

OR/17/001 Geogenic radionuclides in British groundwater

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Introduction

Under natural conditions, the radioactivity of groundwater is produced dominantly by radionuclides in the uranium and thorium decay series. Contributing solutes include uranium, radon and radium. ⁴⁰Potassium also contributes radioactivity from subsurface radioactive decay. Data for naturally-occurring radionuclides in British groundwater are very limited, but vary depending on specific regional studies or their application and utility as tracers. Activities of ²²²Rn and ²²⁶Ra have been investigated as indicators of short-term groundwater residence time or where health concerns have been raised. A limited number of uranium radionuclide studies have also investigated ²³⁴U/²³⁸U activity ratios as indicators of groundwater flow paths, reactions and residence times.

In the absence of data for individual radionuclides, total-element analyses can give indications of occurrence of their respective radionuclides in groundwater, assuming equilibrium proportions. This section describes the distribution and occurrence of naturally-occurring radionuclides of environmental relevance in British groundwater (uranium, radium, radon) where data exist. In the absence of individual radionuclide data, whole-element analyses for potassium, thorium and uranium are described. These latter have been compiled from BGS databases acquired from a number of regional and national baseline studies (e.g. Ó Dochartaigh et al., 2011^[1]; Shand et al., 2007^[2]). Summary statistical data (mean, percentiles) for these elements are provided for various aquifers across Britain, calculated using non-parametric Kaplan Meier (KM) or semi-parametric robust ROS (random on order statistics) methods, depending on percentage of values above detection limits (KM for <50% non-detects). Where >80% of values are non-detects, only observed ranges are given.

Potassium

Occurrence and controls

Potassium is an alkali metal with three naturally-occurring isotopes: ³⁹K (93.3%), ⁴⁰K (0.0117%) and ⁴¹K (6.7%). ⁴⁰K has a half-life of 1.25 x 10⁹ years.

Principal silicate mineral sources of potassium include feldspars and clays. Evaporite minerals may also include potassium salts (e.g. sylvite, polyhalite). Feldspars and clays are ubiquitous rock-forming minerals; occurrences of potassic evaporites in Britain are limited to the Whitby area of North Yorkshire.

Potassium is one of the eight principal ions in groundwater. Nonetheless, dissolved concentrations are limited and fresh groundwater usually has concentrations below around 10 mg/L. Potassium is released to solution substantially by weathering of silicate minerals but its transport is strongly limited by incorporation into secondary clays. Potassium is also a nutrient with an important role in the biosphere. Release of K from agricultural soils occurs via vegetation decomposition, soil leaching

and runoff. Wood ash (potash) is also an enriched source (Hem, 1992^[3]).

Concentrations in groundwater

Concentrations of potassium in British groundwater are typically in the range 1–10 mg/L, but with occasional observed maxima of 100 mg/L or more. Median values are around 2–4 mg/L (Table 2, Figure 34). Anomalously high observed concentrations may be a feature of saline groundwater or water affected by agricultural pollutants. Among the highest concentrations appear to occur within groundwater from the Crag aquifer of East Anglia. These are moderately mineralised with around half in the BGS database having electrical conductance of greater than 1000 $\mu\text{S}/\text{cm}$ (Ander and Shand, 2005^[4]).

Table 2 Statistical summary data for potassium (K) in groundwater from various aquifers in Great Britain (BGS databases)

