5.

On the basis of the availability of data, it was recommended that sampling should concentrate on the following proposed RAPs: duck, bee, earthworm, pine tree and frog. Given the lack of data from Wales, it was proposed that some sampling should be targeted there. Reproductive organs were to be analysed separately wherever possible in recognition of the need to protect the reproductive capacity of non-human species.

2.3 Sampling

Due to time constraints, sampling had to be conducted over the period December 2005 to February 2006. The collection of some species representative of the proposed RAPs over this time period proved difficult; to overcome this, existing sample archives from the CEH were used in combination with the new samples.

Samples obtained comprised:

- a mixed sample of flying insects (predominantly moths) from each of the eight terrestrial Environmental Change Network sites in England and Wales (www.ecn.ac.uk);
- five samples of lodgepole pine (*Pinus contorta*);
- twenty samples of grey heron liver (*Ardea cinerea*) obtained from CEH's Predatory Birds Monitoring Scheme sample archive (http://pbms.ceh.ac.uk/);
- six earthworm samples;
- four rabbits (*Oryctolagus cuniculus*);
- nine pike (*Esox lucius*) obtained from CEH sample archives;
- one toad (*Bufo* spp.);
- five mallards (*Anas platyrhynchos*);
- four samples of trout (*Salmo* spp.) from CEH sample archives.

All rabbit samples, four mallards, three samples of lodgepole pine and two earthworm samples were obtained from Wales, along with nine of the heron liver samples. Lodgepole pine was selected as representative of a species which would have viable seed within its cones during the winter; cone and trunk samples were obtained from all five trees. In addition, two samples of grass snake (*Natrix natrix*) hatchlings were obtained from CEH sample archives. Whilst not a proposed ICRP RAP, reptiles require consideration within radiological environmental impact assessments in the UK. Locations of sampling sites are shown in Figure 2.2.

2.3.1 Sample preparation, analyses and results overview

Samples of lodgepole pine trunk were first washed before being ashed at 450^{o}C. The pine cones were dried at 60^{o}C prior to grinding.

Earthworm samples were kept on wet tissue paper for around two days to allow evacuation of the guts. Samples were freeze dried prior to analyses.

Mallards were plucked and their gastrointestinal tract removed; the liver was removed for separate analysis. Rabbits were skinned and their claws and gastrointestinal tract

removed. Both rabbit and mallard carcasses were washed prior to ashing at 450^{o}C. Herons and mallard liver samples were freeze dried before homogenisation.

The gastrointestinal tract of pike samples was removed prior to ashing the fish at 450^{o}C.

If present, and of suitable size, gonads were removed prior to ashing for separate analyses from pike, mallard and rabbit samples. Gonad samples were freeze dried prior to analysis.

Depending upon sample size, ashed and dried samples were accurately weighed into either 25 ml petri dishes or 150 ml plastic containers. These were sealed and stored for 25 days to allow secular equilibrium to be established. The samples were subsequently analysed by gamma spectrometry on hyper-pure germanium detectors to determine gamma-emitting radionuclide activity concentrations; two-day count times were used for all samples. Due to their small size, toad and grass snake samples were analysed fresh prior to ashing for subsequent analyses to determine total U and Th concentrations.

Samples were finely powdered prior to analysis to determine total U and Th concentrations. Where available, 0.5 g of sample was digested with 10 ml of concentrated nitric acid and two drops of hydrofluoric acid in a microwave oven, dried down and taken up so that the final digest was in 10 ml of 10 per cent nitric acid. Samples were analysed using inductively-coupled plasma mass spectrometry (ICP-MS). Specific activities of 4.07 Bq ^{232}Th mg^{-1} Th and 12.21 Bq ^{238}U mg^{-1} U respectively were used to estimate radionuclide activity concentrations from total U and Th determinations, assuming secular equilibrium in the ^{238}U and ^{232}Th decay series. An assumption of equilibrium between ^{238}U and ^{234}U would appear to be valid for biological samples (see data presented by Eisenbud and Gessel, 1997), enabling ^{234}U activity concentrations to be estimated. Equilibrium between the parents and other members of the two decay chains cannot be assumed for biological samples because of differing environmental and biological behaviours.

Potassium-40 was detectable in the majority of samples. The activity concentrations of ^{238}U and ^{232}Th series gamma-emitting radionuclides were generally below detection limits. Approximately 60 per cent of the 101 samples analysed by ICP-MS had total U concentrations in excess of detection limits; only 25 per cent of samples had measurable total Th concentrations.

Table 2.3 compares the activity concentrations of ^{40}K, ^{232}Th and ^{238}U in reproductive tissues with those in the remainder of the body for rabbits, mallards and pike, and wood for lodgepole pine. For the animals, there are too few samples of testes/ovaries with activity concentrations in excess of detection limits to comment on the data. In the case of lodgepole pine, there is an indication of higher Th and U concentrations in cones compared to wood.

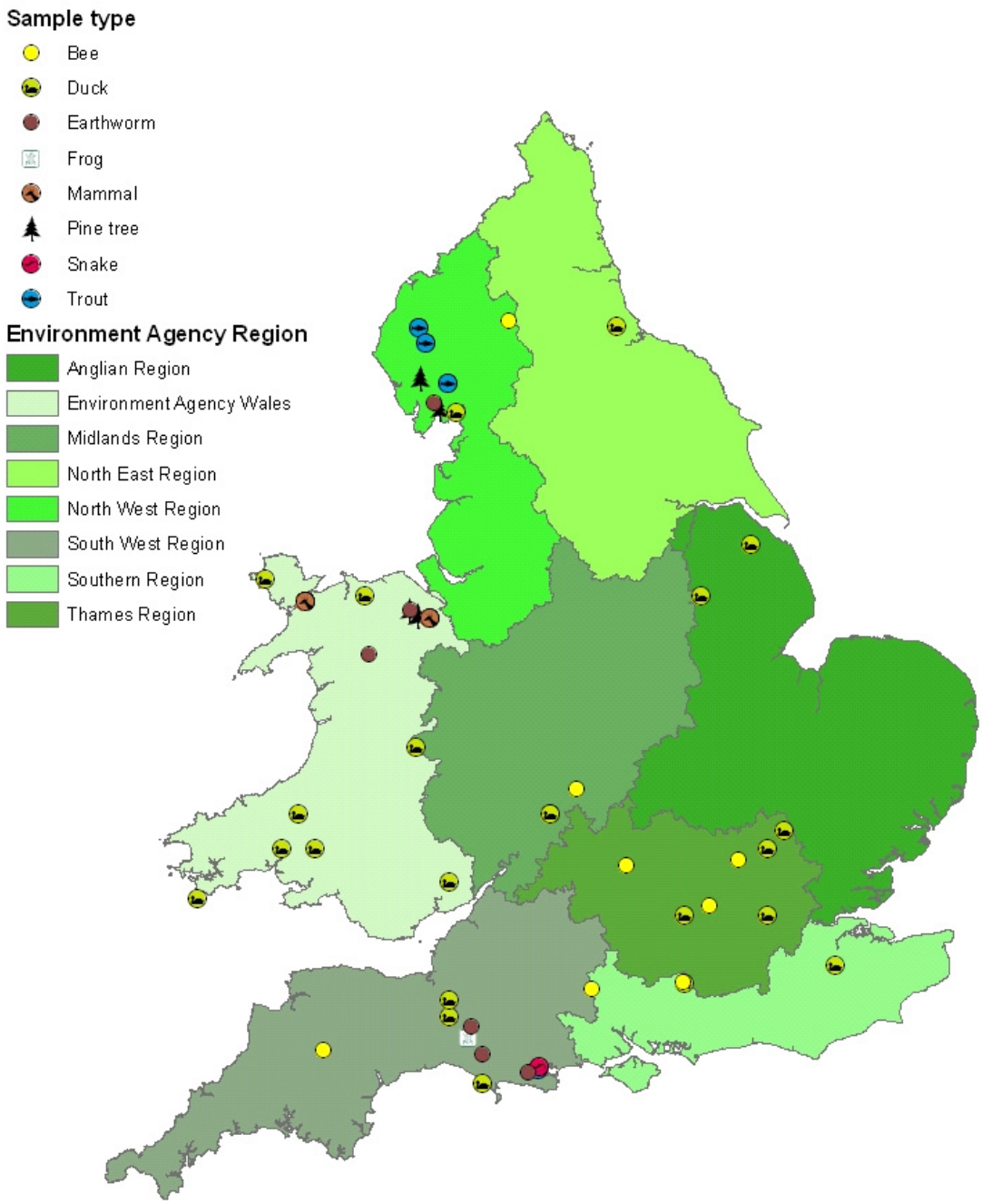

Figure 2.2: Sampling locations of biota analysed to provide additional data

Table 2.3: Comparison of ^{40}K, ^{232}Th and ^{238}U fresh weight (FW) activity concentrations in reproductive tissues and whole body or trunk wood

Sample type and location	^{40}K Bq kg^{-1}		^{232}Th Bq kg^{-1}		^{238}U Bq kg^{-1}	
	Reproductive tissue	Whole body/trunk	Reproductive tissue	Whole body/trunk	Reproductive tissue	Whole body/trunk
Lodgepole pine[1]						
Cumbria	<11	18	6.4×10^{-2}	1.0×10^{-2}	1.3×10^{-2}	2.6×10^{-2}
Cumbria	8.5	12	9.4×10^{-1}	3.4×10^{-3}	1.3×10^{0}	4.5×10^{-3}
Cumbria	<25	10	$<7.9\times10^{-2}$	3.0×10^{-3}	2.8×10^{-2}	3.8×10^{-3}
Flintshire	<16	19	$<8.1\times10^{-2}$	1.4×10^{-2}	2.9×10^{-2}	2.0×10^{-2}
Flintshire	<1.7	18	$<8.3\times10^{-2}$	7.0×10^{-3}	2.5×10^{-2}	9.7×10^{-3}
Denbighshire	<2.5	7.8	$<8.3\times10^{-2}$	2.9×10^{-3}	$<1.7\times10^{-2}$	2.1×10^{-3}
Mallard						
Caernarvonshire[2]	n/a	57	$<3.6\times10^{-2}$	1.8×10^{-2}	9.3×10^{-3}	2.2×10^{-2}
Caernarvonshire[2]	n/a	89	$<3.1\times10^{-2}$	$<6.9\times10^{-3}$	$<6.2\times10^{-3}$	2.1×10^{-2}
Pike						
Bassenthwaite[3]	121	75	$<2.1\times10^{-2}$	$<5.9\times10^{-3}$	$<4.1\times10^{-3}$	$<1.2\times10^{-3}$
Bassenthwaite[2]	<35	90	$<1.9\times10^{-2}$	$<6.9\times10^{-3}$	$<3.8\times10^{-3}$	$<1.4\times10^{-3}$
Bassenthwaite[3]	118	64	$<2.5\times10^{-2}$	$<4.7\times10^{-3}$	$<4.9\times10^{-3}$	$<9.5\times10^{-4}$
Windermere[3]	<40	79	$<2.8\times10^{-2}$	7.7×10^{-3}	$<5.6\times10^{-3}$	4.5×10^{-3}
Windermere[3]	101	103	$<2.6\times10^{-2}$	$<5.3\times10^{-3}$	$<5.2\times10^{-3}$	$<1.1\times10^{-3}$
Derwent Water[3]	n/a	87	$<1.3\times10^{-1}$	$<5.0\times10^{-3}$	$<6.2\times10^{-2}$	$<9.9\times10^{-4}$
Derwent Water[3]	n/a	98	$<2.3\times10^{-1}$	6.1×10^{-3}	$<1.1\times10^{-1}$	1.0×10^{-2}
Rabbit						
Caernarvonshire[3]	<41	80	$<3.3\times10^{-2}$	1.21×10^{-2}	$<6.5\times10^{-3}$	1.1×10^{-2}
Caernarvonshire[3]	<51	<83	$<3.2\times10^{-2}$	$<6.0\times10^{-3}$	$<6.4\times10^{-3}$	$<4.5\times10^{-3}$
Caernarvonshire[2]	n/a	<78	$<4.6\times10^{-2}$	$<7.7\times10^{-3}$	$<9.1\times10^{-3}$	$<5.7\times10^{-3}$
Flintshire[3]	<111	<92	$<3.1\times10^{-2}$	$<6.7\times10^{-3}$	$<6.2\times10^{-3}$	$<2.3\times10^{-3}$
Flintshire[3]	<46	<86	$<3.5\times10^{-2}$	$<6.5\times10^{-3}$	$<7.0\times10^{-3}$	$<7.8\times10^{-3}$
Flintshire[3]	<76	<91	$<3.0\times10^{-2}$	$<6.4\times10^{-3}$	$<6.1\times10^{-3}$	$<3.1\times10^{-3}$

Reproductive tissues analysed: [1]cones; [2]ovaries; [3]testes

n/a = not analysed because of insufficient sample material

2.4 Biota database

All data compiled in the review stage were combined with the results from samples analysed during the course of this work to produce a database (supplied as an MS Excel workbook) together with this report. For biota samples, the quality of data obtained for ^{232}Th and ^{238}U series radionuclides by γ-analyses was poor, generally being below comparatively high detection limits, or having large analytical errors. Consequently, these data have been excluded from the main database although ^{40}K data were retained. Note, however, that some of the ^{238}U and ^{232}Th series data collated in the review were analysed by α-spectrometry and these are included in the database.

Dose conversion coefficients (DCC) for non-human species were derived to estimate absorbed dose to the whole-body based on fresh weight activity concentrations. Some of the compiled data were for specific tissues (for example, much of the collated data were for tissues relevant to the human foodchain). To estimate whole-body activity concentrations from tissue specific values, the same assumptions were made as in the ERICA project (see help functions of ERICA Tool available from http://www.erica-project.org/). Potassium is relatively uniformly distributed within organisms and tissue specific ^{40}K values can therefore be taken to be representative of whole-body activity

concentrations. To convert ^{210}Pb and ^{210}Po activity concentrations reported in the meat of mammals, it was assumed that meat concentrations were representative of all soft tissues, that soft tissues comprised 90 per cent of whole-body liveweight and that 10 per cent and 40 per cent of the whole-body burdens of Pb and Po respectively were in soft tissues. Activity concentrations of U and Th isotopes in meat were assumed to be representative of whole-body concentrations. There was no relationship between either Th or U concentrations in the liver of ducks analysed within this study and those in the remainder of the carcass. Therefore, concentrations determined in the liver of heron samples were taken to be representative of whole-body concentrations.

All concentrations within the database are presented as fresh weight activity concentrations; a dry matter content of 25 per cent was assumed to convert collated dry matter activity concentrations in both flying insects and plant materials. In total, the database contains 585 data lines reporting measured or estimated whole-body activity concentrations; tissue specific results are also retained within the database. For each sample, both species name and attributed ICRP RAP are given.

In addition to activity concentration data, the database contains sampling location information including national grid coordinates, to enable the database to be used with geographical information systems (GIS), together with notes of any assumptions and data manipulations.

Table 2.4 summarises naturally occurring radionuclides in biota within the UK by RAPs. The most abundant data are for ^{40}K followed by ^{232}Th, ^{234}U and ^{238}U; there are relatively few data for ^{210}Pb, ^{210}Po and ^{226}Ra for some RAPs.

Uranium isotopes and ^{232}Th activity concentrations are comparatively high in earthworms, which may be the consequence of residual soil within the gut. The summary values for wild grass in Table 2.4 do not include data from the vicinity of the Springfields plant (Lancashire) which manufactures reactor fuel elements and produces uranium hexafluoride. Uranium-234 and ^{238}U activity concentrations in grass samples from around Springfields have been found to be up to 50 Bq kg^{-1} (FW) (Environment Agency, 2003); these data are retained in the database. The database also contains values for ducks collected close to Springfields, which are within the range of uranium isotope results from elsewhere and are included in the summary values in Table 2.4. Because of the nature of the available data, many are from monitoring activities close to sites discharging radionuclides. The database was inspected to establish whether sites other than Springfields skew the data, but no evidence of this was found.

Table 2.4: Summary of available natural radionuclide activity concentrations in biota within the UK

RAP		Activity concentration (Bq kg^{-1}) FW							
		^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{230}Th	^{232}Th	^{234}U	^{238}U
Wild grass	Mean*	270	2.54	3.81	0.33	2.7×10^{-2}	0.13	0.41^{++}	0.40^{++}
	Min	40.6	8.5×10^{-2}	0.18	3.7×10^{-2}	2.8×10^{-3}	$<5.5\times10^{-2}$	$<2.0\times10^{-2}$	$<2.0\times10^{-2}$
	Max	708	13	17	1.70	7.0×10^{-2}	0.81	2.9	2.3
	n	91	45	46	40	4	46	55	59
Pine tree	Mean	35.2	0.19	0.10	7.8×10^{-2}	6.8×10^{-3}	5.7×10^{-3}	7.6×10^{-3}	7.3×10^{-3}
	Min	7.79	3.4×10^{-2}	3.5×10^{-2}	4.6×10^{-3}	$<4.8\times10^{-3}$	$<4.7\times10^{-3}$	1.1×10^{-3}	7.7×10^{-4}
	Max	135	0.5	0.16	0.18	1.4×10^{-2}	1.4×10^{-2}	2.6×10^{-2}	2.6×10^{-2}
	n	8	5	5	5	5	11	15	15
Brown seaweed	Mean	138	2.1	0.49	n/a	7.7×10^{-2}	4.3×10^{-2}	1.72	1.16
	Min	48.6	0.35	6.5×10^{-2}		7.0×10^{-3}	3.9×10^{-3}	0.19	0.16
	Max	187	8.5	3.6		1.4×10^{-1}	0.18	4.5	4.0
	n	13	62	33		6	19	13	43
Earthworm	Mean	44.6	n/a	n/a	n/a	n/a	0.68	0.74	0.74
	Min	<7.49					0.24	0.36	0.36
	Max	54.6					1.41	1.44	1.44
	n	6					6	6	6
Bee	Mean	229	n/a	n/a	n/a	n/a	$<3.1\times10^{-2}$	1.5×10^{-2}	1.5×10^{-2}
	Min	<38.6					$<3.1\times10^{-2}$	$<6.1\times10^{-3}$	$<6.1\times10^{-3}$
	Max	240					$<3.1\times10^{-2}$	2.4×10^{-2}	2.4×10^{-2}
	n	8					8	8	8
Frog	Mean	<16.1	n/a	n/a	n/a	n/a	1.8×10^{-2}	3.4×10^{-2}	3.4×10^{-2}
	n	1					1	1	1
Duck	Mean	108	n/a	n/a	n/a	1.1×10^{-2}	6.9×10^{-2}	3.8×10^{-2}	4.1×10^{-2}
	Min	<26				$<5.0\times10^{-3}$	$<1.9\times10^{-3}$	$<6.2\times10^{-3}$	$<6.2\times10^{-3}$
	Max	173				1.5×10^{-2}	0.18	0.13	0.13
	n	40				5	32	27	27
All mammals	Mean	104	0.09	0.45	2.1×10^{-2}	2.9×10^{-3}	2.7×10^{-3}	4.7×10^{-3}	5.3×10^{-3}
	Min	36.3	$<1.8\times10^{-2}$	$<9.9\times10^{-2}$	1.3×10^{-3}	1.8×10^{-3}	$<9.0\times10^{-4}$	4.2×10^{-4}	3.3×10^{-4}
	Max	178	0.5	1.62	0.14	3.9×10^{-3}	1.2×10^{-2}	1.1×10^{-2}	1.5×10^{-2}
	n	153	32	31	24	2	38	12	11
Trout	Mean	234	n/a	n/a	n/a	n/a	1.8×10^{-2}	9.0×10^{-3}	9.0×10^{-3}
	Min	<64.6					$<4.0\times10^{-3}$	$<9.5\times10^{-4}$	$<9.5\times10^{-4}$
	Max	1510					4.5×10^{-2}	1.9×10^{-2}	1.9×10^{-2}
	n	44					17	17	17
Snake^{+++}	Mean	<66	n/a	n/a	n/a	n/a	1.2×10^{-2}	1.4×10^{-2}	1.4×10^{-2}
	Min	<54					6.2×10^{-3}	9.7×10^{-3}	9.7×10^{-3}
	Max	<78					1.7×10^{-2}	1.9×10^{-2}	1.9×10^{-2}
	n	2					2	2	2

*Mean does not include values below detection limits. Note the mean is estimated using the number of data entries; it is not weighted for the number of observations (the majority of data entries are n=1). Arithmetic mean is reported.

$^{++}$Does not include data from vicinity of the Springfields plant.

$^{+++}$Not a proposed RAP.

n = number of data entries (not observations) including values below detection limits.

n/a = no data.

2.4.1 Marine biota

Marine biota were outside the remit of this work. However, if data on naturally occurring radionuclides in marine biota within UK waters were found during the course of the literature review, these were collated. The database from a review of naturally occurring radionuclides in marine biota conducted by the FASSET project, considering publications from 1970 to 2002, was also made available (Brown *et al.,* 2004). Data for UK waters were extracted from this and added to those compiled from the refereed literature. These data are provided as an additional worksheet within the database.

Subsequent to the period covered by the Brown *et al.* (2004) review, Young *et al.* (2002) have reported results for naturally occurring radionuclides in various seafoods from around the coast of the UK. These are compared by the authors to a collation of previously reported values. The Young *et al.* (2002) reference represents a potentially useful source of data for marine biota in addition to that compiled here (from the Brown *et al.* (2004) review and *ad hoc* references identified during the collation of primarily terrestrial data).

3 Environmental media

For over 35 years, the British Geological Survey (BGS) have been conducting geochemical surveys of the UK (Table 3.1, Figure 3.1). This work, now referred to as the G-BASE project (Johnson and Breward, 2004; Johnson *et al.,* 2005), includes determinations of K, U and Th concentrations in soils, stream waters and stream sediments. Estimates of ^{40}K, ^{238}U and ^{232}Th series radionuclides in environmental media were based upon this extensive data source, supplemented with data from other BGS projects and the Wolfson geochemical atlas (Webb *et al.,* 1978) as appropriate.

3.1 G-BASE sampling

Sampling procedures are detailed in Johnson (2005). Stream sediments and waters are collected at an optimum density of one per 1.5 to 2 km^2 and are taken ideally from 1st or 2nd order streams, avoiding any potential source of contamination that may lie upstream. Water samples are taken before the stream bed is disturbed. Stream sediment collection involves removing the oxidised layer from the stream bed and removing the sediment using a trowel. This is then sieved through a coarse sieve (two mm) and then a fine sieve (150 µm). The <150 µm fraction is left to settle in a fibreglass pan and is decanted into a sample collection bag.

Soil samples are collected at an optimum density of one every 2 km^2. They are collected using a handheld Dutch soil auger and are taken from the surface (5 to 20 cm) and from a depth of 35 to 50 cm. Each sample is made of a composite of material from auger flights taken from five holes distributed within an area of approximately 20 m^2.

3.2 G-BASE history and the methods used to create consistent national datasets

With a long-running project such as G-BASE (and its precursors), changes in the sampling and analytical techniques used, their sensitivity and the range of elements determined occur and can be significant (see Table 3.1). Over the 35 years since the inception of the regional geochemical survey project that has evolved into G-BASE, considerable advances have been made in instrumental analytical chemistry. The number of elements that can be measured in solid material has increased from sixteen to forty-six, while determinations on stream waters have increased from four to forty elements and parameters. Uranium in both stream sediments and waters was determined from the start of the G-BASE project, as was K in sediments. Potassium in stream waters and Th in all sample media are much later additions. As the project started in northern Scotland, the more recent coverage in central England and Wales generally has a better range of elements and data quality than that further north. The collection of soil samples is also a relatively recent feature.

These historical factors have a significant impact on data coverage and quality. Different methods of analysis require levelling of datasets. Changes in sample processing (such as filtered versus unfiltered waters) and the analysis of either surface or subsurface soils

in different areas have also required normalisation for this project. Such data levelling is a potential source of error that must be borne in mind when making use of the data presented.

Table 3.1: Summary of G-BASE samples collected in chronological order and the analytical methods used for different atlas areas

	Stream sediments			Soils			Waters		
Atlas area	**K**	**Th**	**U**	**K**	**Th**	**U**	**K**	**Th**	**U**
Lake District	DR-OES	NA	DNA	NC	NC	NC	NA	NA	DNA
NE England	DR-OES	NA	DNA	DR-OES	NA	DNA	NA	NA	LF
NW England & N Wales	DR-OES	Part by XRF	DNA	DR-OES	Part by XRF	DNA	NA	NA	LF
Wales & W Midlands	XRF	XRF	XRF	XRF	XRF	XRF	ICP-AES	NA	LF
Humber-Trent	XRF	XRF	XRF	XRF	XRF	XRF	ICP-AES	ICP-MS	ICP-MS
East Midlands	XRF	XRF	XRF	XRF	XRF	XRF	ICP-AES	ICP-MS	ICP-MS
East Anglia	XRF	XRF	XRF	XRF	XRF	XRF	ICP-AES	ICP-MS	ICP-MS

NC, not collected; NA, not analysed.
DR-OES, direct-reading optical emission spectrometry; DNA, delayed neutron activation; LF, laser fluorimetry; XRF, x-ray fluorescence spectrometry; ICP-AES, inductively coupled plasma atomic emission spectrometry; ICP-MS, inductively coupled plasma mass spectrometry.