Stratigraphy	n	ncens	Min	mean	Max	P5	P10	P25	P50	P75	P90	P95
Ashdown and Hastings Beds	8	0	1.34	2.84	11.1	1.34	1.34	1.36	1.56	1.7	5.11	8.09
Bridport Sands	52	0	0.4	2.41	10.5	0.6	0.90	1.40	2.00	3.1	4.1	4.7
Carboniferous	80	0	0.64	8.2	92.2	1.16	1.36	2.43	4.78	8.58	12.7	25
Carboniferous Limestone	223	12	<0.5	2.93	65.3	0.34	0.50	0.9	1.60	2.97	6.25	9.00
Chalk	736	2	<0.4	4.49	175	0.7	0.92	1.3	2.10	3.84	7.8	13.7
Coal Measures	12	0	0.88	5.95	15.7	0.88	0.89	1.17	2.50	7.27	15.5	15.6
Corallian	62	0	0.5	5.23	80.4	0.72	1.1	1.45	2.41	4.8	9.6	11.9
Crag	81	2	<0.5	16.5	220	0.9	1.3	2.6	4.80	20.0	34.8	63.0
Fell Sandstone	3	0	2.08	2.22	2.33	2.08	2.08	2.08	2.26	2.30	2.32	2.32
Granite	197	6	<0.5	4.44	66.8	0.7	0.90	1.5	2.50	4.1	8.7	13.2
Igneous extrusive	13	0	0.52	1.92	10.9	0.52	0.58	0.75	0.850	1.8	2.42	5.80
Igneous intrusive	18	0	0.88	3.05	13.6	0.88	1.02	1.29	2.51	3.78	4.82	6.14
Lincolnshire Limestone	19	0	2.76	5.74	11	2.76	2.84	3.81	4.15	8.82	10.8	10.9
Lower Greensand	154	0	0.83	3.96	20	1.2	1.50	2.1	2.90	5.1	8.2	9.28
Lower Palaeozoic	115	16	<0.5	1.93	33.7	0.094	0.171	0.339	0.80	1.8	4.3	8.99
Magnesian Limestone	120	0	0.613	4.92	169	1.48	1.76	2.19	2.82	3.84	6.56	8.75
Millstone Grit	178	0	0.34	2.76	14.8	0.70	0.77	1.0	1.84	3.7	5.97	7.38
Old Red Sandstone	185	3	<0.5	4.13	201	0.60	0.89	1.47	2.10	3.2	5.44	9.3
Oolite	117	2	<0.5	2.84	71.5	0.40	0.6	1.2	2.06	2.8	3.7	5.3
Ordovician	8	0	0.517	1.63	4.06	0.518	0.518	0.759	1.22	1.52	3.23	3.64
Palaeogene	73	0	0.9	9.08	105	1.20	1.4	2.6	4.61	7.67	18.6	37.6
Permo-Triassic Sandstone	786	3	<0.6	5.47	136	1.60	1.9	2.47	3.50	5.2	8.8	13.0
Precambrian	37	0	0.44	2.14	6.27	0.72	1.02	1.22	1.73	2.63	3.84	5.95
Sand & gravel	18	0	1.25	7.86	49.1	1.25	1.49	3.18	5.54	7.81	13.0	18.4
Silurian	40	0	0.342	1.76	5.7	0.456	0.761	0.996	1.27	1.74	3.61	5.58
Tunbridge Wells Sand	6	0	0.85	12	66.5	0.85	0.85	0.87	1.08	1.59	34.0	50.3
Wealden Group	2	0	0.76	1.19	1.63	0.76	0.76	0.76	1.19	1.41	1.54	1.59

ncens: number of censored points (non-detects); P5-95: percentiles

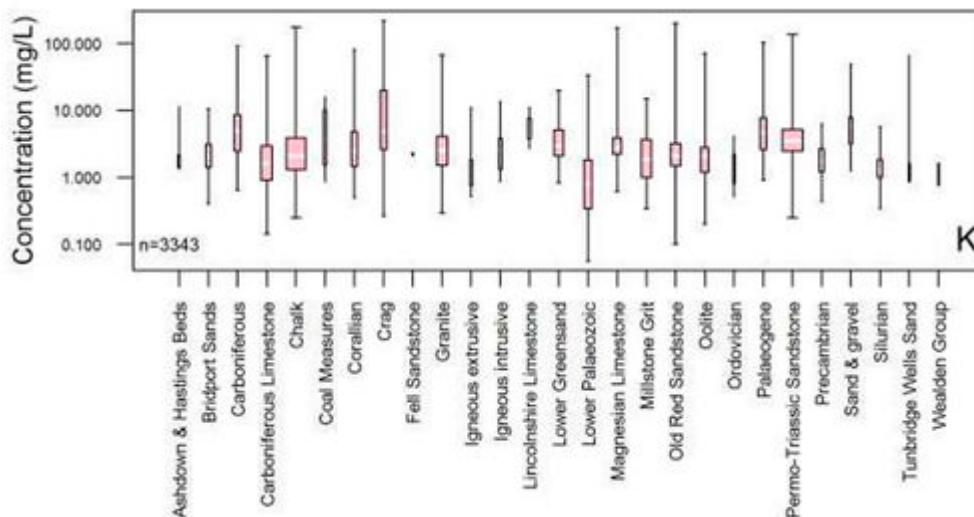


Figure 34 Boxplots showing potassium concentrations in groundwater from British aquifers; whiskers show the range of concentrations (box widths are proportional to the square root of the number of samples).