3.2.1 Potassium in stream sediments and soils

Potassium was originally determined in stream sediments using optical spectrophotometry, then direct-reading optical emission spectrometry, and currently, X-ray fluorescence (XRF). There were relatively few problems in levelling data between areas using these different analytical methods, despite some variation in calibration, and the 'national' dataset was of good quality and integrity (see Johnson *et al.,* 2005). Where available, soil data was similarly reliable.

Stream sediment K concentration data are currently not available from G-BASE for southern England; they are, however, available within the Wolfson geochemical atlas (Figure 3.1; Webb *et al*., 1978). Linear quantile transformation was used to level K in the Wolfson stream sediment data to G-BASE equivalent concentrations (see Figure C.1 in Appendix C.1).

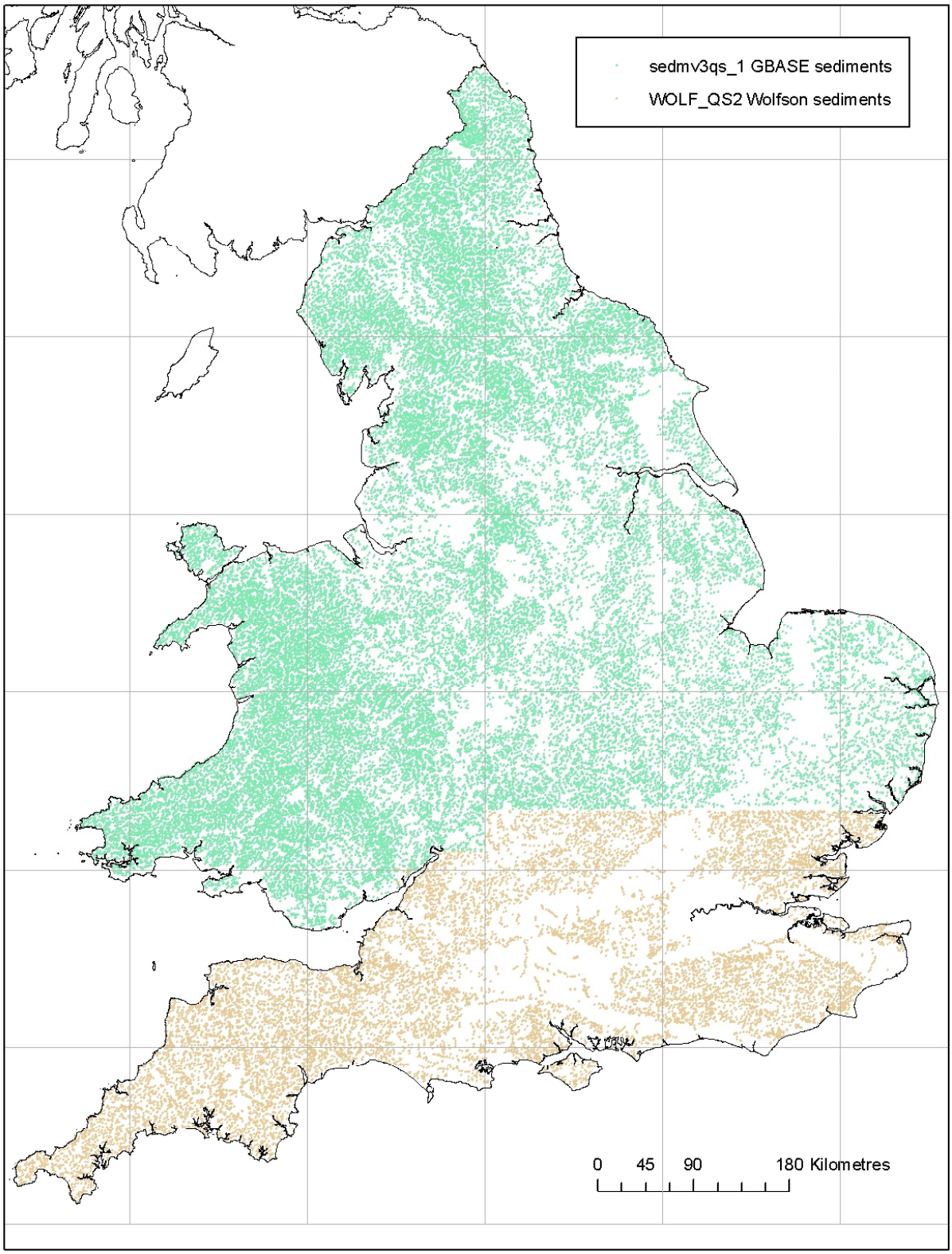

Figure 3.1: Availability of G-BASE geochemical atlas data for K in stream sediments (green points) and coverage of Wolfson atlas data used for southern England (light brown points)

3.2.2 Potassium in stream waters

As a 'major' ion in solution in natural stream waters, K is determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) which gives excellent results with a limit of detection below 0.04 mg l^{-1}. Data are available for Wales and later regional survey areas.

3.2.3 Uranium in stream sediments and soils

Prior to the adoption of XRF as the principal analytical method for G-BASE solid materials (Wales atlas area onwards), uranium in stream sediments was determined by the delayed neutron activation (DNA) method (Table 3.1), originally at the HERALD reactor at Aldermaston and later at ICI Billingham. Detection limits claimed were 0.1 and 0.05 mg kg^{-1} U respectively. With XRF analysis, the quoted limit of detection is 0.5 mg kg^{-1}, but at a much better precision.

Linear quantile transformation was used to level the DNA uranium data from northern England to XRF equivalent uranium data in a similar way to that used for potassium. The same transformation was applied to data for central and North Wales and South West England from BGS's Mineral Reconnaissance Programme (for example, Haslam *et al.,* 1990) as coverage was lacking for these areas within G-BASE. These data were also the result of DNA analyses and a quantile-quantile comparison with XRF data from the same areas suggested a very similar relationship as for the G-BASE data (see Figures C.2 and C.3 in Appendix C.1).

3.2.4 Uranium in stream water

Early determinations of U in stream water were made by the DNA method (Table 3.1) using the HERALD reactor at Aldermaston, as for the U in stream sediments. This was a high neutron-flux research reactor and was capable of a claimed detection limit of 0.5 µg l^{-1} U in waters. However, the difficulties of achieving reliable data at these very low levels due to poor counting statistics during analysis is shown by variable and high blank values in some batches. Coupled with this, the fact that the samples were not filtered and were irradiated complete with their collection bottles gives further sources of random error.

When the HERALD reactor closed, no other UK reactor could achieve a suitable detection limit for U in waters by the DNA method, so for the Tyne-Tees, North West England and Wales atlas areas, laser-induced fluorimetry was used. This should have been capable of a limit of detection of 0.2 µg l^{-1} for 'pure' water samples, but suffered from interferences for some calcium-rich and organic-rich samples. Original samples were not filtered, but if precipitates formed on addition of the required buffer solution, these had to be filtered to avoid serious errors. Precision therefore suffered, and following instrumental problems during the determination of the Wales stream waters, the method was abandoned. Nevertheless, many values above about 0.8 µg l^{-1} are probably reliable in these atlas areas, but some apparently moderately high values may not be.

The relatively recent development of inductively-coupled plasma source mass spectrometry (ICP-MS) analysis has been a major breakthrough in the determination of many elements at extremely low concentrations in solution. Uranium is an excellent example, and on the same filtered water sample as is used for other G-BASE elements

in waters, a detection limit of 0.01 μg l^{-1} is achievable, with no significant interferences. As water samples degrade rapidly with storage, unlike solid materials, samples were not analysed by more than one method. No direct comparison is therefore possible between techniques.

Given this and the disparity in detection limits and data quality between the analytical methods used, the creation of a 'seamless' national dataset for England and Wales was difficult. Attempts were made to normalise the different datasets (derived using different analytical methods) across atlas boundaries using quantile-quantile regression between similar lithologies. The approach was essentially the same as that adopted for K and U in stream sediments. The result, however, was poor, and probably compounded by fact that the ICP-MS data are for filtered waters and the other methods used unfiltered samples that will include colloidal or suspended U species. Whilst some overall patterns seem fairly consistent, obvious boundaries exist, such as that between the Humber-Trent (ICP-MS) and North West England (laser fluorimetry) data along the 400 km East National Grid line. Comparative values across such boundaries are so different that they render the older data unusable for the present purpose. Consequently, only data from ICP-MS analysis are presented here.

3.2.5 Thorium in stream sediments and soils

Thorium was not determined until the adoption of XRF analysis (Table 3.1), so Th data are only available for G-BASE sediments and soils for Humber-Trent, central and eastern England and parts of North East England, North West England and North Wales. However, to provide an estimate of Th levels in sediments for areas surveyed earlier, we have used the known correlation of Th with other similar resistate elements (elements resistant to weathering) to extrapolate the 'missing' Th data to these areas. From observations made on the Wales and Humber-Trent datasets, positive linear correlations were observed between Th and La, Y and Zr. The strongest correlation, that of Th and Ce, cannot be used as Ce is only available on XRF data, while data for the other elements are available by previous methods.

From the correlations examined, a formula was derived which gives a first-order approximation for Th levels in sediments that can be compared with the real data for Wales. The formula used was:

$$Th(estimated) \approx (0.22*La + 0.3*Y + 0.008*Zr)/2.5$$

where all concentrations are in mg kg^{-1} dry weight; $R^2 = 0.22$.

There was a reasonable correlation between estimated values and measured data, but with a fairly high degree of scatter, and trends within the actual data that cannot be duplicated by the estimated data. The extrapolation method is only valid for situations where the proportion of Th strongly correlated with the other resistate phases is high, and cannot deal with those where a good proportion of the Th is in other phases and is behaving independently of La, Y and Zr. However, up to about 20 mg kg^{-1} the similarity is adequate for a first-order estimate, although it should be borne in mind that in some cases the estimated Th is significantly higher than the real data (see Figure C.4 in Appendix C.1).

3.2.6 Thorium in stream water

The determination of Th in stream water is a recent addition to the ICP-MS list of G-BASE elements (Table 3.1) and is only available for Humber-Trent and later areas. There are concerns with the stability of Th in acidified water samples and the accuracy and precision of this data have yet to be fully characterised. Despite a very low quoted limit of detection (0.01 µg l^{-1}), the extremely low natural abundance of Th in surface waters only allows the top 10 per cent of the data (>0.06 µg l^{-1}) to be regarded as reliable.

3.3 Data processing

Data for Th, U and K concentrations are provided on a 5 x 5 km grid square basis as files suitable for import into a GIS.

3.3.1 Stream sediments

For potassium there is almost complete coverage for England and Wales, using G-BASE and Wolfson atlas data, except in some limestone areas where surface drainage is absent, and Greater London (Figure 3.2). Figure 3.2 presents the geometric mean concentrations of K in sediments for 5 x 5 km grid squares (25 km^2) based on the actual data points in each square. Class intervals used for all soil and sediment maps presented in this report are the 10, 25, 50, 75 and 90 percentile values for the raw data distribution.

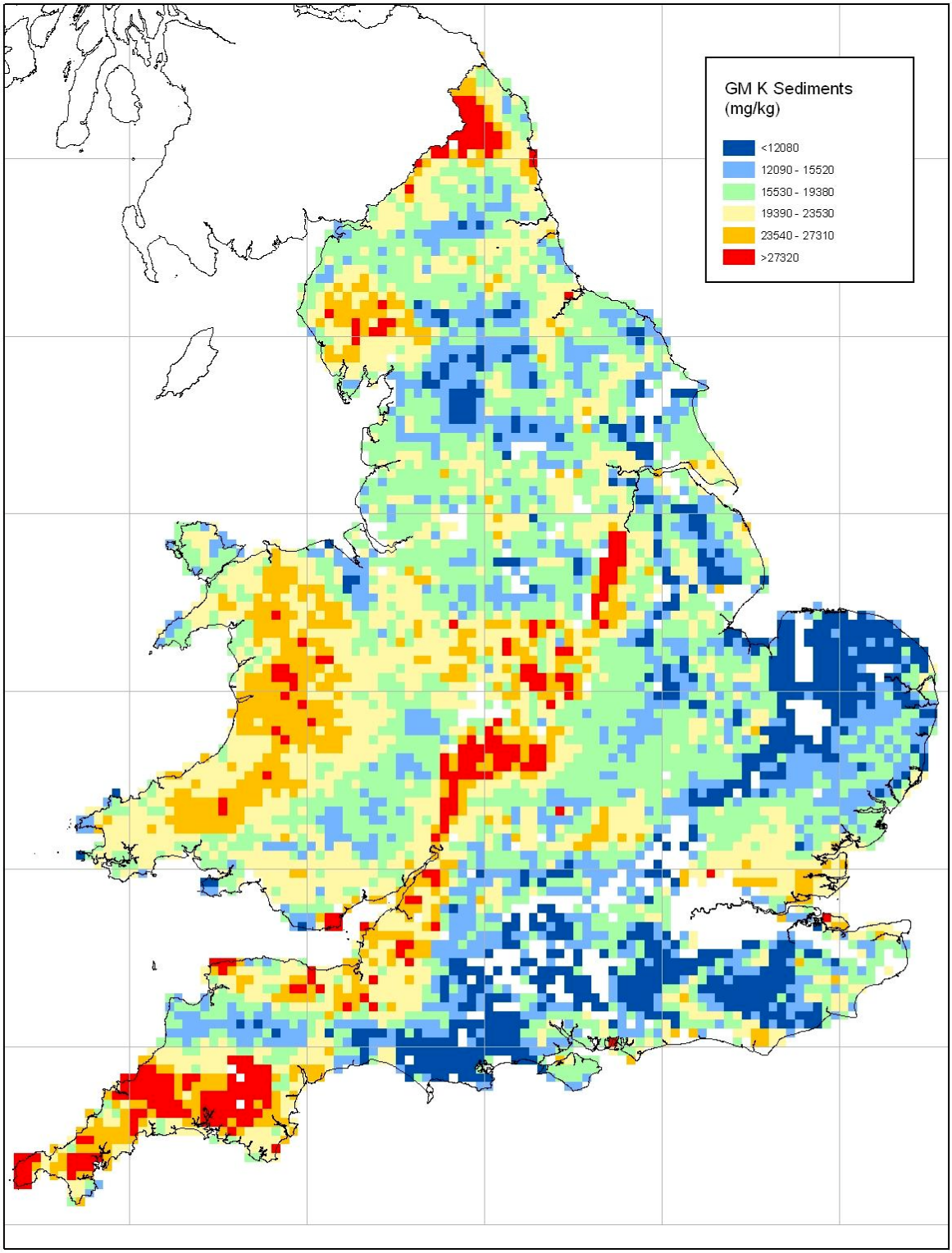

Figure 3.2: Concentrations of K in stream sediments presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

For Th and U there is no coverage in southern England within G-BASE or other datasets, except in parts of the South West. Hence the 5 x 5 km geometric mean coverage for actual samples is more limited (Figures 3.3 and 3.4). To extend Th and U coverage to southern England, geological extrapolation was applied. The strong link between stream sediment geochemistry and geology can be used to provide an estimate of U and Th concentrations for areas currently lacking data. Simplified bedrock and superficial geology codes based on BGS 1:50,000 scale digital geological maps (see Appleton, 2005) were attributed to each stream sediment location. Geometric means for each element were calculated for each one km grid square and parent material (bedrock plus superficial geology) polygon from the nearest five stream sediment values for that parent. These data were then used to compute geometric means for each five km grid square using area-weighted geometric mean values for each parent material found in the square (Figures 3.5 and 3.6). This involved summing the products of the mean radionuclide content for each one km grid square/parent material polygon (derived from the five nearest data points on that parent material) and the area of that polygon and dividing the sum of those products by the total area of the 5 x 5 km square (25 km^2):

$$GM_{5km} = \sum_{1}^{n} \frac{(\overline{X}_1 Area_1) + (\overline{X}_2 Area_2) + ...(\overline{X} n Area_n)}{25km^2}$$

where $\overline{X}$ is the GM for a one km grid square/parent material polygon.

In the files supplied, if the area figure is less than 2.5 x 10^9 m^2, this indicates that some parent material polygons in the square have less than five analysed stream sediment samples. The geometric mean was not calculated for these polygons and hence the area used to compute the overall geometric mean for the 5 x 5 km square is less than the total area.

In most cases, the nearest five points used to calculate geometric means for each one km square and parent material polygon were within five km, but in some cases the furthest point used was over 100 km away. Given that there can be lateral variations in mapped geological units this could lead to poor estimation of element concentrations. The extrapolated values should be taken only as a guide to likely average concentrations.

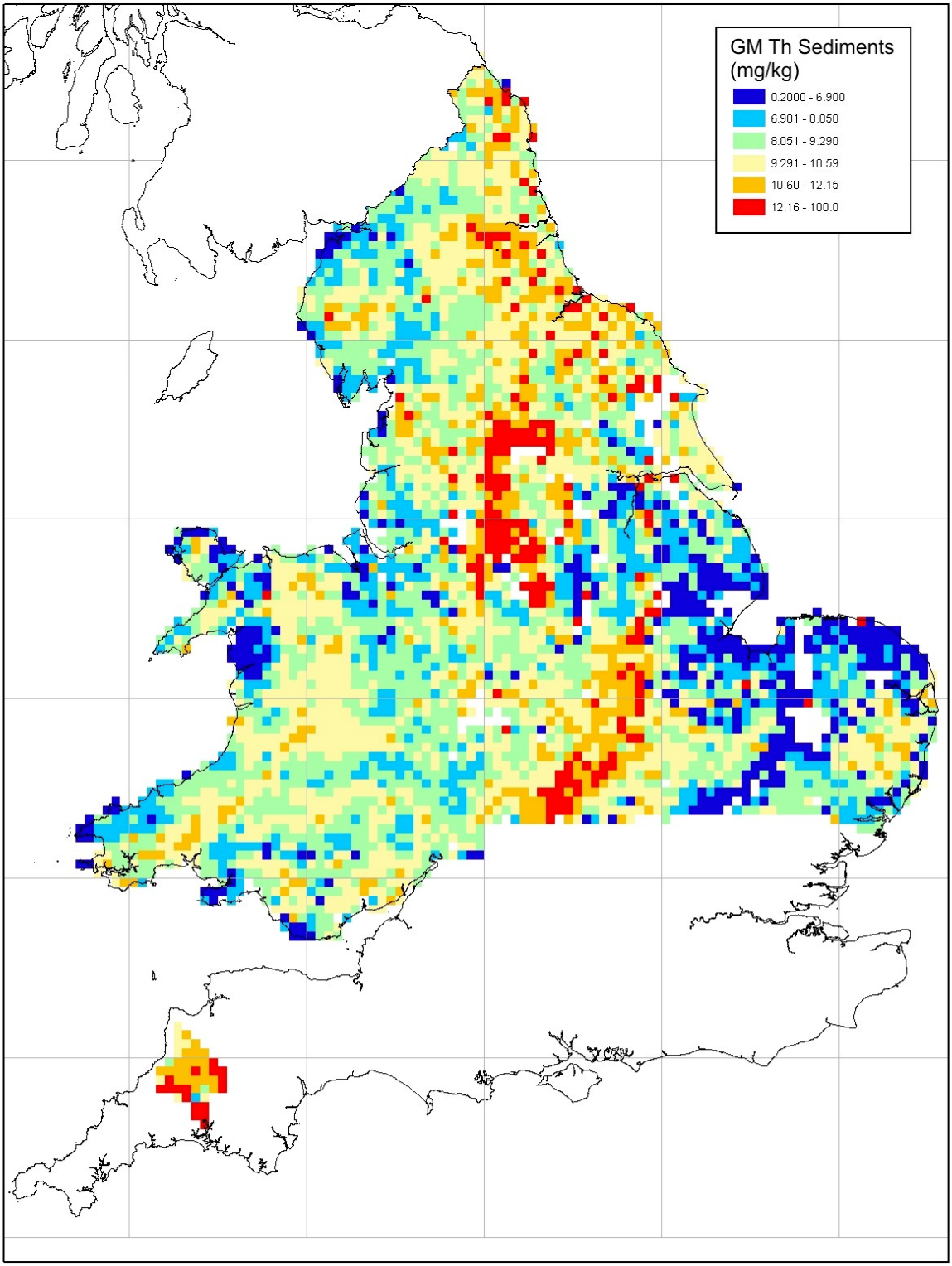

Figure 3.3: Concentrations of Th in stream sediments presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

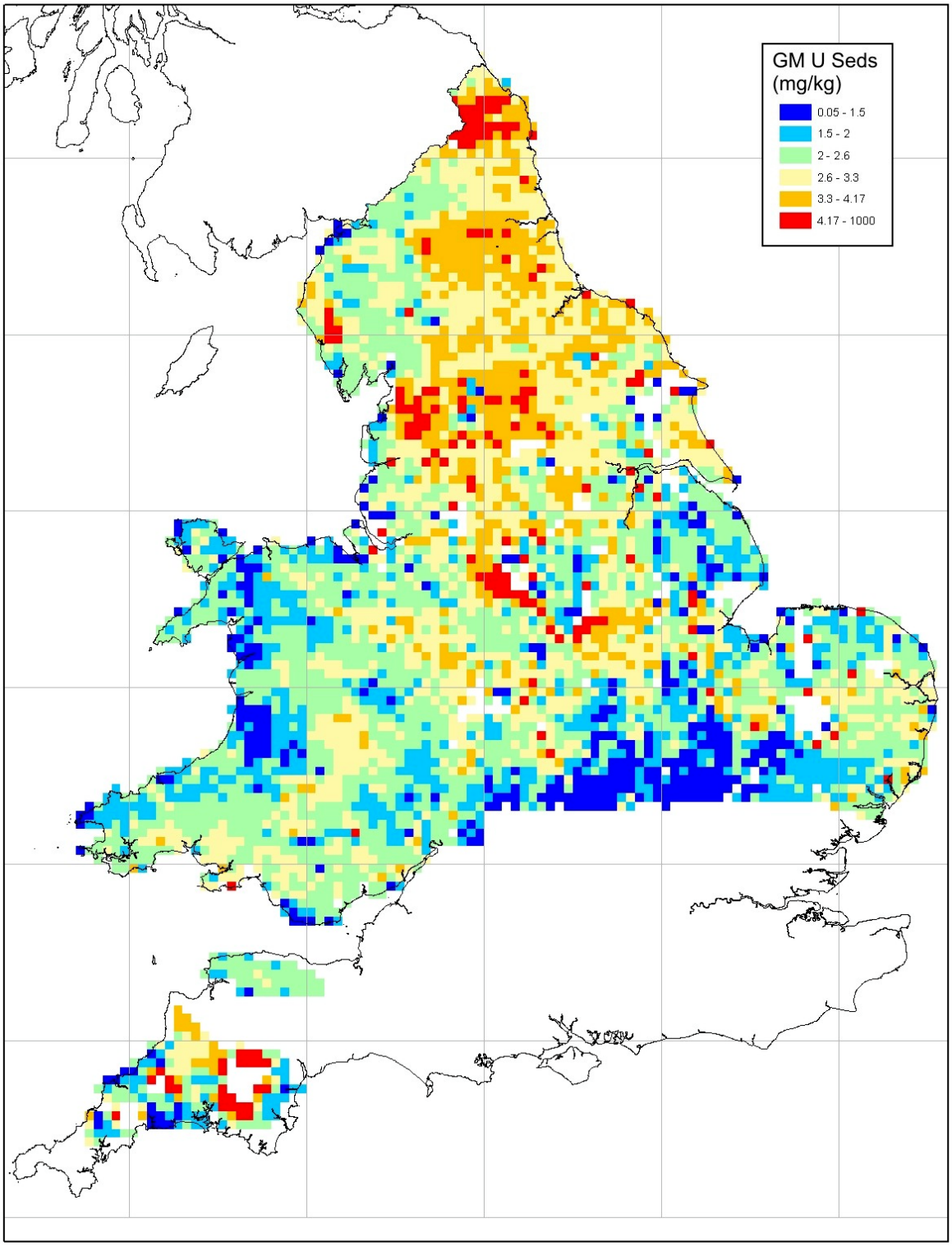

Figure 3.4: Concentrations of U in stream sediments presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

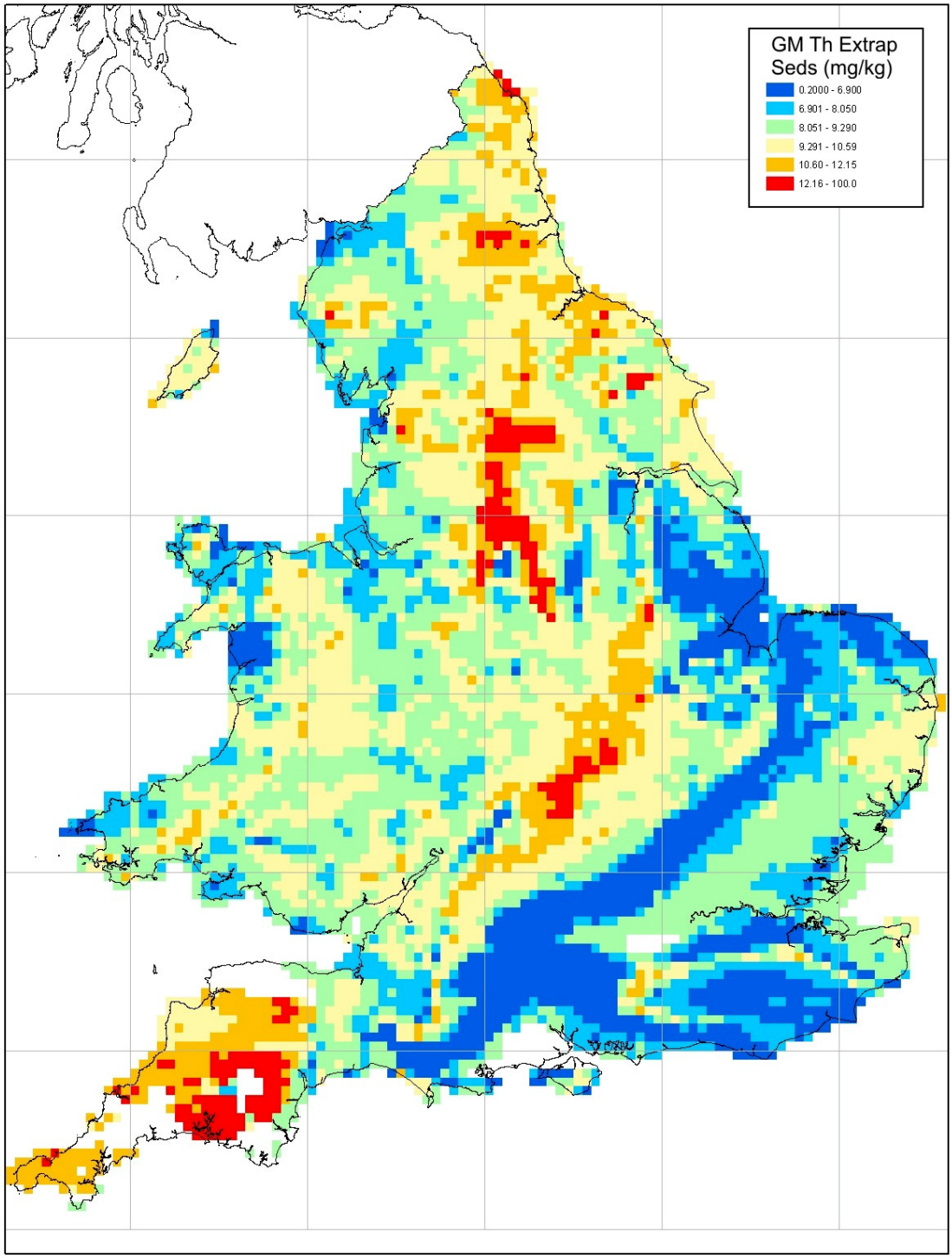

Figure 3.5: Concentrations of Th in stream sediments derived by geological and geochemical extrapolation (mg kg^{-1} dry weight)

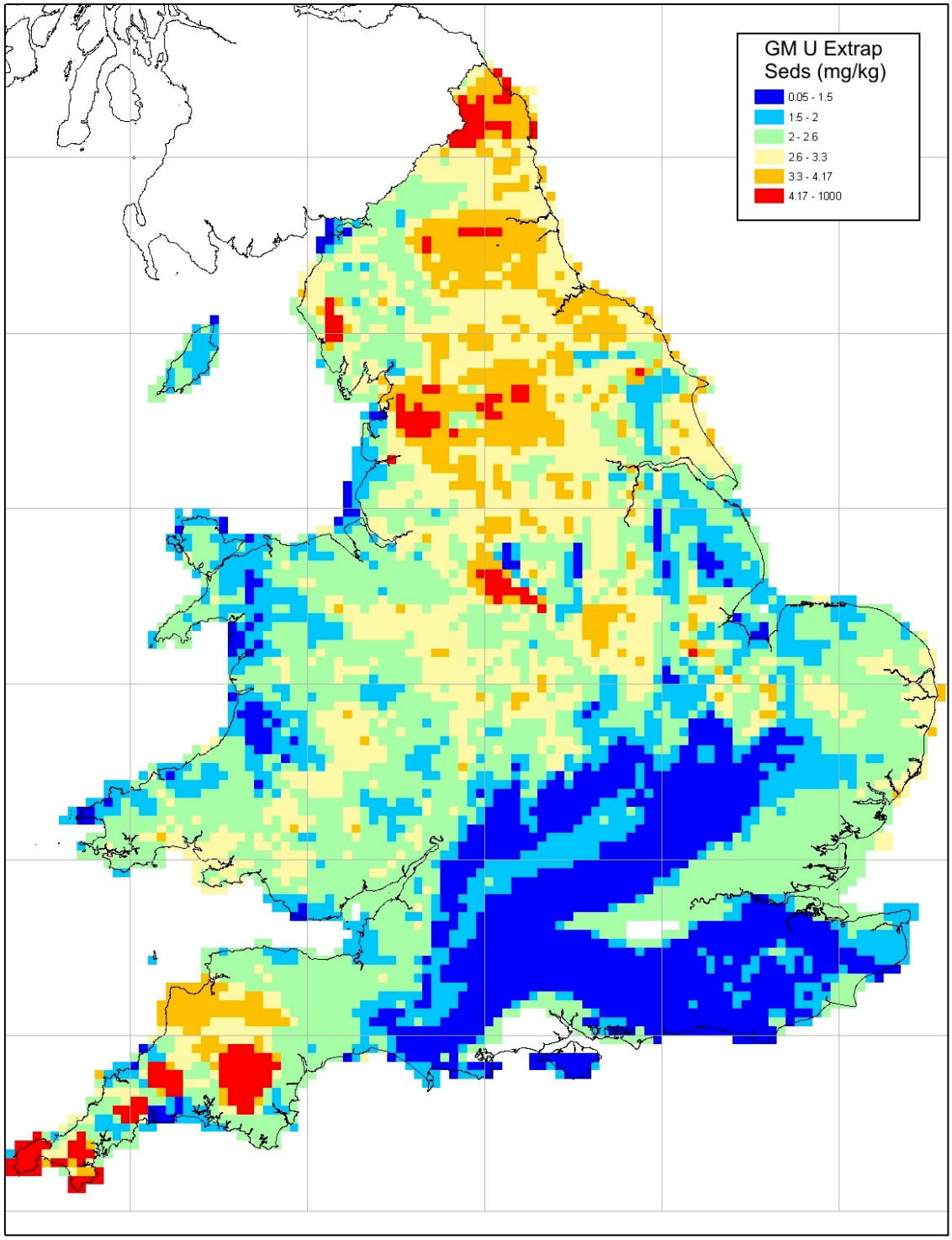

Figure 3.6: Concentrations of U in stream sediments derived by geological and geochemical extrapolation (mg kg^{-1} dry weight)

3.3.2 Soils

The processing of the soils data was complicated by the analysis of different depth samples in different atlas regions due to funding limitations and evolving use of the data. Thus, for the Humber-Trent atlas both surface (A horizon) and subsurface soils were analysed, for East Midlands only the subsurface samples were analysed, whilst for East Anglia only the surface samples were analysed. Parts of the Tyne-Tees and Liverpool Bay atlas areas were covered by soil sampling, but only where surface drainage was sparse (and stream sediment samples lacking). In addition to the levelling of data produced by different methods, normalisation between the different depth samples had to be considered. Where data existed for both surface and subsurface soils, the values were compared. This showed a good linear relationship between surface and subsurface K data, the subsurface values being generally higher (see Figure C.5 in Appendix C.1) and the regression between the two was used to convert the surface soils to equivalent subsurface values. For U data, the two values were so close to a 1:1 relationship that the data could be combined without transformation. In the case of Th, there was a lot of scatter at lower concentrations and a much less clear-cut relationship between the surface and subsurface results. This was due, in large part, to the lack of appropriate data for the comparison, which was limited to a few urban areas. As a result, it was felt that transformation of the data was not justified. Given that the U data were essentially comparable at both depths, the Th data were also used untransformed.

Geometric mean data for soils based on actual samples in 5 x 5 km grid squares are given in Figures 3.7 to 3.9. A similar geological extrapolation to that undertaken for the stream sediments was carried out to extend coverage across England and Wales. As for stream sediments, the well-developed relationship between soil geochemistry and geology should allow reasonable estimates of K, U and Th contents to be derived. Results for each one km square/parent material polygon were computed and used to derive geometric means for each 5 x 5 km grid square using area-weighted geometric mean values for each parent material found in the square (Figures 3.10 to 3.12).

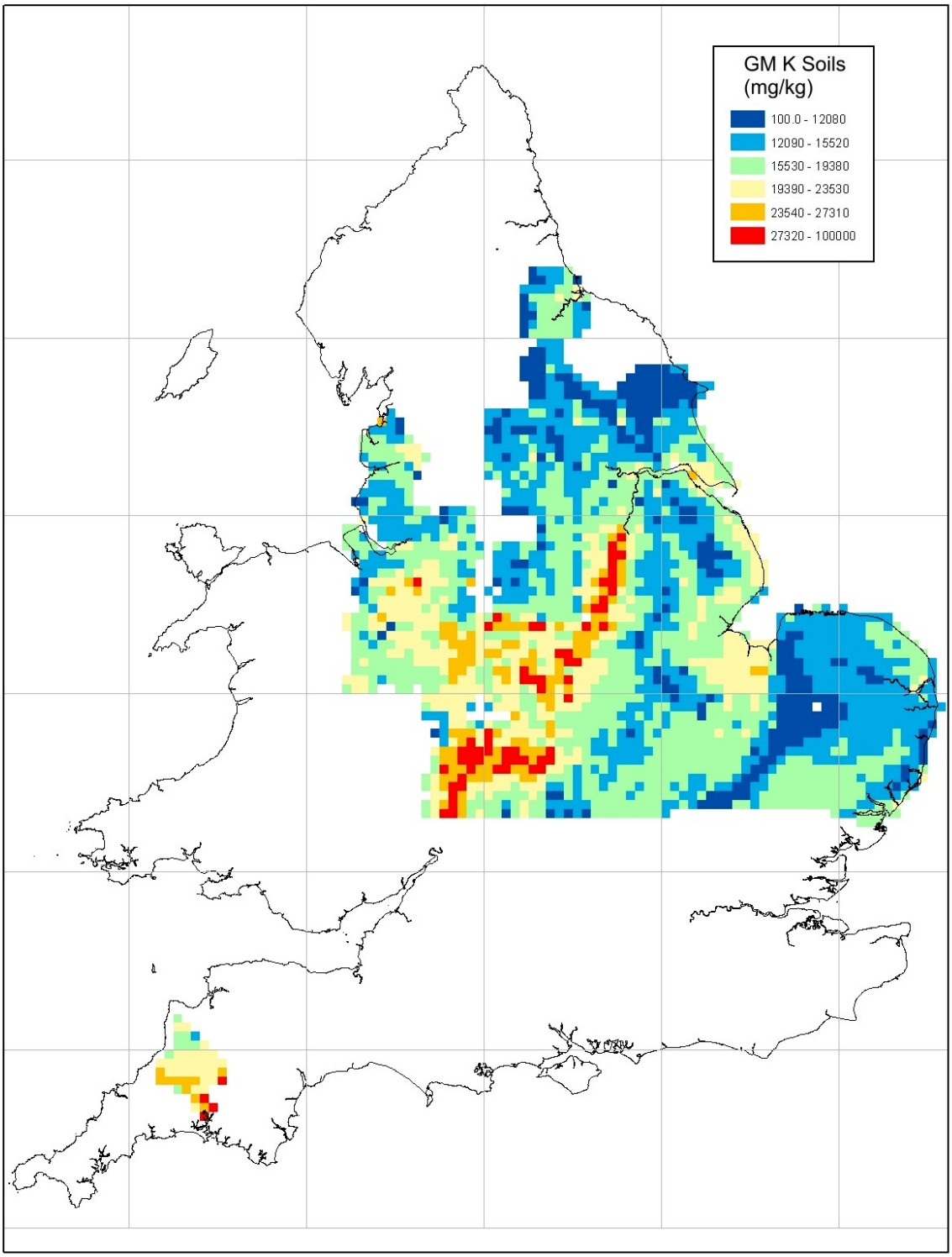

Figure 3.7: Concentrations of K in soils presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

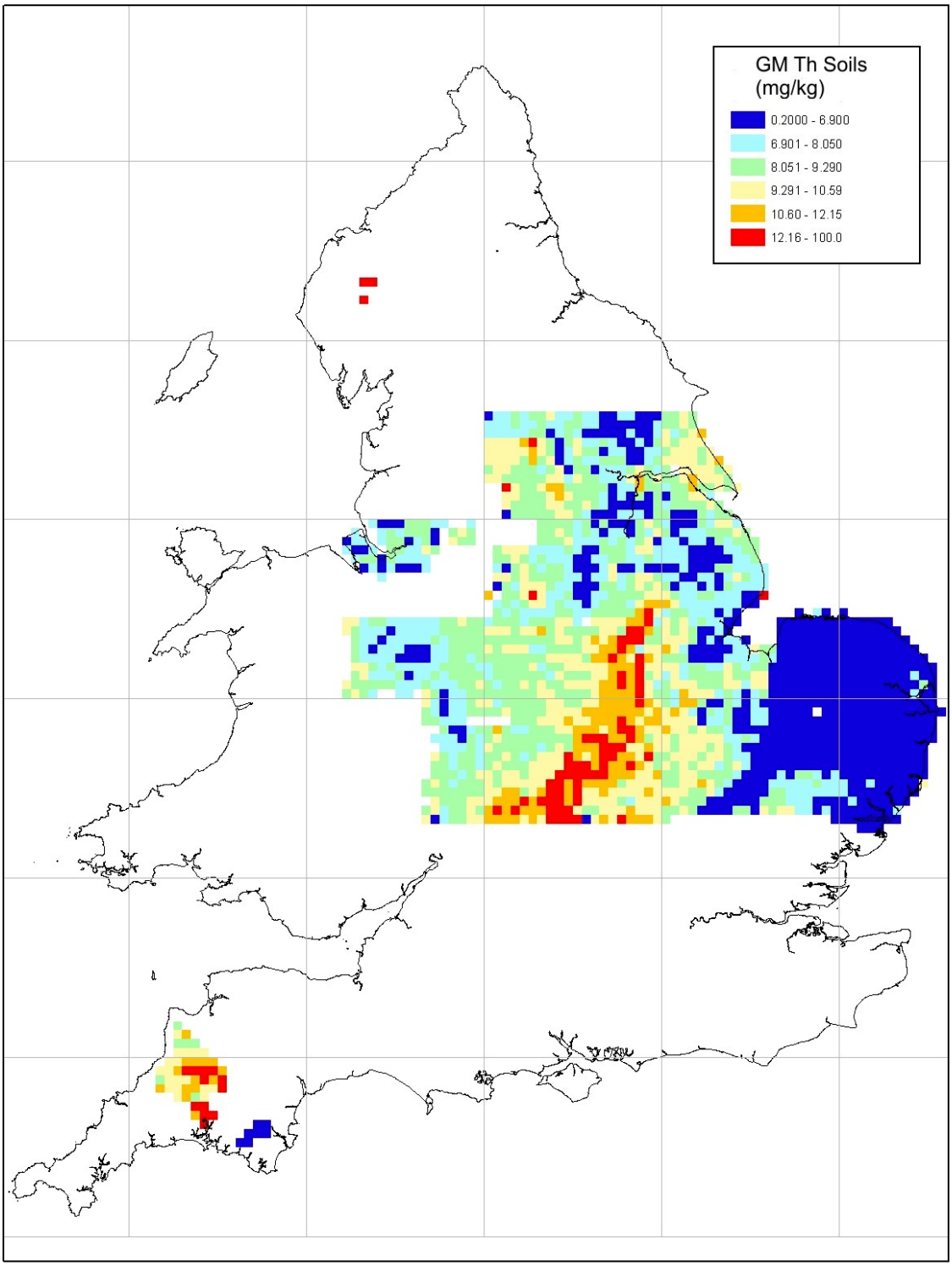

Figure 3.8: Concentrations of Th in soils presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

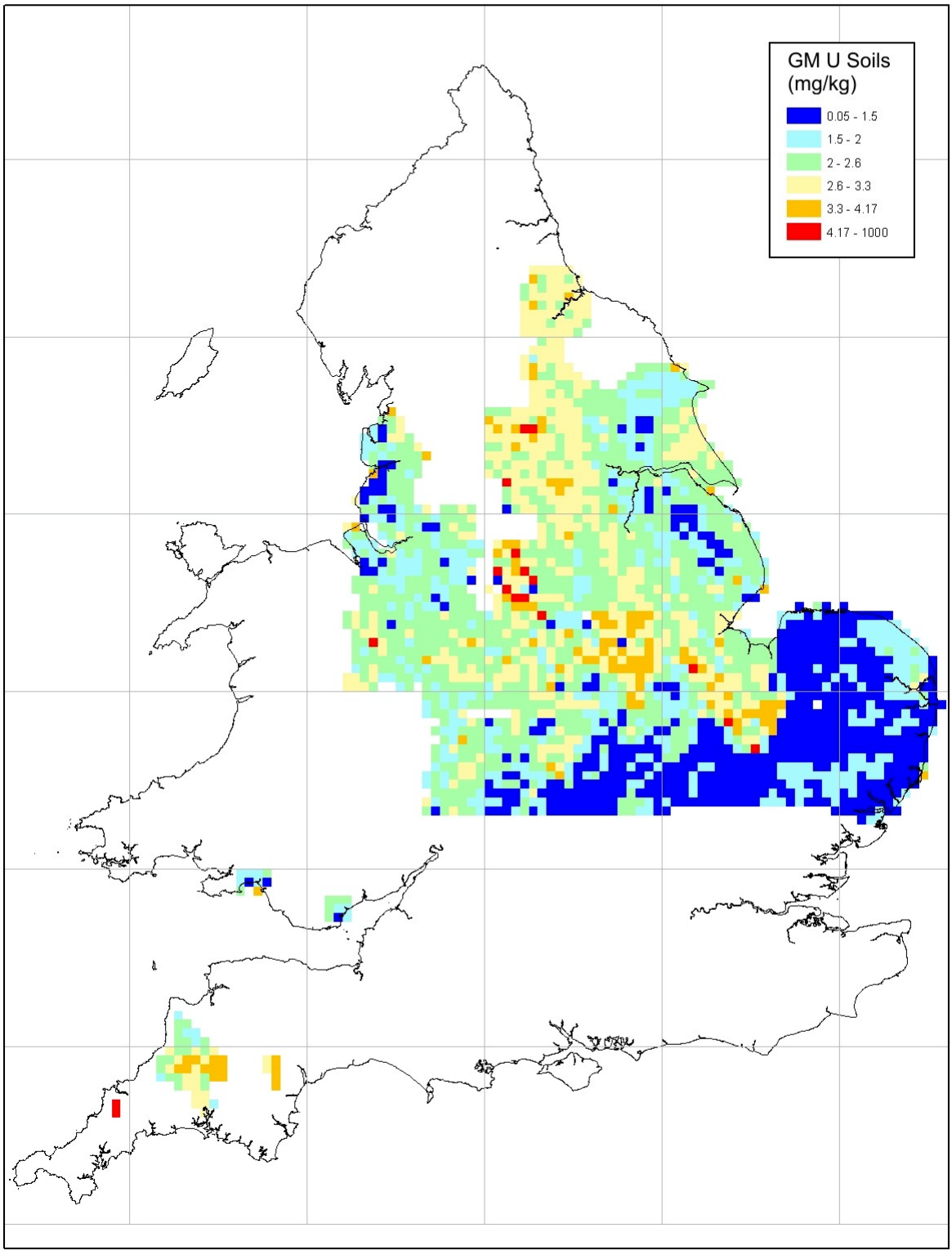

Figure 3.9: Concentrations of U in soils presented as geometric mean of measurements within 5 x 5 km squares (mg kg^{-1} dry weight)

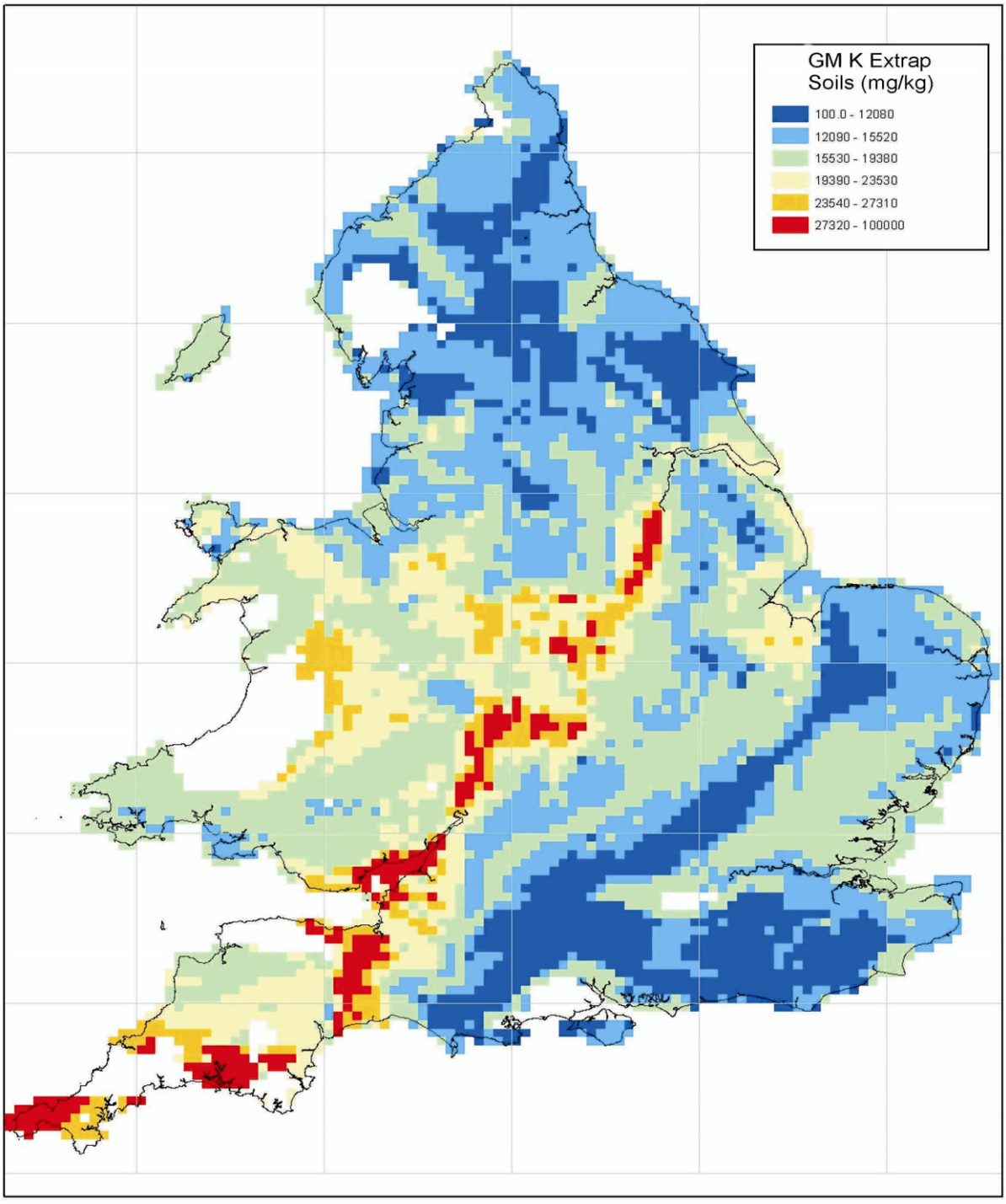

Figure 3.10: Concentrations of K in soils derived by geological extrapolation (mg kg^{-1} dry weight)