Radium

Occurrence and controls

Radioactive decay of U produces a number of daughter products, including radium (^{226}Ra). Radium comprises the naturally-occurring isotopes ^{223}Ra , ^{224}Ra , ^{226}Ra and ^{228}Ra , the most significant of which is ^{226}Ra . The half-life of ^{226}Ra is 1622 years.

Radium is an alkaline earth metal, behaving in the environment in a similar way to barium and strontium. It may be present in clay minerals, micas and metal oxides as well as barite and carbonate minerals. Greater Ra contents have been found in sediments with fine grain sizes than sand grade, suggesting the importance of clay-size minerals (Cuttell et al., 1988^[5]).

Radium has only one oxidation state, Ra(II), in natural systems. Radium compounds are relatively insoluble in water and so concentrations are usually very low, rarely exceeding a few Bq/L (Young et al., 2015^[6]). The element occurs in solution dominantly as free Ra^{2+} or as RaSO_4^0 and also forms complexes with carbonate (Wanty and Nordstrom, 1993^[7]). Mobility may be enhanced by alpha recoil but is also strongly limited by co-precipitation with barite or calcite, as well as by sorption reactions and ion exchange. Radium is known to be present in the mineral scales (calcite, barite) of groundwater systems with elevated uranium contents (Talbot et al., 2000^[8]). Sorption of Ra to metal oxides and clays is pH-dependent, being less significant under acidic conditions (e.g. Almeida et al., 2004^[9]). Higher concentrations of Ra have been found in acidic and anoxic groundwater conditions (USGS, 2016). Radium is also relatively mobile in saline groundwaters (Cuttell et al., 1988^[5]; Tomita et al., 2014^[10]). The mobility of ^{226}Ra influences the mobility of dissolved Rn. $^{226}\text{Ra}/^{238}\text{U}$ activity ratios of rock samples can be less than unity, indicating ^{226}Ra mobilisation relative to the ultimate parent ^{238}U (Cuttell et al., 1988^[5]).

Activity concentrations in groundwater

Very few data exist for radium activities in British groundwater. Cuttell et al (1988) reported activities of ^{226}Ra of 0.07–24 dpm/kg in groundwaters from the Permo-Triassic Sandstone of Merseyside (0.001–0.4 Bq/L). Talbot et al. (2000)^[8] analysed groundwater samples from Devon (Dartmoor Granite, Silurian–Carboniferous sedimentary rock aquifers) but reported all analyses

below the analytical detection limit of 0.1 Bq/L.

Radon

Occurrence and controls

Radon is a radioactive noble gas that exists naturally as three isotopes (^{222}Rn , ^{220}Rn , ^{219}Rn), derived from three radioactive decay chains. The most stable and environmentally relevant isotope of Rn, ^{222}Rn , forms from the alpha decay of ^{226}Ra , and ultimately from ^{238}U . ^{222}Rn is a short-lived isotope with a half-life of 3.8 days. During decay, an alpha particle is ejected, emitting radon in the process. This alpha recoil can alter crystal lattices and increase the leaching potential of Rn from the crystal surfaces (Wanty and Nordstrom, 1993^[17]). By the recoil process, Rn may be transported as a dissolved gas in porewater or retained in the mineral structure. Alpha recoil is likely an important mechanism for transferring Rn to groundwater and has been cited as a cause of radioactive disequilibrium in a number of groundwater studies (Wanty and Nordstrom, 1993^[17]).

As an inert gas, the mobility of radon in water is controlled by physical rather than chemical processes. Radon does not precipitate in mineral phases nor participate significantly in sorption reactions. Radon decays rapidly and does not accumulate in groundwater along flow paths because decay rates are typically faster than rates of groundwater flow. Hence, high Rn concentrations in groundwater are usually indicative of local parent (U, Ra) sources. Dissolved radon is lost from groundwater via release to the atmosphere as well as by radioactive decay.

The emanation efficiency of Rn (proportion of Rn released to groundwater compared to the total Rn produced) is a function of recoil and diffusion processes and is affected by Ra distribution in the host rock, surface geometry (e.g. rock specific surface area and roughness), surface wetness and rock porosity. The emanation efficiency in saturated aquifers is typically lower than 30% (Wanty et al., 1992^[11]). For a given U content of an aquifer host, Rn concentrations in groundwater are expected to be greater in aquifers of lower porosity, higher density or higher emanating efficiency (Wanty et al., 1992^[11]). The extent of transport from a Ra decay site is also determined by the permeability of the host rocks. In crystalline rocks, this is influenced by the extent and connectivity of fractures.