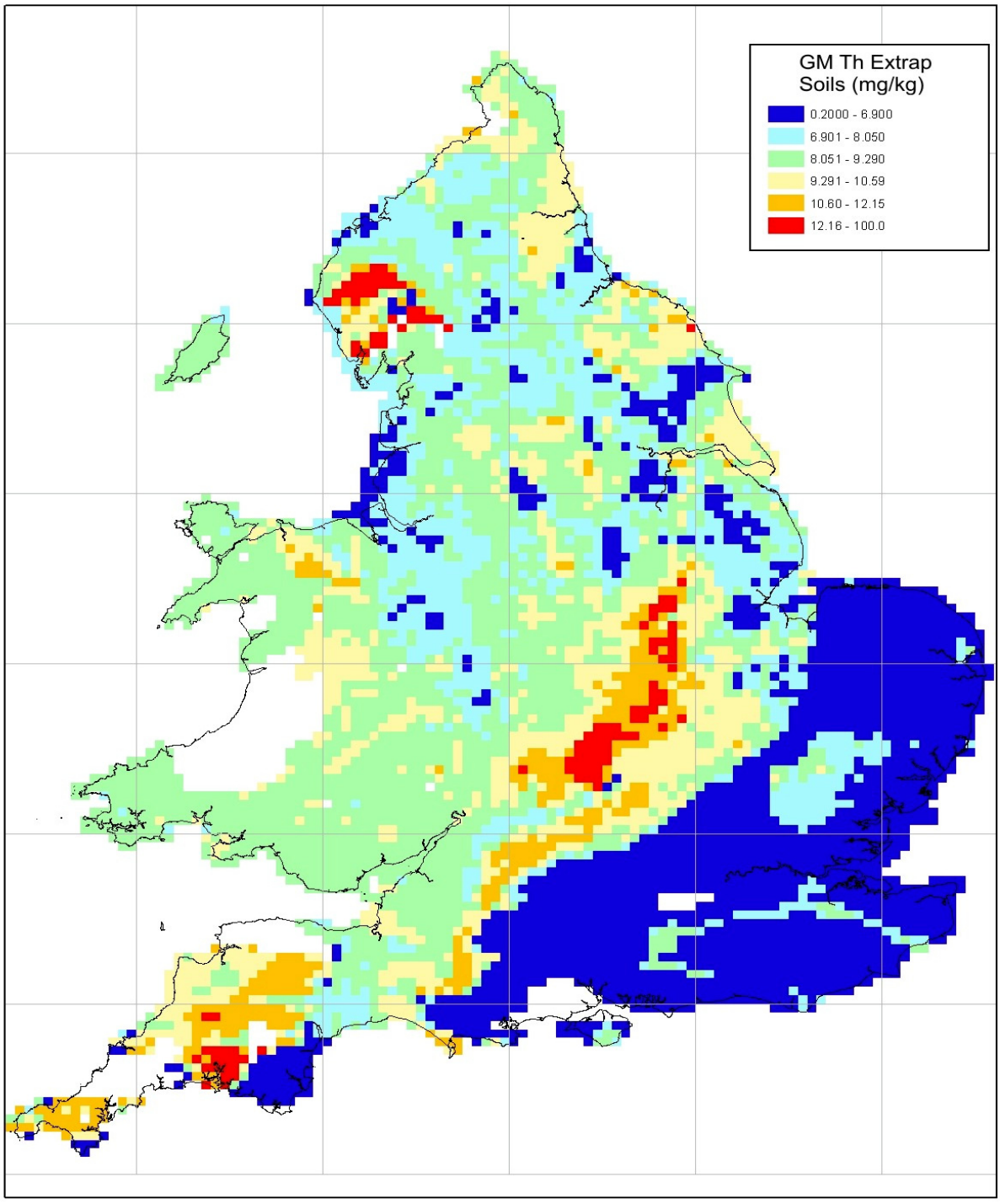

Figure 3.11: Concentrations of Th in soils derived by geological extrapolation (mg kg^{-1} dry weight)

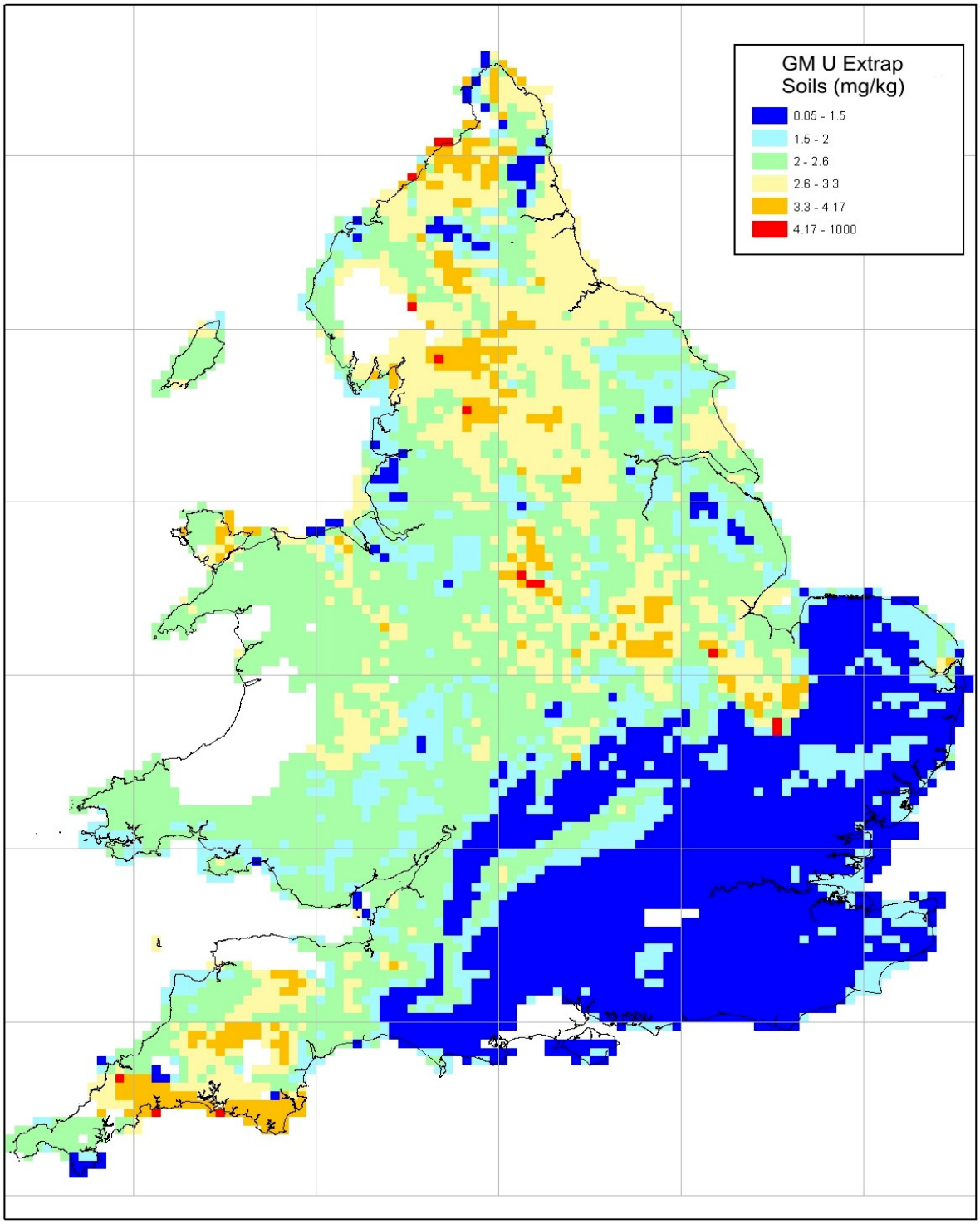

Figure 3.12: Concentrations of U in soils derived by geological extrapolation (mg kg^{-1} dry weight)

3.3.3 Waters

For waters, geometric means were only calculated for 5 x 5 km grid squares based on the actual data points in each square (Figures 3.13 to 3.15). The area used was limited to that covered by the data from ICP analyses, because of the unreliability of earlier results and the problems of trying to level different datasets as described above (Section 3.2).

Class intervals used for the water maps are the 30, 49, 63, 74 and 85th percentile values for the raw data distribution for K; 20, 37, 52, 63 and 78th percentile for U; and 86, 94, 98 and 99th percentile for Th.

The relationship between the water data and geology is much less strong than for stream sediments and soils. For this reason, and because coverage was more limited for waters, it was felt that geological extrapolation could not be justified for surface water data.

3.3.4 Recommendations

For soils and sediments, datasets are provided for both (i) the geometric mean concentrations from measured samples on a 5 x 5 km square basis and (ii) the extrapolated surfaces to extend coverage. It is recommended that where data are available for an area being assessed, these are used rather than the extrapolated values. Note the geometric mean datasets include minimum, maximum and geometric standard deviation values for each square.

More complete geometric mean datasets could be generated once the G-BASE coverage of England and Wales is complete (anticipated by 2021), so that data is provided based on actual values. Total coverage for sediments would require analysis of previously collected samples where U and Th were not analysed. Airborne gamma spectrometry is currently available for parts of Britain but is not extensive enough to materially add to the G-BASE soil data. However, there are plans to extend the coverage in future and consideration could be given to using this data to provide more extensive coverage for soil.

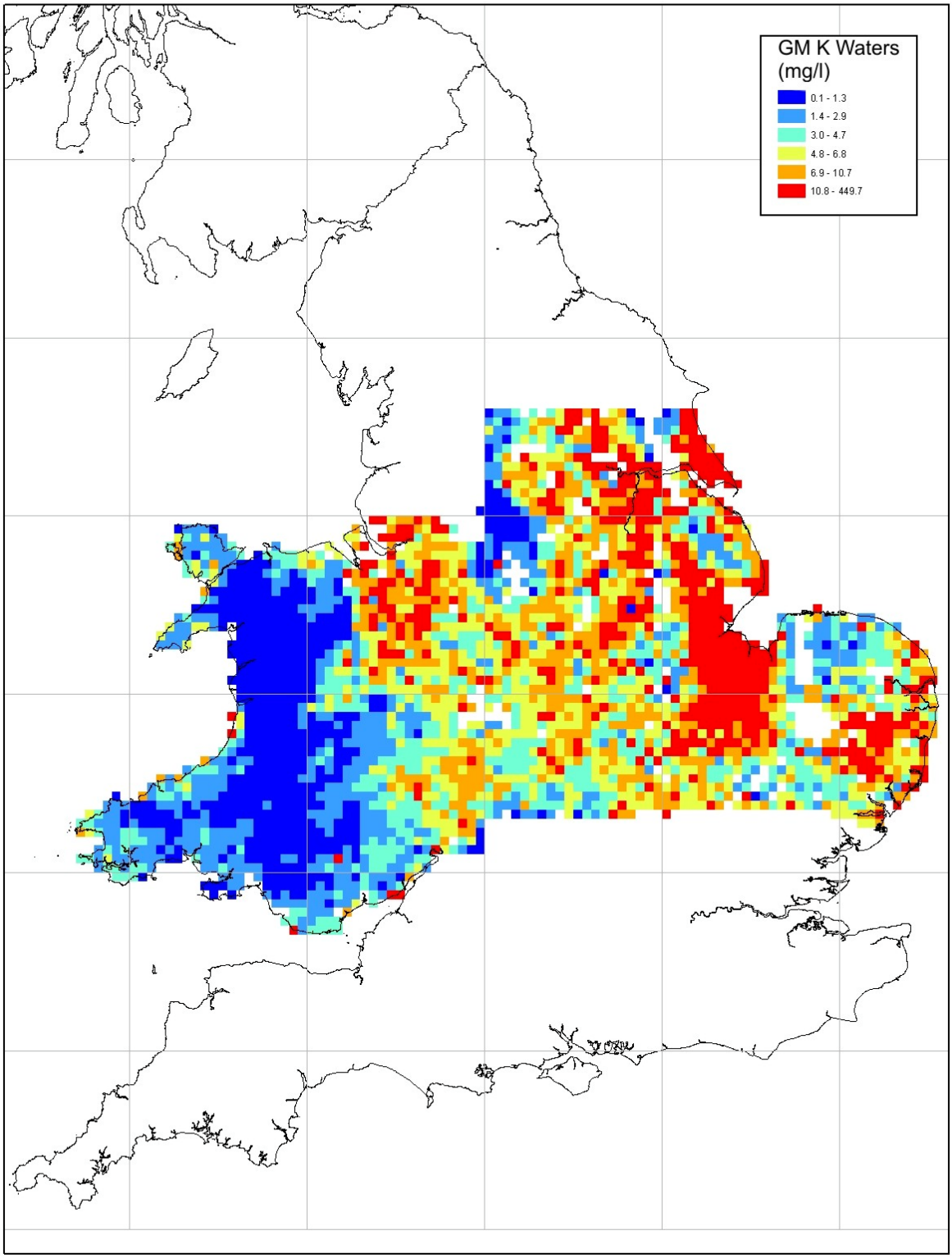

Figure 3.13: Concentrations of K in stream waters presented as geometric mean of measurements within 5 x 5 km squares (µg l^{-1})

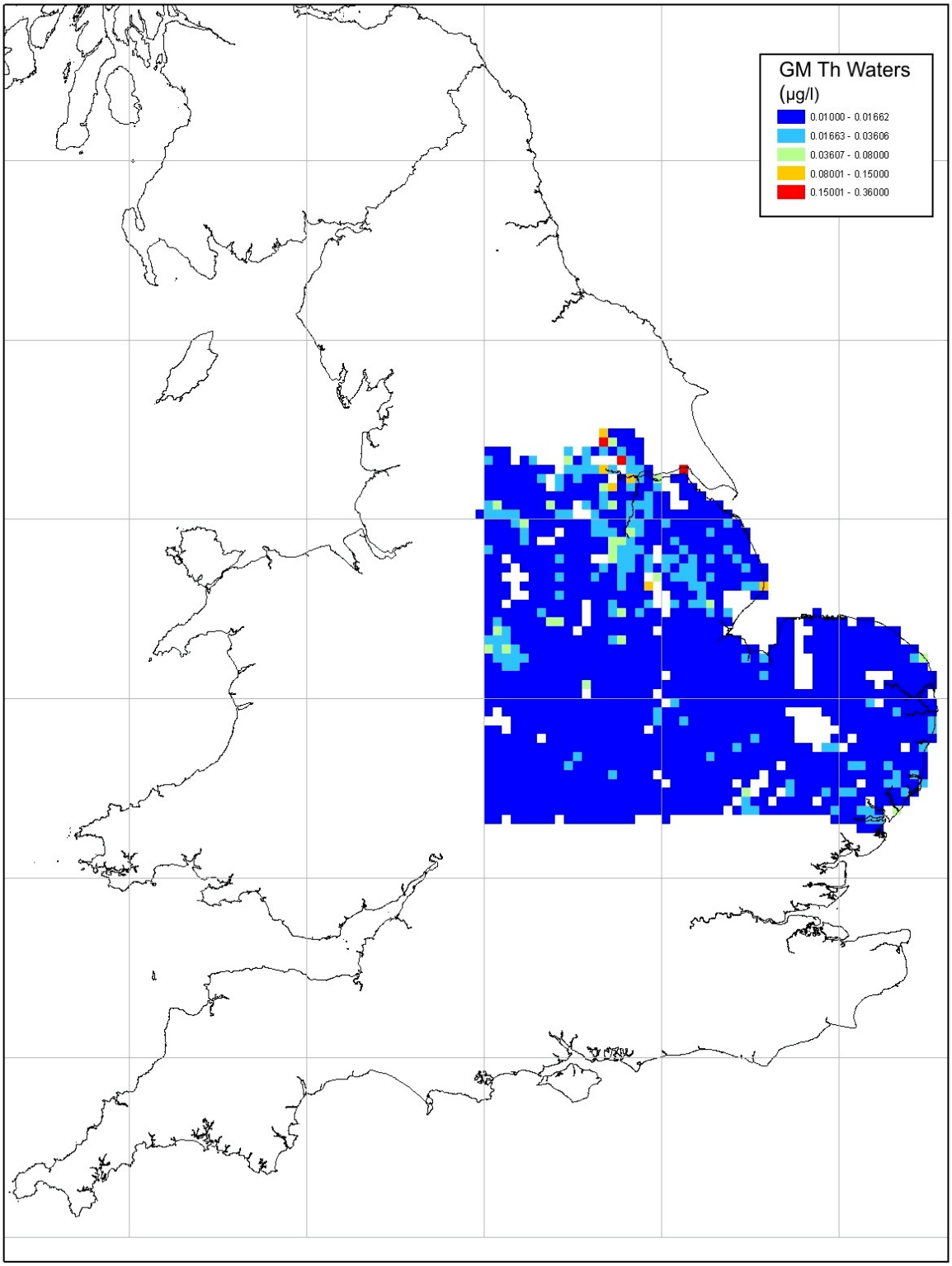

Figure 3.14: Concentrations of Th in stream waters presented as geometric mean of measurements within 5 x 5 km squares (µg l^{-1})

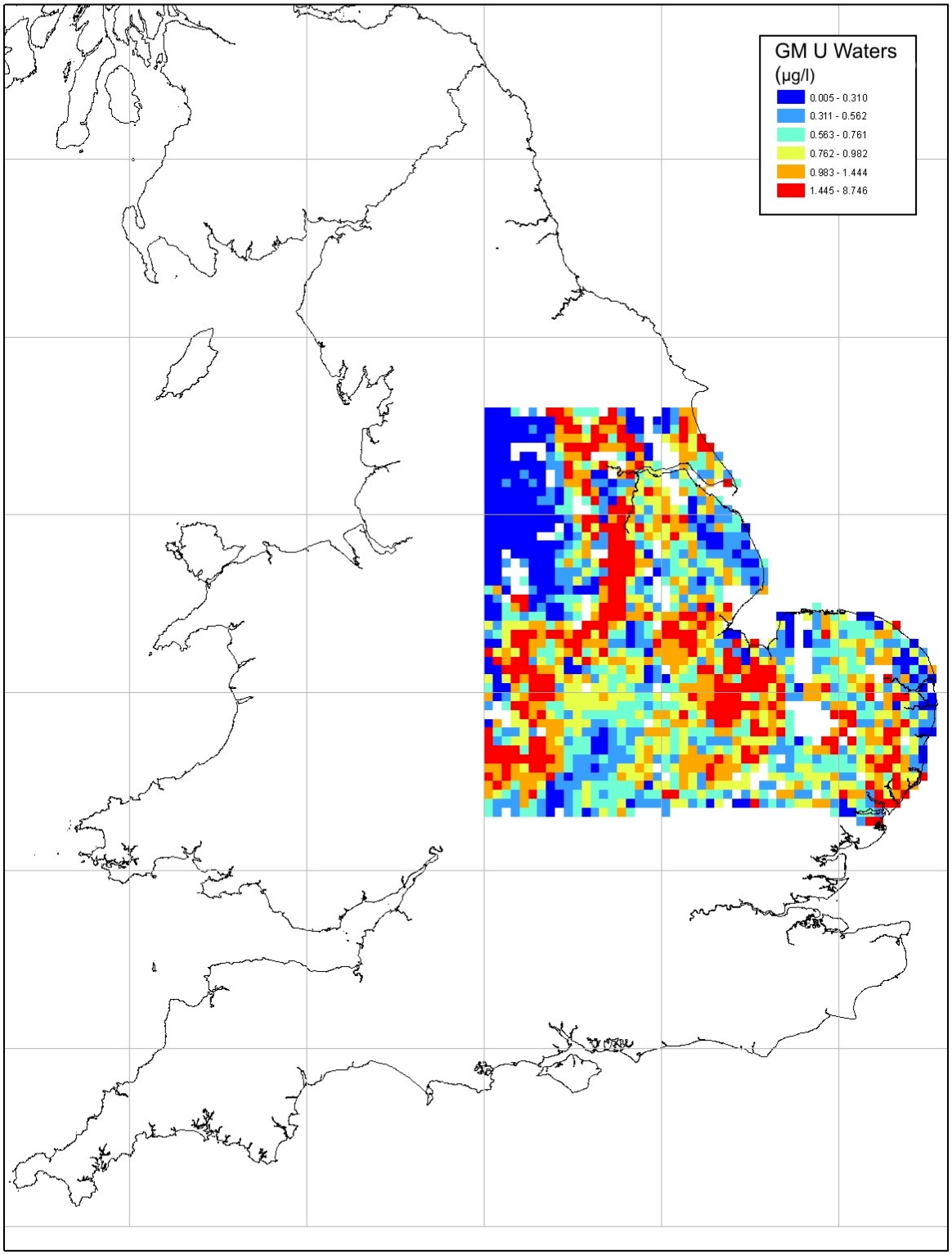

Figure 3.15: Concentrations of U in stream waters presented as geometric mean of measurements within 5 x 5 km squares (µg l^{-1})

3.4 Use of total element data in radiological assessments

The specific activities of ^{40}K, ^{232}Th and ^{238}U are 31.6 Bq g^{-1} K, 4.1 Bq mg^{-1} Th and 12.2 Bq mg^{-1} U respectively. These can be used to estimate the activity concentrations of the three radionuclides from total stable element concentrations of environmental media. In the case of ^{238}U series radionuclides, it can be assumed that activity concentrations of ^{234}U, and the intervening decay products (see Table 1.1), are in equilibrium with those of ^{238}U in soils, sediments and water. Thereafter, the different chemical properties and environmental behaviours of the different elements mean that assumptions of equilibrium may not be valid. Whilst in non-oxidising conditions the activity concentrations of ^{226}Ra may be assumed to be similar to those of ^{238}U, in oxidising conditions ^{238}U is potentially more mobile and this assumption may not be valid. However, all ^{232}Th series radionuclides are likely to be in approximate equilibrium in the environment.

Estimated activity concentrations in water, sediments and soils are summarised by Environment Agency region in Table 3.2; results are not shown where data were available for less than 50 per cent of a region. For this example, the extrapolated datasets for Th and U in sediments and all three elements in soils were used (as they provided more complete coverage). Concentrations of ^{234}U and ^{234}Th were assumed to be equal to those presented for ^{238}U. Activity concentrations of ^{232}Th were assumed to be an approximation of all other radionuclides in its decay series. The activity concentrations of ^{40}K, ^{232}Th and ^{238}U estimated using the approaches outlined above resulted in values within those expected from global reviews cited by Brown *et al.* (2004) and Gómez-Ros *et al.* (2004). An exception was the comparatively high maximum ^{40}K value for stream waters in the Anglian region.

3.5 Other data sources

Gamma air kerma measurements can provide an approximate estimate of external gamma exposure of biota (Beresford and Howard, 2005). Results of air kerma measurements (at one m above surface) from 174 sites throughout the UK (Copplestone *et al.,* in press) are included in the databases supplied. These data have a mean value of 0.75 µGy d^{-1}, ranging from 0.15 to 2.6 µGy d^{-1}. Air kerma rates, measured at one m above ground surface, are also available for 84 tide-washed pastures from measurements along the eastern Irish seaboard of England and Wales by Horrill *et al.* (1992); a mean value of 3.2 µGy d^{-1} (range 1.9 to 6.0 µGy d^{-1}) is estimated from these data. Note that the sites with the highest air kerma rates from the second survey are in Cumbria and North Lancashire and marine discharges from the Sellafield reprocessing plant may make a significant contribution.

Potassium-40 measurements for sediment-soil samples (nine cm deep) taken from the survey points of Horrill *et al.* (1992) are also provided, together with some ^{232}Th and ^{238}U series radionuclides measured by gamma analyses when available from the original analytical reporting sheets. However, the ^{238}U and ^{232}Th series radionuclide results should be treated with caution, as samples were not prepared and analysed for natural series radionuclides (by sealing and allowing for equilibrium to be established) and results are often close to detection limits.

Table 3.2: Summary of estimated ^{40}K, ^{238}U and ^{232}Th in stream sediments, waters and soils summarised by Environment Agency region (based upon the extrapolated datasets which have more complete coverage)

Environment Agency region	Sediment (Bq kg^{-1}DW)		Water (Bq l^{-1})		Soil (Bq kg^{-1} DW)	
	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range
^{40}K						
Anglian	484±137	28–849	(3.56±6.45)x10^{-1}	1.45x10^{-2}–14.2	487±81	267–753
Wales	666±120	230–1,142	(4.68±5.53)x10^{-2}	(0.19–56.7)x10^{-2}	602±87	395–914
Midlands*	647±142	156–1,114	(1.99±1.09)x10^{-1}	(0.16–84.9)x10^{-2}	651±131	345–1,049
North East	569±148	227–1,323			415±71	223–670
North West	594±107	339–1,012			484±89	317–852
South West	621±219	163–1,160			589±207	207–1,159
Southern	459±160	80–908			364±98	94–596
Thames	510±148	69–998			417±110	96–580
^{238}U						
Anglian	26±7	8–57	(1.2±0.83)x10^{-2}	(0.15–6.86)x10^{-2}	22±8	5–59
Wales	27±5	12–44			28±3	19–44
Midlands*	31±7	13–84	(1.51±1.45)x10^{-2}	6.69x10^{-5}–0.11	29±6	7–57
North East	39±9	19–140			32±7	8–51
North West	34±10	19–136			30±6	12–54
South West	35±36	7–253			25±12	2–76
Southern	16±5	8–29			13±5	2–24
Thames	17±7	6–29			13±7	2–33
^{232}Th						
Anglian	33±7	13–58	(4.88±2.20)x10^{-5}	(4.07–36.4)x10^{-5}	29±10	5-69
Wales	37±4	21–46			37±2	30-44
Midlands*	38±5	18–80	(5.37±3.26)x10^{-5}	(4.07–29.1)x10^{-5}	36±4	22-62
North East	41±5	21–100			34±4	16-48
North West	37±4	27–59			36±9	14-75
South West	38±11	16–68			34±11	4-66
Southern	29±5	16–44			21±6	13-36
Thames	33±7	15–58			27±11	12-86

*Approximately 50 per cent of the Midlands Region has extrapolated data for Th and U concentrations in stream waters.

4 Exposure of biota to naturally occurring radionuclides

4.1 Internal exposure estimated from measured biota activity concentrations

The ERICA project has derived unweighted dose conversion coefficients (DCC) specifically for the proposed ICRP RAP geometries (see Table 2.1). The DCC relate whole-body activity concentrations in biota, or media, to absorbed dose (μGy h^{-1} per Bq kg^{-1} fresh weight). For reference, the ERICA DCC values for RAPs can be found in Appendix B.1, where daughter products with a physical half-life of less than 10 days are assumed to be in secular equilibrium with the parent radionuclide (for example, the DCC for ^{228}Th includes contributions from ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po).

To estimate unweighted internal absorbed dose rates, the ERICA DCC values were applied to the mean activity concentrations for each RAP and radionuclide as presented in Table 2.4. The estimated absorbed dose rates are given in Table 4.1. For all organisms, unweighted absorbed doses from ^{40}K predominated; absorbed doses from most other radionuclides for which there were data rarely exceeded 10 per cent of the ^{40}K estimate. Dose rates from the predominantly alpha-emitting radionuclides (^{210}Po, ^{230}Th, ^{232}Th, ^{234}U and ^{238}U) would increase if a radiation weighting factor were applied. Not enough data were available to estimate internal absorbed doses due to ^{228}Th, ^{234}Th and ^{228}Ra for any RAPs, nor for ^{210}Pb, ^{210}Po, ^{230}Th or ^{226}Ra for some RAPs.

Table 4.1: Estimated unweighted absorbed internal dose rates for different RAPs in the UK, calculated using mean measured activity concentrations (Table 2.4) and dose conversion coefficients from the ERICA project (Appendix B.1)

RAP	Unweighted absorbed internal dose (μGy h^{-1})							
	^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{230}Th	^{232}Th	^{234}U	^{238}U
Wild grass	7.9×10^{-2}	8.0×10^{-3}	8.6×10^{-4}	4.9×10^{-3}	7.3×10^{-5}	3.2×10^{-4}	1.1×10^{-3}	9.6×10^{-4}
Pine tree	1.3×10^{-2}	5.9×10^{-4}	2.5×10^{-5}	4.4×10^{-5}	1.8×10^{-5}	1.3×10^{-5}	2.1×10^{-5}	1.8×10^{-5}
Brown seaweed	3.5×10^{-2}	6.5×10^{-3}	9.8×10^{-5}	n/a	2.1×10^{-4}	9.9×10^{-5}	4.8×10^{-3}	2.8×10^{-3}
Earthworm	1.3×10^{-2}	n/a	n/a	n/a	n/a	1.6×10^{-3}	2.1×10^{-3}	1.9×10^{-3}
Bee	6.0×10^{-2}	n/a	n/a	n/a	n/a	7.1×10^{-5}	4.2×10^{-5}	3.6×10^{-5}
Duck	3.7×10^{-2}	n/a	n/a	n/a	2.7×10^{-5}	1.6×10^{-4}	1.1×10^{-4}	9.8×10^{-5}
Rat	3.4×10^{-2}	6.2×10^{-4}	5.5×10^{-5}	3.1×10^{-4}	7.8×10^{-6}	1.0×10^{-5}	1.5×10^{-5}	1.1×10^{-5}
Deer	3.9×10^{-2}	6.2×10^{-4}	5.8×10^{-5}	3.3×10^{-4}	7.8×10^{-6}	1.0×10^{-5}	1.5×10^{-5}	1.1×10^{-5}
Trout	8.0×10^{-2}	n/a	n/a	n/a	n/a	4.1×10^{-5}	2.5×10^{-5}	2.2×10^{-5}

*Rat and deer both assume mammal activity concentrations from Table 2.4.

4.2 External exposure estimated from media activity concentrations

Mean concentrations of ^{40}K, ^{232}Th and ^{238}U by Environment Agency Region shown in Table 3.2 were used to estimate unweighted external absorbed dose rates to RAPs using DCC for organisms on soil, in soil or in water/sediment (see Appendix B.1). It was assumed that:

- rats spend 50 per cent of their time underground;
- trout are in the water column (receiving no exposure from sediments);
- earthworms are permanently within soil;
- frogs spend 50 per cent of their time on the soil surface and 50 per cent within freshwater sediments;
- all other RAPs are on the soil surface only.

Activity concentrations of ^{234}Th and ^{228}Ra were assumed to be equal to those of ^{238}U and ^{232}Th respectively. External dose rates due to radionuclides below ^{234}U in the ^{238}U series were not estimated. Estimated external absorbed dose rates are presented in Table 4.2.

For trout, ^{40}K dominated the absorbed dose rates of the radionuclides for which we were able to make estimates. Whilst ^{40}K was the single largest contributor to the total dose rates estimated for all other RAPs, dose rates estimated for both ^{228}Th and ^{228}Ra were approximately within a factor of two of the ^{40}K contribution. Absorbed dose rates for all other radionuclides were at least two orders of magnitude lower than those estimated for ^{40}K. For the rat, exposure in the soil (assumed to be 50 per cent of the time) dominated the total external absorbed dose rates. Similarly, exposure within sediments (assumed to be the habitat for 50 per cent of the time) dominated the external dose rate for the frog. For terrestrial RAPs, estimated ^{40}K external absorbed dose rates were comparable to internal dose rates predicted using measured biota activity concentrations (compare Tables 4.1 and 4.2). All other external exposure rates were lower (often by more than an order of magnitude) than absorbed dose rates estimated for internal exposure.

The summaries presented in Table 4.2 are based on Environment Agency region mean estimates. The range in values is therefore less than would be obtained from individual 5 x 5 km square data. External dose rates were estimated by applying the DCC to dry weight soil and sediment activity concentrations. However, the DCC are defined on a fresh weight soil and sediment basis, and this will result in an overestimate of external dose rates, although this discrepancy is unlikely to be large.

Table 4.2: Estimated external absorbed doses for terrestrial and freshwater RAPs in England and Wales

RAP		Unweighted absorbed dose (μGy h^{-1})						
		^{40}K	^{228}Ra	^{228}Th	^{232}Th	^{234}Th	^{234}U	^{238}U
Wild grass	Mean	$1.5x10^{-2}$	$6.0x10^{-3}$	$8.9x10^{-3}$	$3.5x10^{-6}$	$1.1x10^{-4}$	$3.3x10^{-6}$	$2.4x10^{-6}$
	Min	$1.1x10^{-2}$	$4.0x10^{-3}$	$5.8x10^{-3}$	$2.3x10^{-6}$	$6.0x10^{-5}$	$1.8x10^{-6}$	$1.3x10^{-6}$
	Max	$1.9x10^{-2}$	$6.9x10^{-3}$	$1.0x10^{-2}$	$4.0x10^{-6}$	$1.5x10^{-4}$	$4.5x10^{-6}$	$3.2x10^{-6}$
Pine tree	Mean	$1.2x10^{-2}$	$4.8x10^{-3}$	$7.3x10^{-3}$	$6.7x10^{-7}$	$8.8x10^{-5}$	$4.3x10^{-7}$	$1.7x10^{-7}$
	Min	$8.7x10^{-3}$	$3.1x10^{-3}$	$4.8x10^{-3}$	$4.4x10^{-7}$	$4.7x10^{-5}$	$2.3x10^{-7}$	$8.8x10^{-8}$
	Max	$1.6x10^{-2}$	$5.5x10^{-3}$	$8.4x10^{-3}$	$7.7x10^{-7}$	$1.2x10^{-4}$	$5.8x10^{-7}$	$2.2x10^{-7}$
Earthworm	Mean	$4.0x10^{-2}$	$1.6x10^{-2}$	$2.5x10^{-2}$	$4.4x10^{-6}$	$2.6x10^{-4}$	$4.1x10^{-6}$	$2.9x10^{-6}$
	Min	$2.9x10^{-2}$	$1.0x10^{-2}$	$1.6x10^{-2}$	$2.9x10^{-6}$	$1.4x10^{-4}$	$2.2x10^{-6}$	$1.5x10^{-6}$
	Max	$5.2x10^{-2}$	$1.8x10^{-2}$	$2.9x10^{-2}$	$5.1x10^{-6}$	$3.5x10^{-4}$	$5.5x10^{-6}$	$3.9x10^{-6}$
Bee	Mean	$1.5x10^{-2}$	$1.1x10^{-2}$	$9.2x10^{-3}$	$1.4x10^{-6}$	$1.1x10^{-4}$	$1.7x10^{-6}$	$1.2x10^{-6}$
	Min	$1.1x10^{-2}$	$7.3x10^{-3}$	$6.0x10^{-3}$	$9.2x10^{-7}$	$5.9x10^{-5}$	$9.1x10^{-6}$	$6.4x10^{-7}$
	Max	$2.0x10^{-2}$	$1.3x10^{-2}$	$1.1x10^{-2}$	$1.6x10^{-6}$	$1.5x10^{-4}$	$2.3x10^{-6}$	$1.6x10^{-6}$
Duck	Mean	$1.4x10^{-2}$	$5.7x10^{-3}$	$8.6x10^{-3}$	$1.2x10^{-6}$	$1.0x10^{-4}$	$1.5x10^{-6}$	$1.0x10^{-6}$
	Min	$1.0x10^{-2}$	$3.8x10^{-3}$	$5.6x10^{-3}$	$8.1x10^{-7}$	$5.5x10^{-5}$	$8.1x10^{-7}$	$5.5x10^{-7}$
	Max	$1.8x10^{-2}$	$6.6x10^{-3}$	$9.9x10^{-3}$	$1.4x10^{-6}$	$1.4x10^{-4}$	$2.0x10^{-6}$	$1.4x10^{-6}$
Rat	Mean	$2.7x10^{-2}$	$1.0x10^{-2}$	$1.6x10^{-2}$	$2.6x10^{-6}$	$1.3x10^{-4}$	$2.6x10^{-6}$	$1.8x10^{-6}$
	Min	$1.9x10^{-2}$	$6.9x10^{-3}$	$1.1x10^{-2}$	$1.7x10^{-6}$	$7.1x10^{-5}$	$1.4x10^{-6}$	$9.4x10^{-7}$
	Max	$3.4x10^{-2}$	$1.2x10^{-2}$	$1.9x10^{-2}$	$3.0x10^{-6}$	$1.8x10^{-4}$	$3.5x10^{-6}$	$2.4x10^{-6}$
Deer	Mean	$8.0x10^{-3}$	$3.1x10^{-3}$	$5.1x10^{-3}$	$4.1x10^{-7}$	$5.3x10^{-5}$	$4.1x10^{-7}$	$2.4x10^{-7}$
	Min	$5.8x10^{-3}$	$2.0x10^{-3}$	$3.3x10^{-3}$	$2.7x10^{-7}$	$2.8x10^{-5}$	$2.2x10^{-7}$	$1.3x10^{-7}$
	Max	$1.0x10^{-2}$	$3.5x10^{-3}$	$5.8x10^{-3}$	$4.7x10^{-7}$	$7.1x10^{-5}$	$5.5x10^{-7}$	$3.2x10^{-7}$
Frog	Mean	$3.9x10^{-2}$	$1.3x10^{-2}$	$2.1x10^{-2}$	$5.9x10^{-6}$	$9.5x10^{-4}$	$6.1x10^{-6}$	$4.4x10^{-6}$
	Min	$3.1x10^{-2}$	$9.9x10^{-3}$	$1.6x10^{-2}$	$4.6x10^{-6}$	$5.4x10^{-4}$	$3.4x10^{-6}$	$2.5x10^{-6}$
	Max	$4.6x10^{-2}$	$1.5x10^{-2}$	$2.4x10^{-2}$	$6.7x10^{-6}$	$1.3x10^{-3}$	$8.4x10^{-6}$	$6.1x10^{-6}$
Trout	Mean	$1.8x10^{-5}$	$2.6x10^{-8}$	$4.3x10^{-8}$	$7.7x10^{-12}$	$4.1x10^{-7}$	$2.1x10^{-9}$	$2.0x10^{-9}$
	Min	$4.1x10^{-6}$	$2.4x10^{-8}$	$4.1x10^{-8}$	$7.3x10^{-12}$	$3.6x10^{-7}$	$1.9x10^{-9}$	$1.7x10^{-9}$
	Max	$3.1x10^{-5}$	$2.7x10^{-8}$	$4.5x10^{-8}$	$8.1x10^{-12}$	$4.7x10^{-7}$	$2.4x10^{-9}$	$2.3x10^{-9}$

*Mean of all Environment Agency regions; +Minimum and maximum Environment Agency region

4.3 Prediction of internal activity concentrations from media activity concentrations

Media concentrations can also be used to predict the activity concentrations in biota using recommended concentration ratios (defined as the ratio between the fresh weight whole-body activity concentration in biota (Bq kg^{-1}) to the activity concentration of water or dry soil (Bq kg^{-1})). Concentration ratios for Th and U isotopes as derived for organisms similar to the ICRP RAPs by the ERICA project are presented in Appendix B.2; no concentration ratio values are recommended for ^{40}K. The concentration ratios were applied to the mean concentrations of ^{232}Th and ^{238}U and predicted whole-body activity concentrations are shown in Table 4.3. Predicted concentrations are compared to measurements (from Table 2.4) in Figures 4.1 and 4.2. The measured and predicted ranges overlap for all RAPs giving some confidence

both in the use of the concentration ratio values and the derived media concentration datasets.

Table 4.3: Predicted whole-body biota ^{232}Th and ^{238}U activity concentrations for England and Wales

RAP	Activity concentration (Bq kg^{-1} FW)					
	^{232}Th			^{238}U		
	Mean^{+}	Minimum*	Maximum*	Mean^{+}	Minimum*	Maximum*
Wild grass	1.39	0.18	3.75	0.35	2.3×10^{-2}	1.11
Pine tree	3.4×10^{-2}	4.4×10^{-3}	9.3×10^{-2}	0.16	1.1×10^{-2}	0.52
Earthworm	0.28	3.6×10^{-2}	0.76	0.21	1.4×10^{-2}	0.67
Duck**	1.2×10^{-2}	1.6×10^{-3}	3.3×10^{-2}	1.2×10^{-2}	7.9×10^{-4}	3.8×10^{-2}
Mammal	3.9×10^{-3}	5.0×10^{-4}	1.0×10^{-2}	2.5×10^{-3}	1.7×10^{-4}	8.1×10^{-3}
Trout	5.6×10^{-3}	4.5×10^{-3}	4.0×10^{-2}	0.40	2.0×10^{-3}	3.2

$^{+}$Mean value is average of Environment Agency region means; *Minimum and maximum are range of predictions across all of 5 x 5 km squares in England and Wales; **Whole-body activity concentrations estimated from soil activity concentrations.

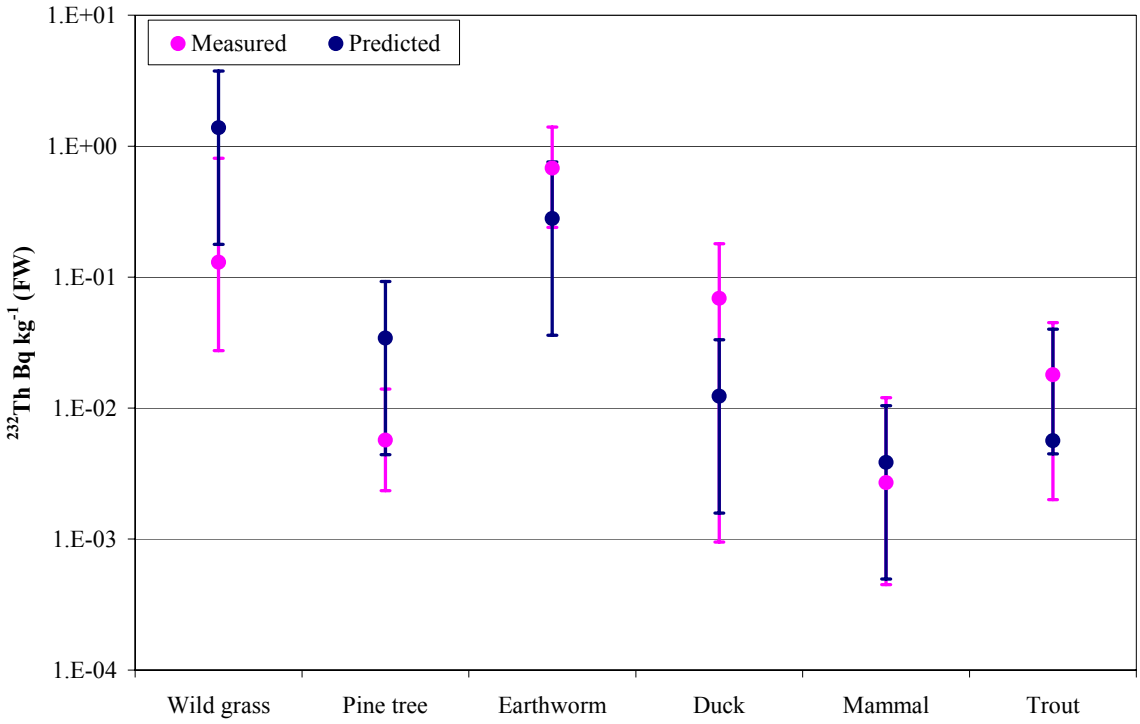

Figure 4.1: Comparison of measured and predicted ^{232}Th biota activity concentrations

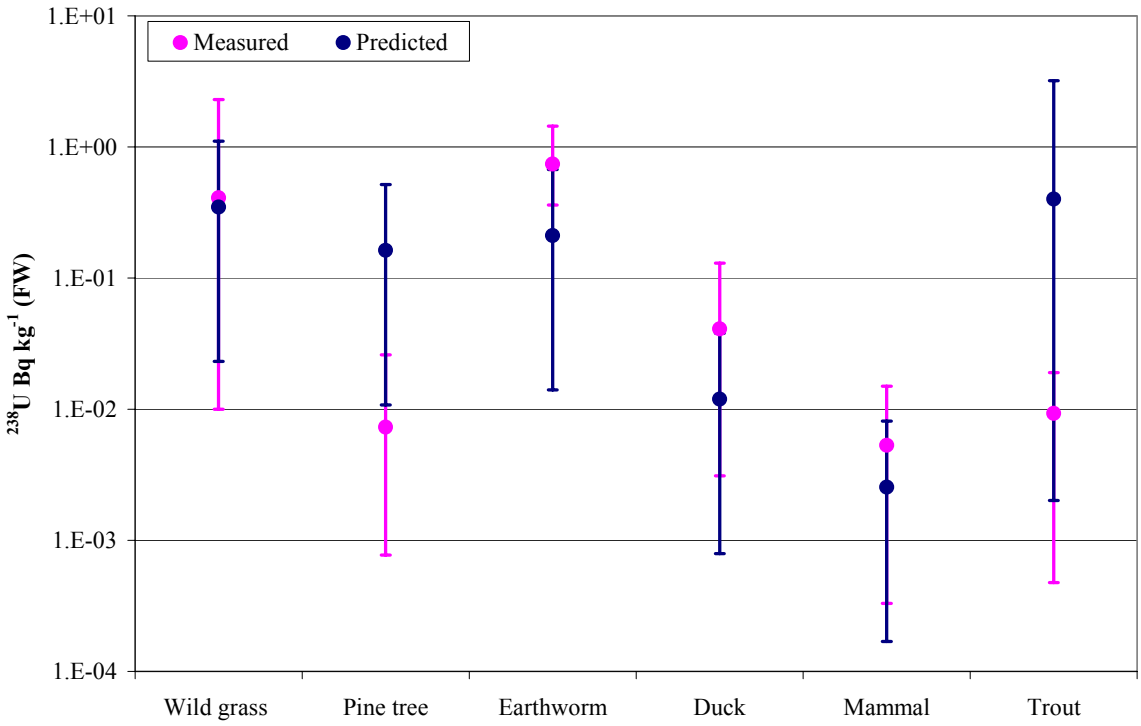

Figure 4.2: Comparison of measured and predicted ^{238}U biota activity concentrations

5 Conclusions and recommendations

The datasets of concentrations of naturally occurring radionuclides in biota and media will help to significantly improve assessments of the background exposure of wild animal and plant species in England and Wales.

To derive data for the RAPs, we compiled values for similar organism types (such as any flying insect data for reference bee) as there were relatively few, if any, data specific to the proposed RAP categories as defined by the ICRP. Whilst there were some data for ^{40}K, ^{232}Th, ^{234}U and ^{238}U for all the organism types considered, data for other naturally occurring radionuclides (^{210}Po, ^{210}Pb, ^{226}Ra, ^{228}Ra) were lacking for many RAPs. The media concentration datasets derived here, together with concentration ratios, could be used to derive some of these missing values.

Average values for 5 x 5 km grid squares were calculated from G-BASE stream sediment, soil and water datasets. As the G-BASE coverage is currently incomplete, it was necessary to use relationships with bedrock and superficial geology to extrapolate stream sediment U and Th data and soil K, U and Th data over all of England and Wales. Complete coverage was achieved for K in stream sediments by combining G-BASE and Wolfson atlas data. More complete geometric mean datasets could be generated once the G-BASE coverage of England and Wales is complete (anticipated by 2021), so that data are based on actual values. Total coverage for sediments would require analysis of previously collected samples where U and Th were not analysed. Other datasets, such as airborne gamma spectrometry, could also be incorporated if more widespread coverage becomes available. Airborne gamma spectrometry is currently available for parts of Britain, but is not extensive enough to materially add to the G-BASE soil data. However, there are plans to extend the coverage in future and consideration could be given to using this data to provide more extensive coverage for soil. Coverage for K, U and Th in stream waters is much more limited and national coverage would be difficult to achieve without extensive re-sampling.

For soils and sediments, datasets are provided for both (i) geometric mean concentrations from measured samples on a 5 x 5 km square basis and (ii) extrapolated surfaces to extend coverage. It is recommended that where data are available for an area being assessed, these are used rather than the extrapolated values. Note that the geometric mean datasets include minimum, maximum and geometric standard deviation values for each square. In the case of Th in stream sediments, extrapolated values may be significant overestimates (see Figure C.4 in Appendix C.1).

Total K, U and Th data from G-BASE were used to estimate ^{40}K, ^{238}U and ^{232}Th media activity concentrations and this generated values comparable to ranges reported in global surveys. For ^{232}Th series radionuclides, equilibrium was assumed with ^{232}Th to derive activity concentrations for the complete series. For ^{238}U series radionuclides, only ^{234}Th and ^{234}U were assumed to be in equilibrium with ^{238}U. Consequently, activity concentrations in media could not be derived for the remaining radionuclides in this series, including ^{226}Ra, ^{210}Po and ^{210}Pb. Some values for England and Wales for these radionuclides in media could be collated from sources such as the RIFE reports.

Acknowledgements

The authors would like to thank: Lee Walker (CEH Monks Wood), Phil Gould (Rothamsted Research), Ian Winfield (CEH Lancaster) and Chris Reading (CEH Dorset) for provision of archived samples; Sophie Everett (CEH Dorset) for sample collections in South West England; Jenny Cook, Kate Breward and Tom Barlow (BGS Keyworth) for conducting total Th and U analyses; and the ERICA consortium for providing DCC and concentration ratio values and Imperial College of Science and Technology for permission to use some of the Wolfson Geochemical Atlas data under licence.