High Rn concentrations are typically not found in zones of high aquifer transmissivity as a result of lower rock/water ratios and dilution of emanating Rn (Lawrence et al., 1991^[12]). Inverse relationships have been found between ^{222}Rn concentration and well yield (Hall et al., 1987^[13]).

The solubility of Rn in water is temperature dependent, decreasing by about 3% per °C as temperature decreases from 20°C to 10°C. Barometric pressure and rainfall also have an influence, though less than other physical processes (Lindsey and Ator, 1996^[14]).

Temporal variations in Rn concentration have been observed in association with fluctuations in water level and groundwater pumping rates. The variation may be attributed to changing relative influences of different water-producing horizons (Lawrence et al., 1991^[12]).

In groundwater, although associated, the properties of Rn and U differ significantly. Radon is a dissolved inert gas and uranium a redox-sensitive metal with strong adsorption and complexation controls. Hence, although the two elements can be derived from a common geogenic source and can occur together in local groundwaters, they have very differing transport behaviour (Veeger and Ruderman, 1998) and may therefore become separated (Dowdall et al., 2013^[15]). The concentrations of uranium in groundwater are therefore not a reliable indicator of the concentrations of dissolved radon gas.

High Rn concentrations in groundwater are typically associated with U-rich rocks such as granite, granite gneiss and pegmatite (Banks et al., 1998a^[16]; Przylibski et al., 2004^[17]; Skeppström and Olofsson, 2007^[18]; Veeger and Ruderman, 1998^[19]; Vinson et al., 2009^[20]). In Britain, highest concentrations of Rn in the environment are associated with the Cornubian granite batholith of south-west England (Talbot et al., 2001^[21]) and the Grampian and Helmsdale areas of Scotland. High levels are also associated with the Carboniferous Limestone, Carboniferous shales of northern England, ironstone deposits (e.g. the Jurassic Northampton Sand), Palaeozoic mudrocks, and the Middle Old Red Sandstone of north-east Scotland (Appleton, 2005^[22]).

Activity concentrations in groundwater

The concentrations of Rn in groundwater typically vary over a large range, from around 3 Bq/L to 80 000 Bq/L (Appleton, 2005^[22]). Data for Rn in British groundwater are sparse. The most comprehensive recent review of available information is that carried out by Young et al. (2015)^[6]. They concluded that air radon was likely a good indicator of dissolved radon in groundwater. Mean values for dissolved Rn in groundwater from Denbighshire, Ceredigion and Anglesey in Wales were recorded as 30.2 Bq/L, 47.1 Bq/L and 24.8 Bq/L respectively, the former two designated as Radon Affected Areas in terms of atmospheric concentrations.

In areas of granitic rocks, ²²²Rn activities greater than around 50 Bq/L are not uncommon. Activities up to 5300 Bq/L have been found in granitic groundwater from south-west England (Young et al., 2015^[6]). In a Devon groundwater study, 9% of groundwater samples analysed for Rn were found to contain more than 1000 Bq/L (n=105) (Talbot et al., 2000^[21]). Mean activity for groundwater from the Dartmoor Granite was >700 Bq/L. Banks et al. (1998b)^[23] reported values in the range 44–200 Bq/L (median 140 Bq/L) from groundwaters in the granite of the Isles of Scilly. Activities of 40–76 Bq/L (mean 61 Bq/L) have also been reported in areas of granite and granite pegmatite in Aberdeenshire, Scotland. The same study found lower activities of 3–35 Bq/L (mean 23 Bq/L) in areas of metasedimentary rock (Al-Doorie, 1993^[24]).

In the Permo-Triassic Sandstone aquifer, Rn activities are also relatively high, though lower than in groundwater from granitic rocks. A range of 7.1–35.4 Bq/L was found in groundwaters from Merseyside (Cuttell et al., 1988^[5]), while activities of 5.8–36.6 Bq/L were found in the equivalent lithologies of Northern Ireland (Gibbons and Kalin, 1997^[25]). Values up to 50 Bq/L have also been found for Triassic sandstone groundwater from Lancashire (BGS, unpublished data, 2016). Young et al. (2015)^[6] reported activities up to 1350 Bq/L in undifferentiated Upper Devonian rocks from Great Britain, presumably including the red-bed Old Red Sandstone aquifer. In Chalk groundwater, activities <10 Bq/L are more typical. In the Chalk aquifer of south-east England, radon activities in the range 3.5–9 Bq/L were found in public-supply boreholes and 3.8–10.5 Bq/L in groundwater from springs (Low, 1996)^[26].