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6 Glossary of terms

Absorbed dose	Quantity of energy imparted by ionizing radiation to unit mass of matter such as tissue. Unit gray, symbol Gy. 1 Gy = 1 joule per kilogram.
Becquerel (Bq)	The International System of Units (SI) definition of activity. 1 Bq = 1 disintegration per second.
G-BASE	Geochemical surveys of the UK conducted by British Geological Survey for over 35 years. See http://www.bgs.ac.uk/gbase/home.html
Gamma air kerma	Kerma is the kinetic energy released in material measured in Gy. Kerma can be quoted for any specified material at a point in free space or in an absorbing medium. Gamma air kerma is the exposure measured in air (in the case of values quoted here, 1 m above the soil surface), which is in effect the absorbed dose measured in air.
Reference animals and plants (RAPs)	A reference animal or plant (as proposed by the ICRP) is a hypothetical entity with the assumed basic biological characteristics of a particular type of animal or plant, as described to the generality of the taxonomic level of Family with defined anatomical, physiological, and life-history properties, that can be used for the purposes of relating exposure to radiation dose and relating dose to different categories of effect, for that type of living organism.
Natura 2000 site	The European network of protected sites established under the Birds Directive and the Habitats Directive.

7 List of abbreviations

BGS: British Geological Survey

CEH: Centre for Ecology and Hydrology

DCC: Dose conversion coefficient

DNA: Delayed neutron activation

DR-OES: Direct-reading optical emission spectrometry

DW: Dry weight

ERICA: Environmental risk from ionising contaminants: assessment and management (EC 6th framework project see http://www.erica-project.org/)

EURATOM: European Atomic Energy Community

FASSET: Framework for assessment of environmental impact (EC 5th framework project)

FP5: Fifth Framework Programme of the European Commission

FP6: Sixth Framework Programme of the European Commission

FW: Fresh weight

GIS: Geographical information system

HERALD: Highly enriched reactor Aldermaston

ICI: Imperial Chemical Industries

ICP-AES: Inductively-coupled plasma atomic emission spectrometry

ICP-MS: Inductively-coupled plasma mass spectrometry

ICRP: International Commission on Radiation Protection

LF: Laser Fluorimetry

RAPs: Reference animals and plants (as proposed by ICRP, 2005)

RIFE: *Radioactivity in Food and the Environment* (annual report series published by UK agencies)

UK: United Kingdom

XRF: X-ray fluorescence spectrometry

Appendix A.1: Grey literature sources reviewed for available data

Grey literature sources (in addition to RIFE reports) from which data were used

Environment Agency, 2001. *Radioactivity in the Environment*, Report for 1999. Environment Agency: Bristol.

Environment Agency, 2001. *Radioactivity in the Environment*, Report for 2000. Environment Agency: Bristol.

Environment Agency, 2003. *Radioactivity in the Environment*, Report for 2001. Environment Agency: Bristol.

Fulker, M.J., McKay, K., John, C. and Jackson, D., 1995. *Radioactivity in terrestrial wild foods near Sellafield*. Westlakes Research Ltd.: Moor Row.

Green, N., Hammond, D.J., Davidson, M.F., Wilkins, B.T. and Williams, B., 2002. *The radiological impact of naturally occurring radionuclides in foods from the wild*. NRPB-W30. NRPB: Didcot.

Smith, K.R., Crockett, G.M., Oatway, W.B., Harvey, M.P., Penfold, J.S.S. and Mobbs, S.F., 2001. *Radiological impact on the UK population of industries which use or produce materials containing enhanced levels of naturally occurring radionuclides*. NRPB-R327. NRPB: Didcot.

Grey literature sources reviewed which did not yield useful data

The Royal Society, 2002. *The health hazards of depleted uranium munitions*. (Part 1, 2 and summary). Available from: www.royalsoc.ac.uk

Ewers, L.W., Ham, G.J. and Wilkins, B.T., 2003. *Review of the Transfer of Naturally Occurring Radionuclides to Terrestrial Plants and Domestic Animals*. NRPB-W49. NRPB:Didcot.

Watson, S.J., Jones, A.L., Oatway, W.B. and Hughes, J.S., 2005. *Ionising Radiation Exposure of the UK Population: 2005 Review*. ISBN 0-85951-558-3. HPA-RPD-001. Didcot: Health protection agency.

Harvey, M.P. and Simmonds, J.R., 2002. *Generalised Derived Constraints for Radioisotopes of Polonium, Lead, Radium and Uranium*. Volume 13, No. 2. Didcot: NRPB.

Environment Agency, Scottish Environment Protection Agency, Department of Environment, Northern Ireland, National Radiological Protection Board, Food Standards Agency. December 2002. Radioactive Substances Regulation. Authorisation of Discharges of Radioactive Waste to the Environment. Principles for the Assessment of

Prospective Public Doses. Interim Guidance. Available from: http://www.food.gov.uk/safereating/rad_in_food/111902

Green, N., Hammond, D.J. and Wilkins, B.T., 2005. *A Long-term Study of the Transfer of Radionuclides from Soil to Fruit*. ISBN 0-85951-565-6. HPA-RPD-006. Didcot: Health Protection Agency.

Hughes, J.S., 1999. *Ionising Radiation Exposure of the UK Population: 1999 Review*. NRPB-R311. ISBN 0-85951-439-0. Didcot: NRPB.

Appendix A.2: Refereed literature sources reviewed for available data

Beardsley, T., 1983. Windscale 1957 Accident - Polonium Not A Hazard. *Nature,* 305, 351.

Beardsley, T., 1983. Windscale 1957 Accident - Forgotten Polonium Is Traced. *Nature,* 302, 470.

Bellis, D., Mcleod, C.W. and Satake, K., 2001. The potential of elemental and isotopic analysis of tree bark for discriminating sources of airborne lead contamination in the UK. *Journal of Environmental Monitoring,* 3, 194-197.

Abstract: Samples of tree bark, which accumulate airborne material, were collected from seven locations in the UK to provide an indication of the magnitude and source of lead pollution. Measurement of the Pb content and Pb-206/207 stable isotope ratio by inductively-coupled plasma mass spectrometry revealed significant differences between the sites. The concentration of Pb varied over almost four orders of magnitude from 7.2 to 9,600 μg g^{-1}, the maximum values being found near a 'secondary' Pb smelter. The Pb-206/207 isotope ratios varied from 1.108 +/- 0.002 to 1.169 +/- 0.001. The lowest Pb concentrations and highest isotope ratios were detected in bark samples from the Scilly Isles, reflecting the low level of industry and road traffic. In contrast, samples obtained from a city centre (Sheffield) and near a motorway (M1) contained 25 to 46 μg g^{-1} Pb and recorded the lowest Pb-206/207 ratios. Higher concentrations in the vicinity of a coal-fired power station recorded a Pb-206/207 ratio of 1.14, suggesting a significant contribution from fly ash. The relative contribution of lead from petrol (Pb-206/207 = 1.08) and other sources such as coal (Pb-206/207 = 1.18) were thus estimated using mass balance equations. Tree bark near the lead smelter recorded an intermediate Pb-206/207 ratio of 1.13, reflecting the processing of material of mixed origin.

Bellis, D.J., Mcleod, C.W. and Satake, K., 2002. Pb and Pb-206/Pb-207 isotopic analysis of a tree bark pocket near Sheffield, UK, recording historical change in airborne pollution during the 20th Century. *Science of the Total Environment,* 289, 169-176.

Abstract: A section of tree trunk (beech, *Fagus sylvatica*) containing a bark pocket progressively enclosed at the junction of two branches was collected from a semi-rural location near Sheffield, UK. According to the annual growth rings, the bark pocket formed between *circa* 1919 and 1998 (the date of felling). The bark pocket was divided into consecutive samples of differing radial depth (and thus age), that were analysed by ICP mass spectrometry. The Pb concentration varied from 7 to 78 mg kg^{-1} and the Pb-206/Pb-207 isotope ratio from 1.11 to 1.15. In contrast, the current surface bark contained 46 mg kg^{-1} Pb and recorded a Pb-206/Pb-207 ratio of 1.11. The changing elemental and isotopic composition of the bark pocket recorded historical change in the level and sources of airborne Pb pollution. An overall increase in Pb concentration with time was accompanied by a progressive reduction in Pb-206/Pb-207 from *circa* 1935 to 1943. Mass balance calculations indicated that Pb additives in petrol contributed significantly to the rise in concentration, accounting for a maximum of 50 per cent of the total Pb for *circa* 1986 to 1998, but that other sources were generally dominant. The highest Pb concentrations were recorded from *circa* 1951 to 1973, suggesting a high

level of industrial pollution. A reduction in Pb concentration and reversal of the trend in Pb-206/Pb-207 was observed in the current bark.

Bellis, D.J., Satake, K. and Mcleod, C.W., 2004. A comparison of lead isotope ratios in the bark pockets and annual rings of two beech trees collected in Derbyshire and South Yorkshire, UK. *Science of the Total Environment,* 321, 105-113.

Abstract: ICP-MS analysis of the bark pockets and annual rings of two beech (*Fagus sylvatica* L.) trees collected from Longshaw, Derbyshire and Swinton, South Yorkshire in the UK recorded differences in the Pb-206/Pb-207 isotope ratio. In the Longshaw sample, the Pb-206/Pb-207 isotope ratio of the bark pockets (1914 to 1998, 78 to 260 μg g^{-1} Pb) declined from approximately 1.16 to 1.12, whilst the annual rings (1899 to 1998, 0.2 to 2.5 μg g^{-1} Pb) had a Pb-206/ Pb-207 ratio of approximately 1.18. In the Swinton sample, the bark pockets (1919-1998, 7-78 μg g^{-1} Pb) declined from 1.15 to 1.11 and the annual rings (1899-1998, 0.2-0.5 μg g^{-1} Pb) from 1.18 to 1.15. The data implied that the bark pockets accumulated lead directly from the atmosphere through wet and dry deposition, whilst the annual rings accumulated lead from the soil via the roots. The bark pockets recorded a relative decline in the accumulation of lead from indigenous sources, such as lead smelting and coal combustion (1.17-1.19), and an increase in imported sources such as the smelting of Australian ores (1.04) and leaded petrol usage (1.06-1.09). In contrast, the annual rings at Longshaw recorded ratios typical of indigenous lead, whilst the annual rings in Swinton recorded a relatively small decrease in Pb-206/Pb-207 reflecting leaded petrol usage. The decline in Pb-206/Pb-207 of the bark pockets was consistent with the historical decline in Pb-216/Pb-217 of atmospheric lead recorded in peat, lake sediments and archival herbage at other UK locations.

Bradford, W.R., Curtis, E.J.C. and Popplewell, D.S., 1984. Radioactivity in Environmental-Samples Taken in the Sellafield and Ravenglass Areas of West Cumbria, 1977-1982. *Science of the Total Environment,* 35, 267-283.

Camplin, V.C., Baxter, A.J. and Round, G.D., 1996. The radiological impact of discharges of natural radionuclides from a phosphate plant in the United Kingdom. *Environment International*, 22, S259-S270.

Abstract: Discharges of natural radionuclides in liquid effluents from a phosphate ore processing plant in the United Kingdom (UK) have been made to the Irish Sea since 1954. Recently, the plant has begun to import crude phosphoric acid and discharges have been substantially reduced. Environmental monitoring of seafood began in the late 1980s. Initial estimates showed that a high rate of consumers of seafood were receiving exposures of similar to 0.3 mSv y^{-1}. In this paper, an oceanographic model is used to hindcast exposures since 1954 and to forecast trends in the future. Using new dosimetric data derived from a human uptake study at this laboratory, exposures of local seafood consumers were cautiously predicted to have risen to 3 mSv y^{-1} in 1963, peaking at 6 mSv y^{-1} in 1983 and falling to 0.04 mSv y^{-1}after the reduction in discharges in the early 1990s. Most of these doses were due to Po-210 in crustaceans and molluscs. The model predicts a quick return to near background concentrations in the environment during the early 1990s. Environmental measurements have shown a slower, though significant, reduction in concentrations. External radiation from radionuclides absorbed onto fine sediments is identified as another source of exposure deserving further study. Taking into account the pessimistic nature of the assumptions in the assessment, it is unlikely that exposures exceeded the dose limit of 5 mSv y^{-1} due to the operation of the plant.

Chadwick, R.C. and Chamberlain, A.C., 1970. Field Loss of Radionuclides from Grass. *Atmospheric Environment*, 4, 51.

Chamberlain, A.C., 1996. Emissions from Sellafield and activities in soil. *Science of the Total Environment*, 177, 259-280.

Abstract: The discharges of radioactivity from the Windscale piles and other installations at Sellafield remain a matter of concern, though 30 or 40 years have elapsed since most of the emissions occurred. New data on emissions and activities in soil have become available in the past few years. They are reviewed, together with the older data, with particular reference to the emissions of irradiated uranium oxide from the Windscale piles, the releases in the 1957 accident, and the aerial emissions from the reprocessing plant.

Copplestone, D., Johnson, M.S., Jackson, D. and Jones, S.R., 2000. Doses to terrestrial biota in the vicinity of BNFL Sellafield, Cumbria, UK. *Radiation Protection Dosimetry,* 92, 177-182.

Abstract: Source terms and corresponding radionuclide activity concentrations in biota for Cs-134, Cs-137, Pu-238, Pu-230+240 and Am-241 have been assessed for three semi-natural ecosystems in the vicinity of BNFL Sellafield, Cumbria, UK. Estimates of absorbed doses (mGy d^{-1}) have been calculated. Doses to key indicator species, *Oniscus asellus* (detritivorous invertebrate), *Carabus violaceous* (predatory invertebrate) and *Apodemus sylvaticus* (granivorous wood mouse) are discussed with reference to the 1 mGy d^{-1} level, below which it is postulated that no observable effects on populations in a terrestrial ecosystem occur. Implications for the 'critical group' and 'reference model' approaches for a framework of radiological environmental protection are discussed. The need to assess the most highly exposed species is advanced. New research focused on the application of biomarker techniques as a mechanism for determining the interactions and effects of environmental contaminants on ecosystem structure and functioning is presented.

Curtis, E.J.C., Popplewell, D.S. and Ham, G.J., 1991. Radioactivity in Environmental-Samples Taken in the Sellafield, Ravenglass and Morecambe Bay Areas of West Cumbria. *Science of the Total Environment,* 105, 211-231.

Abstract: Seaborne sediments deposited in the estuaries of the Esk, Duddon, Leven and Kent have been analysed for fission products and actinides discharged in waste from the Sellafield processing works in West Cumbria, and the values compared with the generally expected values due to fallout from atmospheric nuclear weapons tests. Analyses of tissues from sheep grazing the marshes of these estuaries show that the internal radiation dose of the general public through eating mutton or liver from these animals would be at most a few percent of recommended limits. Analytical data are presented on the actinide content of beef cattle, and on potato crops grown under field conditions; these data show that, as with the sheep data, the radiation dose to the consumer would be small.

Dollard, G.J. and Lepp, N.W., 1980. Differential Mobility of Lead and Zinc in Phloem Tissue of Sycamore (*Acer-Pseudoplatanus* L). *Zeitschrift fur Pflanzenphysiologie,* 97, 409-415.

Farmer, J.G., 1979. Lead in Wild Blackberries from Suburban Roadsides. *Journal of the Science of Food and Agriculture,* 30, 816-818.

Farmer, J.G., Eades, L.J., Atkins, H. and Chamberlain, D.F., 2002. Historical trends in the lead isotopic composition of archival Sphagnum mosses from Scotland (1838-2000). *Environmental Science and Technology,* 36, 152-157.

Abstract: The analysis of almost 200 Scottish Sphagnum moss samples collected over the past 170 years has revealed trends in the isotopic composition of lead similar to those previously established for dated Scottish lake sediments and peat bogs, lending credibility to these proxy records of atmospheric lead contamination and deposition. The effect of temporal variations in contributions from sources such as smelting of indigenous lead ores (Pb-206/Pb-207 similar to 1.16-1.18), coal combustion (Pb-206/Pb-207 similar to 1.17-1.19), and the use of imported Australian lead (Pb-206/Pb-207 similar to 1.04) was clearly seen in the Scottish moss Pb-206/Pb-207 record. This showed some differences from the corresponding archival herbage record for the south of England, where the initial influence of Australian lead occurred earlier, at the end of the 19th century. A significant decline from a Pb-206/Pb-207 value of similar to 1.17 in the Scottish moss record began in the 1920s and continued until the 1980s (Pb-206/Pb-207 similar to 1.12). The success of measures to reduce lead emissions to the atmosphere over the past 20 years in the UK, in particular from petrol engine vehicles using alkyl lead additives manufactured primarily from Australian lead, is evident in both the increasing Pb-206/Pb-207 ratio and falling lead concentration data for Scottish moss.

Garland, J.A. and Mckay, W.A., 1992. The Influence of Grassland Management on the Radionuclide Inventory of Soils in West Cumbria, UK Comment. *Journal of Environmental Radioactivity,* 15, 81-83.

Haas, G., Schupfner, R. and Muller, A., 1995. Transfer of Natural and Man-Made Radionuclides from Plants to Roe Deer and Farm-Animals. *Journal of Radioanalytical and Nuclear Chemistry Articles,* 194, 269-276.

Abstract: In this work, the transfer behaviour of long living radionuclides from the thorium decay series (Ra-228, Th-228, Th-232) as well as of K-40 and Cs-137 is studied. In a small area of middle Europe (South East Germany) showing an increased thorium content of soil, the activity concentrations in samples of feed plants, farm animals, farm animal products and roe deer has been determined. The concentration ratios feed-to-animal tissue and to animal products are calculated, indicating a significantly enhanced transfer from feed to roe deer tissues. Determinations of the activity concentrations in fish (carp), pig (tissues), egg and milk complete this examination. Among all studied samples which are important for human nourishment, eggs and carp cause the greatest exposure by ingestion.

Ham, G.J., Wilkins, B.T. and Ewers, L.W., 2001. Pb-210, Po-210, Ra-226, U and Th in arable crops and ovine liver: Variations in concentrations in the United Kingdom and resultant doses. *Radiation Protection Dosimetry,* 93, 151-159.

Abstract: Concentrations of a range of naturally occurring radionuclides have been determined in the same crops grown at two sites in the UK. Ovine liver has also been studied. One site was in an area where concentrations in soil are typical of the UK (the 'control' site) and the other in an area where levels were well above average (the 'test' site). For an average adult consumer of all of the foodstuffs studied, the doses from consumption for the test site were about four times higher than those for the control site. However, the differences were small compared with the variability in overall doses from natural background across the UK. Pb-210 and Po-210 were important contributors to doses for both sites, but at the test site the contribution from Ra-226 was also significant.

Of the foodstuffs studied, consumption of leafy vegetables and liver gave the highest doses. The doses from leafy vegetables were sensitive to the weather conditions prior to harvesting. Consequently, rigorous monitoring programmes should be based on several samples collected throughout the year; extrapolations based on a single annual sample are unlikely to be reliable.

Hamilton, E.L., 1980. Concentration and distribution of Uranium in *Mytilus edulis* and associated materials. *Marine Ecology Progress Series*, 2, 61-73.

Hill, C.R., 1960. Lead-210 and Polonium-210 in Grass. *Nature,* 187, 211-212.

Horrill, A.D. and Mudge, S., 1990. The Influence of Grassland Management on the Radionuclide Inventory of Soils in West Cumbria, UK. *Journal of Environmental Radioactivity,* 12, 143-165.

Howard, B.J., 1985. Aspects of the Uptake of Radionuclides by Sheep Grazing on an Estuarine Saltmarsh. 1. The Influence of Grazing Behaviour and Environmental Variability on Daily Intake. *Journal of Environmental Radioactivity,* 2, 183-198.

Howard, B.J. and Lindley, D.K., 1985. Aspects of the Uptake of Radionuclides by Sheep Grazing on an Estuarine Saltmarsh. 2. Radionuclides in Sheep Tissues. *Journal of Environmental Radioactivity*, 2, 199-213.

Kalac, P., 2001. A review of edible mushroom radioactivity. *Food Chemistry,* 75, 29-35.

Abstract: The review deals mainly with the situation in Europe, where wild-growing mushrooms are widely consumed as a delicacy and some species have been found to be extensively contaminated by radioactive fallout from the Chernobyl disaster in 1986. The natural isotope K-40 usually causes activities of 0.8-1.5 kBq kg^{-1} dry matter. Activities of Cs-137 from nuclear weapons testing were commonly reported below 1 kBq kg^{-1} dry matter until 1985. The situation changed dramatically after the Chernobyl accident and activities up to tens of kBq kg^{-1} dry matter of Cs-137 and to a lesser extent of Cs-134 were observed in the following years in some edible species. Among the heavily accumulating species belong *Xerocomus* (*Boletus*) *badius*, *Xerocomus chrysenteron*, *Suillus variegatus*, *Rozites caperata* and *Hydnum repandum*. Activity concentrations have been affected by several environmental factors, such as the rate of soil contamination with fallout, the horizon from which mycelium takes nutrients, soil moisture and time from the disaster. Wild mushroom consumption contributed up to 0.2 mSv to the effective dose in individuals consuming about 10 kg (fresh weight) of heavily contaminated species per year. The radioactivity of cultivated mushrooms is negligible. Contamination can be considerably decreased by soaking or cooking of dried or frozen mushroom slices. Animals such as deer, which eat mushrooms, have elevated levels of radionuclides in their tissues.

Keating, G.E., McCartney, M. and Davidson, C.M., 1996. Investigation of the technological enhancement of natural decay series radionuclides by the manufacture of phosphates on the Cumbrian coast. *Journal of Environmental Radioactivity,* 32, 53-66.

Abstract: The aim of this study is to investigate the response of the aquatic environment following the cessation of discharges from a phosphate-ore processing plant, situated at Whitehaven, North West England. A preliminary survey was carried out to determine Po-210, Pb-210 and U-238 activities in intertidal biota and sediment in the vicinity of the plant in November 1992. The maximum concentrations observed in the samples are all above the reported natural ranges and are clearly indicative of technological enhancement. The highest Po-210 (7,749 +/- 201 Bq kg^{-1} dry weight) and U-238 (390 +/- 12 Bq kg^{-1} dry

weight) activities were found in the sediment collected from Whitehaven Harbour, the sampling site closest to the outfall pipe. At all sites, with the exception of Whitehaven itself, Po-210 levels in mussels were greater than winkles which, in turn, were greater than levels in sediments which exceeded the levels observed in seaweed. Uranium levels were found to be greatest in sediment, while the rest of the sample types exhibited similar levels of activity. Initial Pb-210 results show similar levels in mussels and winkles, which suggests the uptake mechanism to be different than that of Po-210. Further surveys have since been carried out at approximately four to five-month intervals and the temporal trends obtained suggest an overall decrease in radionuclide concentrations in all sample types.

Lepp, N.W. and Dollard, G.J., 1974. Studies on Behaviour of Lead in Wood - Binding of Free and Complexed Pb-210 to Xylem Tissue. *Oecologia*, 16, 369-373.

Lowe, V.P.W., 1991. Radionuclides and the Birds at Ravenglass. *Environmental Pollution*, 70, 1-26.

Abstract: Since 1983, concern has been expressed about the apparent decline in numbers of birds in the Ravenglass estuary in West Cumbria, particularly of the black-headed gull colony on the Drigg dunes, and suggestions have been made that this decline might be due to excessive radiation in the birds' food and their general environment. Twelve species of marine invertebrates from Ravenglass, most of them known to be important foods for birds, were analysed, and further samples were taken from sites along the West Cumbrian coast. None of these samples showed excessive contamination with any of the radionuclides analysed. Analysis of a sample of bird carcasses from the areas showed oystercatchers (*Haematopus ostralegus*) and shelduck (*Tadorna tadorna*) to have some of the highest concentrations of ^{137}Cs in their tissues; yet their breeding success and populations were not affected. Black-headed gulls, on the other hand, were found to be feeding mainly inland, and were the least contaminated with radionuclides of all the birds at Ravenglass, yet this species and its breeding success were in decline. Calculations of the total dose equivalent rate to the whole body of the most contaminated black-headed gull amounted to 9.8 x 10^{-4} mSv h^{-1} (equivalent to 8.4 x 10^{-4} mGy h^{-1}, whole-body absorbed dose rate), and the background exposure dose was of the order of 8.3 x 10^{-4} mGy h^{-1}. As a minimum chronic dose of 1,000 mGy d^{-1} has been found necessary to retard growth of nestling birds, and 9,600 mGy over 20 days of incubation to cause the death of 50 per cent of embryos in black-headed gulls' eggs, the concentrations of radionuclides in the foods, body tissues and general environment were at least three orders of magnitude too low to have had any effect.

McCartney, M., Davidson, C.M., Howe, S.E. and Keating, G.E., 2000. Temporal changes in the distribution of natural radionuclides along the Cumbrian coast following the reduction of discharges from a phosphoric acid production plant. *Journal of Environmental Radioactivity*, 49, 279-291.

Abstract: The concentrations of a range of natural radionuclides (U-238, U-234, Th-232, Th-230, Th-228, Pb-210 and Po-210) in the marine environment near a phosphoric acid production plant have been monitored over the period 1992-1997. At the beginning of the study period, concentrations of U-238 and its daughters were clearly enhanced in mussel, winkle and sediment samples. The highest concentrations of U-238, Th-230, Pb-210 and Po-210 in the sediment of Whitehaven harbour were 685 +/- 17, 1,290 +/- 35, 140 +/- 2 and 7,750 +/- 200 Bq kg^{-1}, respectively. Since 1992, discharges of these radionuclides have been greatly reduced and levels in the environment have decreased accordingly. The radiation dose to the general public is mainly due to the ingestion of Po-

210 and to a lesser extent Pb-210 in molluscs. The committed effective dose to a group of local seafood consumers via this route has decreased from 1.0 mSv yr^{-1} in 1993 to 0.19 mSv yr^{-1} in 1997, although the latter is still above the expected background level of 0.1 mSv yr^{-1}. The enhanced levels are maintained by the reduced but not insignificant discharges from the plant and, at Whitehaven perhaps, by the continuing presence of phosphate ore in the sediment.

McDonald, P., Cook, G.T. and Baxter, M.S., 1992. Natural and Anthropogenic Radioactivity in Coastal Regions of the UK. *Radiation Protection Dosimetry,* 45, 707-710.

Abstract: The radionuclide contents of a range of coastal marine samples (sediment, sea water, seaweed, mussels and winkles) collected from 15 UK sites have been investigated to provide a perspective on the relative contributions to enhanced marine radioactivity in the UK by the nuclear and non-nuclear industries. Sites typifying high levels of anthropogenic radioactivity (Irish Sea area) were influenced by the authorised discharges from the nuclear fuel reprocessing plant at Sellafield into the North East Irish Sea. Enhanced concentrations of natural radionuclides were due to: (1) discharges from a phosphate ore processing plant (Whitehaven) and (2) dumping of coal spoil directly into the North Sea (Blackhall Colliery). New site-specific concentration factor (CF) values derived here are consistent with existing literature data except for Pb-210 and Po-210, for which the results suggest that the upper and lower values of the currently recommended CF ranges should be extended. Radiologically, the highest potential exposure to the public calculated from the results derives from the ingestion of Whitehaven mussels (3.2 mSv y^{-1}), the greatest single contribution being from technologically enhanced Po-210.

McDonald, P., Baxter, M.S. and Fowler, S.W., 1993. Distribution of Radionuclides in Mussels, Winkles and Prawns. 1. Study of Organisms Under Environmental-Conditions Using Conventional Radio-Analytical Techniques. *Journal of Environmental Radioactivity,* 18, 181-202.

Abstract: Mussels (*Mytilus edulis*) and winkles (*Littorina littorea*) collected from Ravenglass, Cumbria, England in the vicinity of the British Nuclear Fuels plc nuclear reprocessing plant at Sellafield, and prawns (*Palaemon serratus*) landed nearby at Whitehaven, have been investigated to determine the distributions of alpha-emitting (Po-210, Pu-238, Pu-239+240, Am-241) and gamma-emitting (Nb-95, Zr-95, Ru-103, Ru-106, Cs-137, Am-241) radionuclides in their tissues and organs. Ravenglass mussels exhibited Pu-239+240 concentrations ranging from 43 Bq kg^{-1} dry weight in muscle tissue to 1,658 Bq kg^{-1} dry in byssal threads, the corresponding Cs-137 range being 131-1,340 Bq kg^{-1}. Although Po-210 concentrations were not determined in byssal threads, muscle tissue still displayed the lowest polonium concentration (124 Bq kg^{-1}), whilst the viscera (containing digestive gland, stomach and kidneys) contained the highest (596 Bq kg^{-1}). Subsequent concentration factor estimates for Cs-137, Po-210 and Pu-239+240 in the total soft parts of Ravenglass mussels were, respectively, 9, 25, 800 and 1,400. In Cumbrian winkles, nuclide concentrations ranged for Pu-239+240, from 18.5 Bq kg^{-1} dry weight (muscle tissue) to 45.7 Bq kg^{-1} dry weight (pallial complex); for Cs-137, from 103 (foot tissue) to 1,495 Bq kg^{-1} (pallial complex) and for Po-210, from 12.2 (muscle tissue) to 145 Bq kg^{-1} (digestive gland). Total soft part concentration factors (CF) were calculated to be 16 for Cs-137, 5,500 for Po-210 and 5,700 for Pu-239+240. The magnitude of these CF, as for those in the mussel, is consistent with the respective CF values recommended by the International Atomic Energy Agency for molluscs. Radionuclide concentrations in Whitehaven-landed prawns were much lower than those observed in mussels or winkles; no artificial gamma-emitter activities were present above

detection limits and the highest Pu-239+240 concentration was 5.96 Bq kg^{-1} dry weight in the carapace. Po-210 activities, however, were more readily detectable throughout the prawn tissues, with concentrations ranging from 2.7 Bq kg^{-1} (abdomen muscle) to 144 Bq kg^{-1} (cardiac foregut), producing CF of the order of 2 x 10^4 in tissues associated with feeding and digestion. Previous studies have attempted to determine the principal nuclide source to marine organisms by comparing nuclide activity ratios in their tissues, sea water and particulate material. From the environmental samples studied here, no single transport medium appears to dominate uptake. The primary radiological implication of the observed radionuclide concentrations in Ravenglass mussels and winkles is that, from seafood ingestion, the critical group receives only a small percentage (around 10 per cent) of the ICRP-recommended subsidiary dose limit. Dose contributions from Po-210 are higher than those from Pu-239+240 in mussels, but are less than those from Pu-239+240 in winkles.

McDonald, P., Baxter, M.S. and Scott, E.M., 1996. Technological enhancement of natural radionuclides in the marine environment. *Journal of Environmental Radioactivity,* 32, 67-90.

Abstract: This review summarizes aspects of technologically enhanced radioactivity in the UK marine environment and considers briefly related investigations in western Europe. It also discusses some models for the kinetics of series decay and ingrowth which can be applied to technological inputs of series members to the marine environment and to their differential elemental biogeochemistries.

McDonald, P., Jackson, D., Leonard, D.R.P. and Mckay, K. 1999. An assessment of Pb-210 and Po-210 in terrestrial foodstuffs from regions of England and Wales. *Journal of Environmental Radioactivity*, 43, 15-29.

Abstract: Based on data for nationally available foodstuffs, naturally occurring Pb-210 and Po-210 contribute significantly to the UK radiation dose from dietary intake. To provide a more complete overview of radiological implications to the public, samples of offal, cereal, fruit, root and green vegetables were collected from 11 sites, including regions of potential Pb-210 and Po-210 enhancement. Considerable variability was evident in levels of Pb-210 and Po-210 from all sites and in all food types investigated. Higher concentrations were typically found in offal and cereals. Lowest concentrations were generally found in root vegetables. Between sites, the only evidence of elevated concentrations of Po-210 occurred at Helston (Po-210, 3.0 Bq kg^{-1} in bovine liver, no other offal sample exceeding 0.8 Bq kg^{-1}) and Holyhead (Po-210, 0.4 Bq kg^{-1} in blackberries, no other fruit sample exceeding 0.1 Bq kg^{-1}). Other foodstuffs from these sites did not show similar enhanced levels of Po-210. The maximum dose arising from consumption of Pb-210 and Po-210 in foods surveyed in this study is estimated to be around 120 µSv y^{-1} to adults. This compares with an estimated UK average consumption dose from all nuclides and foodstuffs of 300 µSv y^{-1} (broad range 100-1,000 µSv y^{-1}).

Mcgee, E.J., Synnott, H.J., O'keefe, C. and Colgan, P.A., 1995. Radionuclide Uptake by Red Deer (*Cervus-Elaphus*) on Mountain Grazing. *British Veterinary Journal,* 151, 671-682.

Abstract: Forty-two red deer (*Cervus elaphus*) were shot during the 1992 annual cull in Glenveagh National Park, Ireland. Samples of rumen, kidney and faeces were removed from each animal. Kidney samples were used to estimate flesh radiocaesium (Cs-137) concentrations and the 95 per cent confidence interval for the mean was 203 +/- 12 Bq kg^{-1}. The maximum recorded Cs-137 concentration in kidney was 367 Bq kg^{-1}(fresh

weight). The altitude of the cull and the age and sex of each animal were recorded. Neither age nor sex correlated with concentrations of Cs-137 in rumen, kidneys or faeces. Despite the limited altitudinal range of the study and the free ranging behaviour of deer, there was a highly significant positive correlation between rumen, kidney and faecal Cs-137 concentrations and the altitude of the cull. K-40 concentrations in rumen, kidney and faeces did not correlate with the altitude of cull, age or sex of slaughtered animals. Significant Cs-137 concentration differences were identified in the sequence: rumen < faeces = faeces. Cs-137 concentrations in rumen, kidney and faeces for individual animals were all significantly correlated. Statistical testing showed that the concentration sequence for K-40 was: rumen < kidney = faeces; a sequence which differs from that of Cs-137. A comparison of rumen:faecal ratios demonstrated that significantly more Cs-137 was excreted in faeces than was the case for K-40. The concentration of Cs-137 excreted in faeces relative to concentrations in forage (rumen), is approximately twice that for K-40. Linear regression of faecal Cs-137 concentrations (y) on kidney concentrations (x) was carried out, the regression equation is y = -86.90 + 0.97x. This equation (R^2) = 0.73, (F-1, F-40 = 107) may be used to predict Cs-137 concentrations in flesh by measurement of faecal concentrations. This is a useful preliminary assessment method, particularly with herds of wild animals that prove difficult to capture for in vivo monitoring.

Otlet, R.L., Walker, A.J., Fulker, M.J. and Collins, C., 1997. Background carbon-14 levels in UK foodstuffs, 1981-1995, based upon a 1992 survey. *Journal of Environmental Radioactivity*, 34, 91-101.

Abstract: Knowledge of the current natural background level is important to carbon-14 related studies, especially dose assessments in the vicinity of and distant from nuclear establishments, since its value must be subtracted from measured levels to determine local enhancement. Although measurements have been made world-wide to monitor the decline of the background level, from its peak in the early 1960s to the present, there exists a paucity of precise data for UK natural materials. Accordingly, this study was carried out to establish an average figure for the natural C-14 level in foodstuffs in 1992 from a survey of materials collected from a wide range of sites over England and Wales, using this single year value, to fill in missing years prior to 1992 and to extrapolate for some years afterwards. Results are presented from the survey and from the derived extrapolation, with comparison made with measurements from other laboratories.

Pearce, F., 1983. Polonium Cloud Engulfs Windscale. *New Scientist,* 97, 867.

Purvis, O.W., Bailey, E.H., McLean, J., Kasama, T. and Williamson, B.J., 2004. Uranium biosorption by the lichen *Trapelia involuta* at a uranium mine. *Geomicrobiology Journal,* 21, 159-167.

Abstract: Metal localisation was investigated in the lichenised ascomycete *Trapelia involuta* growing on a range of uraniferous minerals including metazeunerite [$Cu(UO_2)_2(AsO_4)_2.8H_2O$], metatorbernite [$Cu(UO_2)_2(PO_4)_2.8H_2O$], autunite [$Ca(UO_2)_2(PO_4)_2.10H_2O$] and uranium-enriched iron oxide and hydroxide minerals at the abandoned South Terras mine site, Cornwall, UK. Apothecia from samples collected from waste dumps at the mine had an unusually dark colour that decolorized with NaOCl, an observation which together with Fourier Transform Infrared Spectroscopy of apothecial extracts, suggested the presence of melanin-like pigments. X-ray element mapping and probe traverses across the lichen-rock interface identified the highest U, Fe, and Cu concentrations in the outer parts of melanised apothecia. Accumulation of mineral particulates and complexing with lichen acids were not considered responsible

for this, since element ratios in the traverses did not correspond with those of likely mineral phases and lichen metabolites were localised in different tissues. Metal biosorption by melanin-like pigments was likely to be responsible for the observed metal fixation. No detectable U or Cu was observed in control samples, although Fe showed a similar localisation in some specimens. The high concentrations of mucopolysaccharides and P recorded inside apothecia (within asci containing reproductive spores and hypothecium) suggests that the formation of melanised tissues may help protect vital reproductive tissues from the toxic effects of U and other metals, since the uranyl ion complexes strongly with phosphate species.

Rollo, S.F.N., Camplin, W.C., Allington, D.J. and Young, A.K., 1992. Natural Radionuclides in the UK Marine-Environment. *Radiation Protection Dosimetry*, 45, 203-209.

Abstract: The importance of natural radionuclides giving rise to radiation exposure of man from marine consumption pathways has been known for some time. However, the extent of surveys of levels in marine biota has been limited. This paper presents new data on concentrations of natural radionuclides in fish, shellfish and seaweeds taken from coastal sampling locations in the UK. Sampling included areas where levels due to natural sources would be predominant, but efforts were made to study potential sources of technologically enhanced discharges to seas and rivers, particularly the phosphogypsum plant at Whitehaven in Cumbria. The highest concentrations (up to 371 Bq kg^{-1} (wet) Po-210) were observed in winkles near Whitehaven. The general levels at sites remote from known sources were much lower. Monthly concentrations in molluscs at a single location were elevated by approximately a factor of two during the summer months. An assessment of the expected doses to members of the public from marine consumption pathways is made.

Ryan, T., 2002. Transuranic biokinetic parameters for marine invertebrates - a review. *Environment International*, 28, 83-96.

Abstract: A catalogue of biokinetic parameters for the transuranic elements plutonium, americium, curium, neptunium, and californium in marine invertebrates is presented. The parameters considered are: the seawater-animal concentration factor (CF); the sediment-animal concentration ratio (CR); transuranic assimilation efficiency; transuranic tissue distribution and transuranic elimination rates. With respect to the seawater-animal CF, authors differ considerably on how they define this parameter and a seven-point reporting system is suggested. Transuranic uptake from sediment by animals is characterised by low CR. The assimilation efficiencies of transuranic elements in marine invertebrates are high compared to vertebrates and mammals in general, and the distribution of transuranics within the body tissue of an animal is dependent on the uptake path. The elimination of transuranics from most species examined conformed to a standard biphasic exponential model, though some examples with three elimination phases were identified.

Side, J., 1985. Radioactivity on UK Coasts. *Marine Pollution Bulletin*, 16, 340-341.

Smithbriggs, J.L. and Bradley, E.J., 1984. Measurement of Natural Radionuclides in UK Diet. *Science of the Total Environment*, 35, 431-440.

Smithbriggs, J.L., Bradley, E.J. and Potter, M.D., 1986. The Ratio of Pb-210 to Po-210 in UK Diet. *Science of the Total Environment,* 54, 127-133.

Sugden, C.L., Farmer, J.G. and Mackenzie, A.B., 1993. Isotopic-Ratios of Lead in Contemporary Environmental Material from Scotland. *Environmental Geochemistry and Health,* 15, 59-65.

Abstract: Lead emitted into the environment, primarily from the combustion of leaded petrol and industrial activities, retains the isotopic signature of the ore(s) from which it is derived. Leaded petrol, atmospheric particulates and street dust sampled in central Edinburgh between February 1989 and December 1991 had mean Pb-206/Pb-207 ratios of 1.082 +/- 0.024, 1.092 +/- 0.011 and 1.109 +/- 0.016 respectively. These isotope ratios were found to be depleted in Pb-206 compared with a mean of 1.160 +/- 0.012 for tap water in contact with lead pipes and typical ratios of 1.17-1.19 for British lead ore deposits and coal. Paint, with an observed wide range of Pb-206/Pb-207 ratios (1.083-1.183), appears to have significantly influenced house dust and some street dust Pb-206/Pb-207 values. Such overlaps and influences may hinder the quantitative apportionment, via isotope data, of source and route in general population surveys of human exposure to lead.

Sumerling, T.J., 1984. The Use of Mosses as Indicators of Airborne Radionuclides Near a Major Nuclear Installation. *Science of the Total Environment,* 35, 251-265.

Swift, D.J., Smith, D.L., Allington, D.J. and Ives, M.J., 1994. The Po-210 Content of North Sea Edible Crab, *Cancer pagurus L*, and Common Shrimp, *Crangon crangon L* and the Potential Radiological Impact. *Journal of Environmental Radioactivity,* 23, 213-230.

Abstract: The Po-210 content of brown shrimp and edible crab was measured in monthly samples over one year to investigate possible seasonal changes. The highest value measured in shrimp was found in August for hepatopancreas tissue. Tail muscle values were significantly lower than those in the hepatopancreas but followed a similar trend, with the highest value being measured in August. However, in neither tissue were these statistically significant changes correlated with time. No statistically significant difference was found between male and female crab brown meat Po-210. There was no statistically significant variation in the brown meat Po-210 content of female crabs over the period of the study. However, there was a statistically significant variation in the brown meat Po-210 content for males, but there was no clear pattern with time. Male claw muscle contained less Po-210 than female and was more variable with time, although, again, without a clear pattern being visible. The median individual effective dose equivalent for Po-210 from eating North Sea shrimp tail muscle was estimated at about 0.001 mSv y^{-1} by using the NRPB recommended dose coefficient of 4.35 x 10^{-7} Sv Bq^{-1}. The equivalent value for eating dressed crab (mixed crab brown meat and claw muscle) was about 0.02 mSv y^{-1}. A measure of whole shrimp, traditionally one pint (568 ml), was estimated to represent an effective dose equivalent of about 0.007 µSv. The equivalent mean value for an average weight of dressed crab (135 g wet weight) was 1.1 µSv (range 0.6-2.2 µSv). The mean concentration for Po-210 in edible crab brown meat was calculated as 2.7 x 10^4 Bq kg^{-1} (range 1.0 x 10^4 to 4.7 x 10^4) and for claw muscle 590 Bq kg^{-1} (range 3.0 x 10^2 to 1.3x10^3). These results show that the IAEA recommended value for crustacea, 5 x10^4 Bq kg^{-1}, can overestimate the accumulation of Po-210 in the edible fractions of some commercially important crustacean species.

Swift, D.J., Smith, D.L., Allington, D.J. and Winpenny, K., 1995. A Laboratory-Study and Field-Study of Po-210 Depuration by Edible Winkles (*Littorina littorea L*) from the Cumbrian Coast (North Eastern Irish Sea). *Journal of Environmental Radioactivity*, 26, 119-133.

Abstract: Edible winkles from Saltom Bay, Cumbria (North East Irish Sea) had flesh Po-210 concentrations in excess of 200 Bq kg^{-1} (wet). This came from liquid waste discharged under authorization from a chemical plant producing phosphoric acid. This labelling of the winkles under natural conditions was exploited to determine the Po-210 depuration rate and biological half-time. Winkles transferred to Lowestoft and depurated in flowing seawater had biological half-times ranging from 82 to 119 days, depending on temperature. These values seem long from the few data available in the literature. Cadmium was also discharged in the liquid waste and cadmium was measured in the depurating winkles. The hypothesis is put forward that long half-times were due to Po-210 binding to sob-cellular proteins induced by the presence of cadmium. After the chemical plant was shut, changes in Saltom Bay winkle flesh contents of Po-210, Pb-210 and cadmium were followed by monthly samples. The biological half-time in the environment for the decrease in Po-210 after the plant closed was estimated at about 92 days.

Watmough, S.A. and Hutchinson, T.C., 2002. Historical changes in lead concentrations in tree rings of sycamore, oak and Scots pine in North West England. *Science of the Total Environment,* 293, 85-96.

Abstract: Lead concentrations in tree rings of sycamore (*Acer pseudoplatanus L*.), oak (*Quercus robur L*.) and Scots pine (*Pinus sylvestris L*.) sampled at a parkland in North West England were measured in wood formed since the mid-1800s. Concentrations of Pb in Scots pine and oak peaked in wood formed between 1900 and 1940, most likely because of Pb accumulation in heartwood, indicating that oak and Scots pine are unsuitable for monitoring temporal changes in Pb deposition at the study site. In contrast, Pb concentrations in sycamore, a species that has similar heartwood and sapwood chemistry, were relatively constant in wood formed between the mid-1800s and 1950. Lead concentrations decreased steadily in sycamore tree rings formed after the 1950s, and decreased more abruptly in wood formed after 1985. This sharp decrease in wood Pb cannot be due to decreases in soil Pb concentration. Stable Pb isotope analysis was used to further investigate Pb patterns in sycamore wood. Excess Pb-206/Pb-207 ratios in tree rings of sycamore were relatively constant, approximately 1.17, in wood formed prior to the 1930s, but decreased steadily thereafter, reaching a minimum value of approximately 1.16 in wood formed between 1975 and 1985. after which time Pb-206/Pb-207 ratios increased. This pattern is consistent with changes in Pb isotope ratios measured in peat, sediment and aerosol samples in the UK. However, the magnitude of the decrease in Pb-206/Pb-207 (largely due to gasoline Pb) is considerably lower than in other studies and our estimates indicate that less than 20 per cent of the total Pb in sycamore wood measured since the mid-1800s is derived from gasoline emissions. A more likely explanation for the pattern of Pb observed in sycamore tree rings is that soil Pb accumulates within rings of the diffuse porous wood over a number of years. Such uptake patterns would result in lower Pb concentrations in the outer (more recently formed) tree rings, which coincide with recent reductions in Pb deposition in the UK. Overall, this study indicates that tree ring chemistry is unsuitable for monitoring historical changes in Pb deposition at the study site.

Watson, W.S., Sumner, D.J., Baker, J.R., Kennedy, S., Reid, R. and Robinson, I., 1999. Radionuclides in seals and porpoises in the coastal waters around the UK. *Science of the Total Environment*, 234, 1-13.

Abstract: It has been suggested that marine predators be assessed for biologically relevant contamination levels because of their trophic position. Accordingly, in studying

radioactive contamination in the marine environment around the UK, tissues from seals and porpoises were chosen. Liver and muscle tissue from dead seals and porpoises found stranded around the UK coast were analysed for the following radionuclides: Cs-134, Cs-137, Pu-238, Pu-239 and Pu-240. Multifactor analysis of variance indicated that, for radiocaesium, there was no significant difference for harbour seals, grey seals or porpoises in terms of species or gender; however, the tissue activity concentration increased with body weight and decreased with distance from Sellafield, the major nuclear reprocessing plant in the UK. Levels of radiocaesium in muscle were higher than those in liver, while there appeared to be a concentration factor of approximately three to four for muscle radiocaesium when compared to radiocaesium levels reported for fish, the main food source of the marine mammals under study. Approximate radiation dose calculations indicated that the average dose from radiocaesium was less than 10 per cent of the dose from the naturally occurring radioisotope of potassium, K-40. The highest tissue activity concentration for plutonium of 0.037 Bq kg^{-1} (Pu-239 and Pu-240) was detected in a grey seal stranded at Rathlin Island in Northern Ireland. Calculation of approximate radiation doses from plutonium contamination showed that, as with radiocaesium, the average dose was small compared with that from ^{40}K. In summary, the radiocaesium contamination in seals and porpoises decreased with distance from Sellafield, indicating that the BNFL reprocessing plant was the major source of the contamination. Marine mammals concentrated radiocaesium from their environment by a factor of 300 relative to the concentration in seawater, indicating the value of using marine mammal tissue to measure radiocaesium contamination in the marine environment. The maximum radiation dose to marine mammals from radiocaesium was higher than doses previously assessed for critical groups of humans living near Sellafield, while the maximum dose from plutonium was comparable to the doses for humans.

Wildgust, M.A, McDonald, P. and White, K.N., 1998. Temporal changes of Po-210 in temperate coastal waters. *Science of the Total Environment,* 214, 1-10.

Abstract: The temporal variation of Polonium-210 (Po-210) was examined in coastal sea water, the mussel *Mytilus edulis*, the winkle *Littorina littorea* and green alga *Ulva lactuca* in order to investigate the entry of Po-210 into the marine food chain. More than 99 per cent of Po-210 in the water column occurred in the particulate phase. Dissolved Po-210 concentrations peaked during the spring phytoplankton bloom and it is suggested this is related to preferential scavenging of Po-210 by the increased numbers of bacteria, viruses and small dissolved particulates. Changes in *L. littorea* Po-210 specific activity are thought not to be related to food, but to a drop in body weight following spawning. Much of the Po-210 accumulated by *M. edulis* was located in the digestive gland. The specific activity of Po-210 in the digestive gland of *M. edulis* was shown to be strongly correlated with changes in seawater suspended particulate specific activity. Examination of other trace metal (Ag, Al, As, Ca, Cd, Cr, Co, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, Sb, Se, Sn and Zn) variations in the digestive gland revealed that class B and borderline metals had a strong positive correlation with Po-210. Ongoing work is investigating whether the accumulation and loss of Po-210 is affected by the presence of metallothioneins.

Yoshitome, R., Kunito, T., Ikemoto, T., Tanabe, S., Zenke, H., Yamauchi, M. and Miyazaki, N., 2003. Global distribution of radionuclides concentrations (Cs-137 and K-40) in marine mammals. *Environmental Science and Technology,* 37, 4597-4602.

Abstract: Concentrations of anthropogenic radionuclides were measured in the muscle of marine mammals collected from various locations all over the world, and the global distribution of Cs-137 in marine mammals was investigated. K-40 was detected in all the

specimens of marine mammals with no apparent difference between regions. An anthropogenic radionuclide, Cs-137, was detected in most of the species of marine mammals. With regard to the worldwide distribution of Cs-137, the highest concentration was noticed in the UK coast, followed by Lake Baikal, and decreased toward the southern sampling points. A strong positive correlation was observed between Cs-137 levels in the muscle of marine mammals and the ambient seawater. Marine mammals feeding on fish showed a higher concentration factor (CF) for Cs-137 than those feeding on cephalopods. To our knowledge, this is the first report on the global distribution of Cs-137 and the effect of feeding habits on the CF values of Cs-137 in marine mammals.

Appendix B.1: Dose conversion coefficients

Naturally occurring radionuclide unweighted dose conversion coefficients (DCC) for the ICRP proposed RAPs are derived for the ERICA project and used in this report (see Section 4). Note that geometries used to derive the DCC are as specified in ICRP (2005), see Table 2.1. Internal DCC are relative to whole-body fresh weight activity concentrations, external DCC are relative to soil or sediment fresh weight or water activity concentrations as appropriate. The values presented here are those available from the ERICA project in October 2006. As the ERICA project will not be completed until March 2007, it is possible some values may change (see the ERICA Tool available from http://www.erica-project.org).

Aquatic RAPs (μGy h^{-1} per Bq kg^{-1} (fresh weight))

ICRP RAP	^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{228}Ra	^{228}Th	^{230}Th	^{232}Th	^{234}Th	^{234}U	^{238}U
Trout											
Internal	$3.4x10^{-4}$	$3.1x10^{-3}$	$2.5x10^{-4}$	$1.5x10^{-2}$	$3.5x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$5.0x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
External	$8.8x10^{-5}$	$4.3x10^{-9}$	$4.0x10^{-6}$	$9.2x10^{-4}$	$5.0x10^{-4}$	$8.3x10^{-4}$	$2.5x10^{-7}$	$1.5x10^{-7}$	$3.1x10^{-5}$	$1.6x10^{-7}$	$1.0x10^{-7}$
Duck											
Internal	See terrestrial animals										
External	$8.6x10^{-5}$	$4.2x10^{-9}$	$3.9x10^{-6}$	$9.0x10^{-4}$	$4.9x10^{-4}$	$8.1x10^{-4}$	$2.4x10^{-7}$	$1.4x10^{-7}$	$2.8x10^{-5}$	$1.5x10^{-7}$	$9.5x10^{-8}$
Brown seaweed											
Internal	$2.5x10^{-4}$	$3.1x10^{-3}$	$2.0x10^{-4}$	$1.4x10^{-2}$	$2.4x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$3.5x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
External	$1.8x10^{-4}$	$4.9x10^{-9}$	$4.7x10^{-5}$	$1.1x10^{-3}$	$6.0x10^{-4}$	$1.1x10^{-3}$	$6.7x10^{-7}$	$5.0x10^{-7}$	$1.7x10^{-4}$	$6.6x10^{-7}$	$5.1x10^{-7}$
Frog											
Internal	See terrestrial animals										
External	$1.1x10^{-4}$	$4.7x10^{-9}$	$1.1x10^{-5}$	$1.0x10^{-3}$	$5.5x10^{-4}$	$9.2x10^{-4}$	$4.1x10^{-7}$	$2.9x10^{-7}$	$6.3x10^{-5}$	$3.7x10^{-7}$	$2.7x10^{-7}$

Terrestrial RAPs (μGy h^{-1} per Bq kg^{-1} (fresh weight))

ICRP RAP	^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{228}Ra	^{228}Th	^{230}Th	^{232}Th	^{234}Th	^{234}U	^{238}U
Earth-worm											
Internal	$2.9x10^{-4}$	$3.1x10^{-3}$	$2.3x10^{-4}$	$1.4x10^{-2}$	$2.7x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$4.1x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
In soil	$8.0x10^{-5}$	$4.5x10^{-9}$	$6.0x10^{-7}$	$9.0x10^{-4}$	$5.0x10^{-4}$	$7.9x10^{-4}$	$2.1x10^{-7}$	$1.4x10^{-7}$	$1.1x10^{-5}$	$1.7x10^{-7}$	$1.2x10^{-7}$
Bee											
Internal	$2.6x10^{-4}$	$3.1x10^{-3}$	$2.1x10^{-4}$	$1.4x10^{-2}$	$2.5x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$3.4x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$3.0x10^{-5}$	$1.7x10^{-9}$	$2.9x10^{-7}$	$3.5x10^{-4}$	$1.9x10^{-4}$	$2.9x10^{-4}$	$7.2x10^{-8}$	$4.4x10^{-8}$	$4.6x10^{-6}$	$7.1x10^{-8}$	$5.0x10^{-8}$
Wild grass											
Internal	$2.9x10^{-4}$	$3.1x10^{-3}$	$2.2x10^{-4}$	$1.4x10^{-2}$	$2.6x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$4.0x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$2.9x10^{-5}$	$1.7x10^{-9}$	$4.0x10^{-7}$	$3.3x10^{-4}$	$1.9x10^{-4}$	$2.8x10^{-4}$	$1.4x10^{-7}$	$1.1x10^{-7}$	$4.7x10^{-6}$	$1.4x10^{-7}$	$1.0x10^{-7}$
Pine tree											
Internal	$3.8x10^{-4}$	$3.1x10^{-3}$	$2.5x10^{-4}$	$1.5x10^{-2}$	$5.7x10^{-4}$	$2.0x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$5.2x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$2.4x10^{-5}$	$1.4x10^{-9}$	$1.3x10^{-7}$	$2.7x10^{-4}$	$1.5x10^{-4}$	$2.3x10^{-4}$	$4.4x10^{-8}$	$2.1x10^{-8}$	$3.7x10^{-6}$	$1.8x10^{-8}$	$6.9x10^{-9}$
Rat											
Internal	$3.3x10^{-4}$	$3.1x10^{-3}$	$2.4x10^{-4}$	$1.4x10^{-2}$	$3.3x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$4.9x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$3.0x10^{-5}$	$1.7x10^{-9}$	$2.8x10^{-7}$	$3.4x10^{-4}$	$1.9x10^{-4}$	$2.9x10^{-4}$	$6.9x10^{-8}$	$4.2x10^{-8}$	$4.5x10^{-6}$	$6.7x10^{-8}$	$4.7x10^{-8}$
In soil	$7.6x10^{-5}$	$4.3x10^{-9}$	$5.2x10^{-7}$	$8.5x10^{-4}$	$4.7x10^{-4}$	$7.4x10^{-4}$	$1.8x10^{-7}$	$1.2x10^{-7}$	$1.1x10^{-5}$	$1.5x10^{-7}$	$1.0x10^{-7}$
Deer											
Internal	$3.8x10^{-4}$	$3.1x10^{-3}$	$2.5x10^{-4}$	$1.5x10^{-2}$	$6.1x10^{-4}$	$2.0x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$5.2x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$1.6x10^{-5}$	$8.6x10^{-10}$	$7.7x10^{-8}$	$1.8x10^{-4}$	$9.7x10^{-5}$	$1.6x10^{-4}$	$2.5x10^{-8}$	$1.3x10^{-8}$	$2.2x10^{-6}$	$1.7x10^{-8}$	$1.0x10^{-8}$
Frog											
Internal	$3.2x10^{-4}$	$3.1x10^{-3}$	$2.4x10^{-4}$	$1.4x10^{-2}$	$3.0x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$4.6X10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$3.0x10^{-5}$	$1.7x10^{-9}$	$2.8x10^{-7}$	$3.4x10^{-4}$	$1.9x10^{-4}$	$2.9x10^{-4}$	$7.1x10^{-8}$	$4.3x10^{-8}$	$4.6x10^{-6}$	$6.9x10^{-8}$	$4.8x10^{-8}$
In soil	$7.9x10^{-5}$	$4.5x10^{-9}$	$5.8x10^{-7}$	$8.9x10^{-4}$	$4.9x10^{-4}$	$7.8x10^{-4}$	$2.0x10^{-7}$	$1.4x10^{-7}$	$1.1x10^{-5}$	$1.7x10^{-7}$	$1.2x10^{-7}$
Duck											
Internal	$3.4x10^{-4}$	$3.1x10^{-3}$	$2.5x10^{-4}$	$1.5x10^{-2}$	$3.6x10^{-4}$	$1.9x10^{-2}$	$2.7x10^{-3}$	$2.3x10^{-3}$	$5.0x10^{-4}$	$2.8x10^{-3}$	$2.4x10^{-3}$
On soil	$2.8x10^{-5}$	$1.6x10^{-9}$	$2.6x10^{-7}$	$3.2x10^{-4}$	$1.8x10^{-4}$	$2.7x10^{-4}$	$6.5x10^{-8}$	$3.9x10^{-8}$	$4.3x10^{-6}$	$6.3x10^{-8}$	$4.3x10^{-8}$

Appendix B.2: Concentration ratios

Uranium and thorium concentration ratios are derived by the ERICA project and used in this report (see Section 4). Only those values used in this work are presented. The values presented here were available from the ERICA project in October 2006. As the ERICA project will not be completed until March 2007, it is possible some values may change (see the ERICA Tool available from http://www.erica-project.org).

ERICA reference organism	ICRP RAPs	Uranium	Thorium
		Bq kg^{-1} (FW) whole-body: Bq kg^{-1} media*	
Grasses and herbs	Wild grass	$1.46x10^{-2}$	$4.37x10^{-2}$
Tree	Pine tree	$6.79x10^{-3}$	$1.08x10^{-3}$
Terrestrial invertebrate	Earthworm	$8.84x10^{-3}$	$8.84x10^{-3+}$
Mammal	Deer & rat	$1.06x10^{-4}$	$1.22x10^{-4}$
Terrestrial bird	Duck	$4.98x10^{-4}$	$3.89x10^{-4}$
Pelagic fish	Trout	$3.00x10^{+1}$	$1.10x10^{+2}$

*For all organisms other than pelagic fish, the media concentration is defined as dry weight activity concentration in soil; media concentrations for pelagic fish are those in water.
$^{+}$Given the lack of data for the transfer of Th to terrestrial invertebrates, the ERICA default value is assumed to be the same as that for U.

Appendix C.1: Further detail on manipulation of G-BASE data

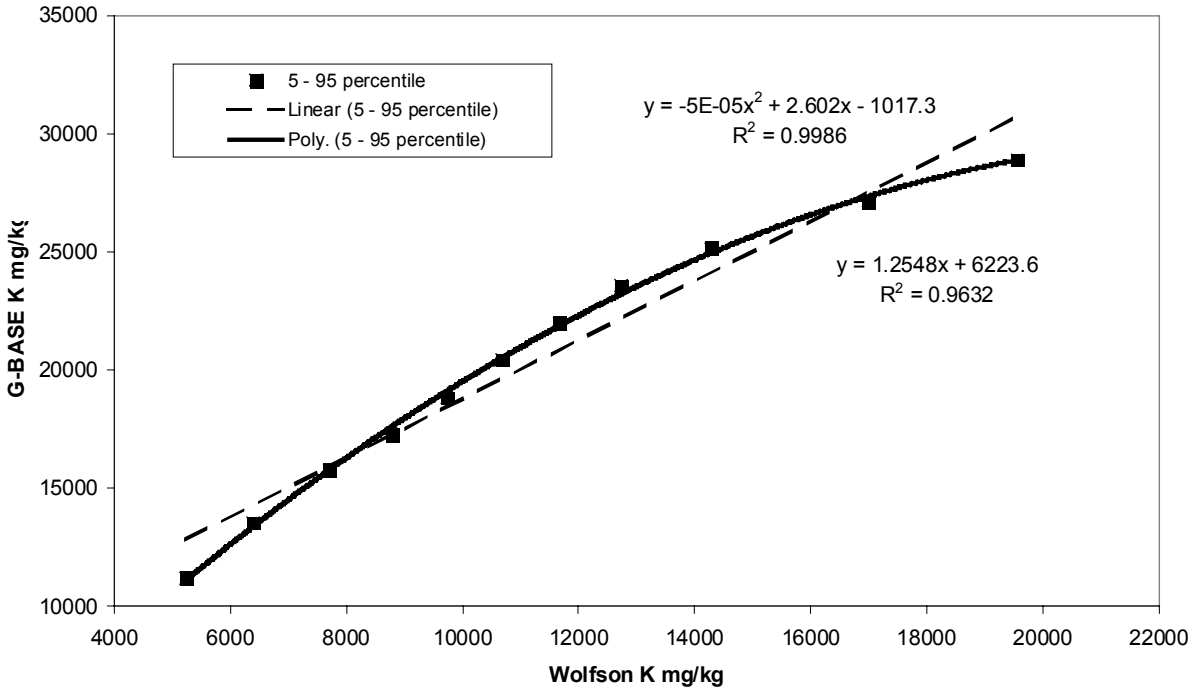

Figure C.1: Linear quantile transformation of Wolfson potassium in stream sediment to G-BASE XRF equivalent potassium in stream sediments (5 - 95 percentile range; data from this study)

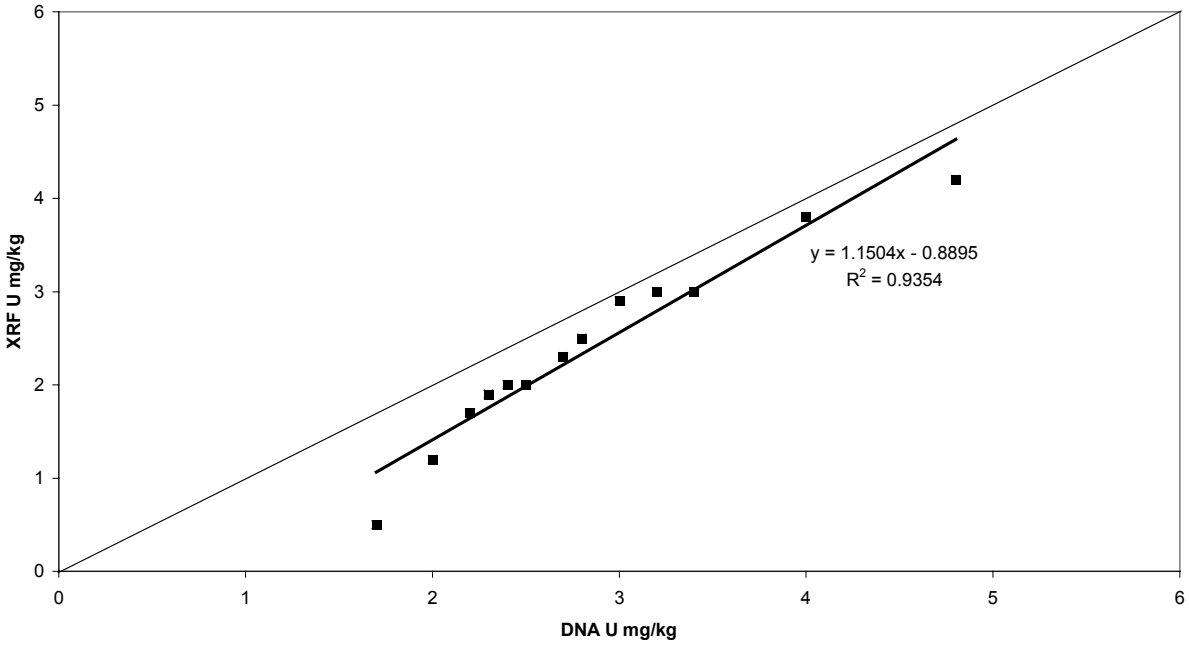

Figure C.2: Linear quantile transformation of DNA uranium in stream sediment to G-BASE XRF equivalent uranium in stream sediments (transformation applied only up to 6 mg kg^{-1})

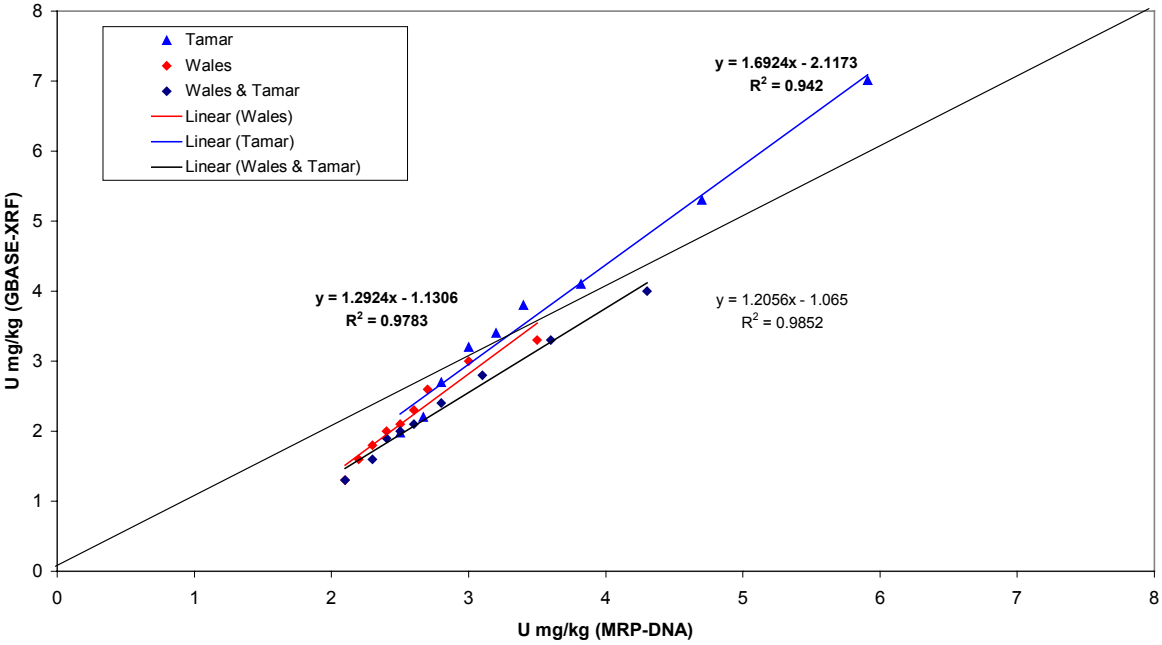

Figure C.3: Linear quantile plot of MRP-DNA uranium in stream sediment and G-BASE-XRF uranium in stream sediments (quantiles less than 20 percentile not plotted as LoD for most of XRF data is 1 mg kg^{-1}; values below this all given a nominal concentration of 0.5 mg/kg). The combined Wales and Tamar data gave a very similar relationship to that derived from the much larger G-BASE data comparison, so the G-BASE transformation was also applied to the MRP data.

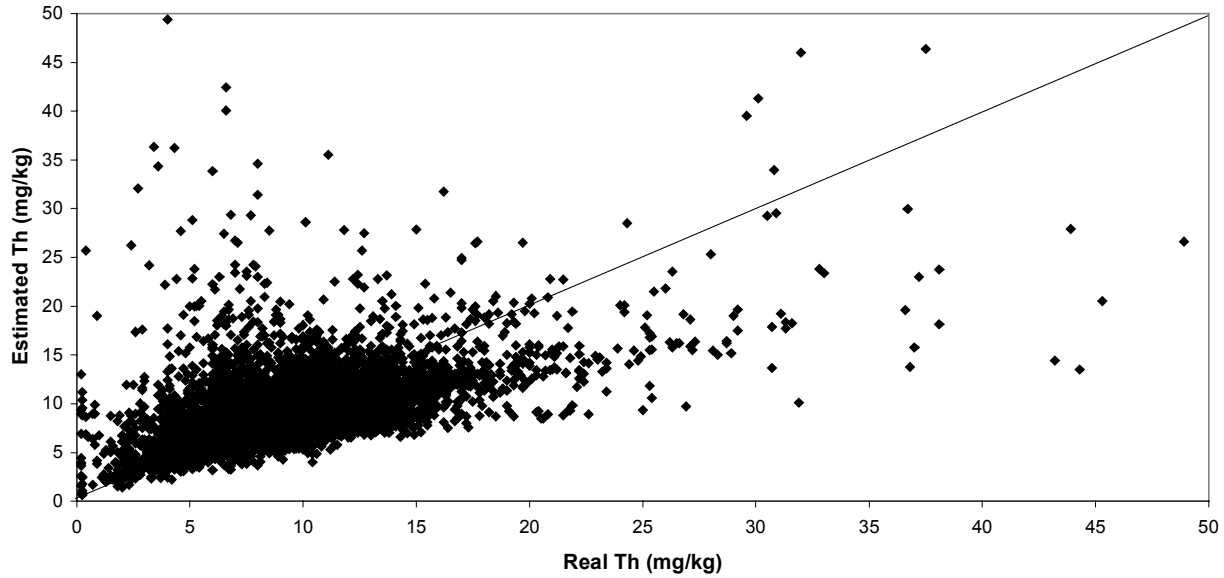

Figure C.4: Comparison of actual (real) and estimated Th values for stream sediments from Wales. The estimated values were derived by multiple linear regression using La, Y and Zr data compared with Th.

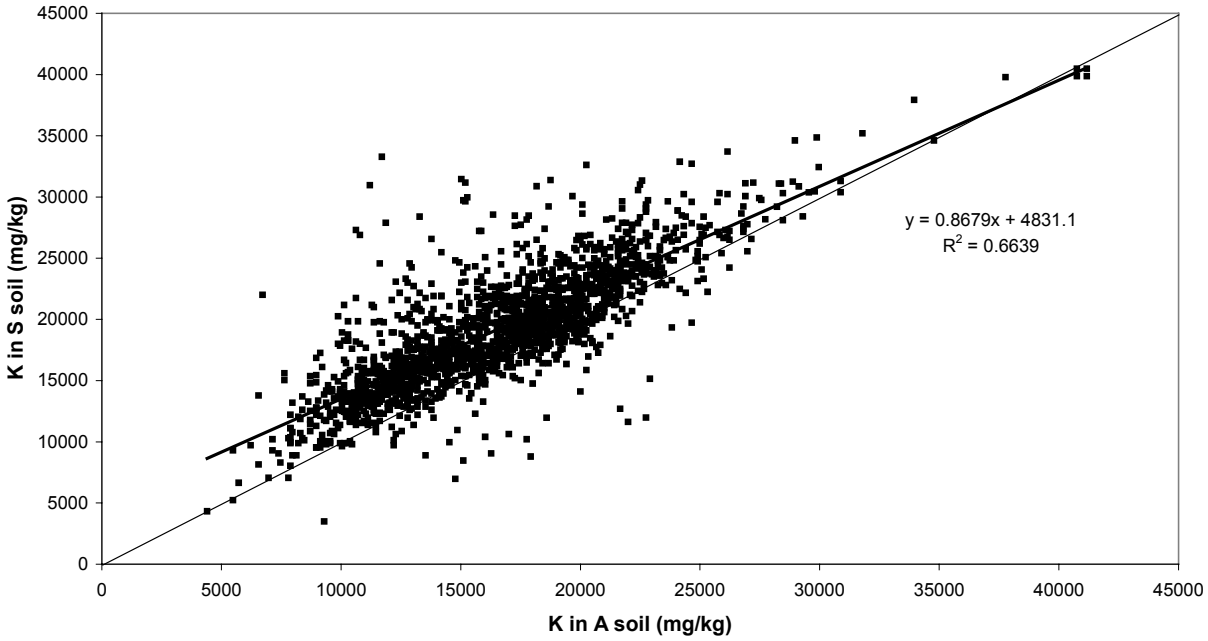

Figure C.5: Comparison of K content of surface (A horizon) and subsurface soils from urban G-BASE sampling in Coventry, Leicester and Stoke. The subsurface soils have generally higher levels of K and the regression was used to convert the surface soil data to estimated subsurface equivalents. The method involves plotting the matching percentiles of each dataset, assuming the data distribution should be similar when the same types of samples are collected from the same area. Thus, the 5th percentile of one dataset is plotted against the 5th percentile of the other dataset and the relationship used to transform one dataset to as closely match the other as possible.

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