The investigation by Young et al. (2015^[6]) summarised groundwater data from databases held by BGS and Public Health England. A number of aquifers were represented, providing maximum observations of: Teign Valley Group (657 Bq/L), Hawick Group (57 Bq/L), Holsworthy Group (353 Bq/L), Dinantian volcanics (103 Bq/L), Ordovician volcanics (43 Bq/L), Palaeogene intrusions (64 Bq/L), Gala Group (35 Bq/L), Bridport Sand (10 Bq/L) and Gault (10 Bq/L) (Young et al., 2015^[6]). Ranges of ²²²Rn activity reported for groundwater (wells, boreholes) from local authorities/water companies were in the range 0.01–382 Bq/L (mean 9.9 Bq/L, n=1561, England & Wales only). Highest values were from igneous intrusions and Caradoc, Pridoli and Triassic aquifers (Young et al., 2015^[6]).

Thorium

Occurrence and controls

The principal isotope of thorium is ^{232}Th , with a half-life of 1.39×10^{10} years. Thorium is around three times more abundant in the earth's crust than uranium, but it is much less soluble in water and its concentrations are therefore usually low. It is commonly undetectable and so data for thorium in groundwater are limited.

Thorium occurs in the tetravalent (Th(IV)) state (Hem, 1992^[3]). The element's occurrence as a major constituent in minerals is restricted to thorianite and thorite (Langmuir and Herman, 1980^[27]). Thorium also occurs in the zircon, monazite and uraninite. The low solubility of these minerals is a major factor in the low abundance of Th in groundwater. It is present as a trace constituent in phosphate, oxide and silicate minerals and sorbs to clays. Thorium tends to be more concentrated in sediments with fine grain sizes (Cuttell et al., 1988^[5]).

Thorium in solution is invariably complexed, dominant ions being $\text{Th}(\text{SO}_4)_2^0$, ThF_2^{2+} and $\text{Th}(\text{HPO}_4)_2^0$. Complexation can increase the mobility of Th by some orders of magnitude (Langmuir and Herman, 1980^[27]). Complexing is more important at $\text{pH} < 7$. Thorium mobility is likely most strongly controlled by slow mineral dissolution kinetics and by sorption (Langmuir and Herman, 1980^[27]). Sorption of thorium to clays, metal oxides and organic matter increases with increasing pH, and is practically complete at pH 6.5 (Langmuir and Herman, 1980^[27]).

Concentrations in groundwater

Available BGS data for thorium concentrations in British groundwater are in the range <0.001 – 1.85 $\mu\text{g/L}$ (Table 3, Figure 35). The highest recorded observations occur in groundwater from the Permo-Triassic Sandstone and Chalk aquifers. These also had the largest ranges (and numbers of samples). Median and mean values (where calculable) across the aquifers are <0.03 $\mu\text{g/L}$.

Few other data are available for thorium in the literature. A range of 0.4 – 0.5 $\mu\text{g/L}$ was determined in groundwater from granite in Altnabreac, Caithness (Ivanovich and Kay, 1983^[28]). Talbot et al. (2000)^[8] reported Th concentrations in the range 0.02 – 0.066 $\mu\text{g/L}$ in groundwater from Dartmoor Granite of Devon.

$^{230}\text{Th}/^{234}\text{U}$ ratios of 0 – 0.037 were reported by Cuttell et al. (1988)^[5] from the Permo-Triassic Sandstone aquifer of Merseyside.

Table 3 Statistical summary data for thorium (Th) in groundwater from various aquifers in Great Britain (BGS databases)

Stratigraphy	n	ncens	Min	mean	Max	P5	P10	P25	P50	P75	P90	P95
Ashdown and Hastings Beds	8	8	<0.005	<0.005								
Bridport Sands	52	48	<0.016	0.0789								
Carboniferous	78	55	<0.001	0.0238	0.979	0.00016	0.00030	0.000844	0.003	0.00995	0.027	0.083
Carboniferous Limestone	100	91	<0.005		0.23							
Chalk	305	269	<0.001		1.47							
Coal Measures	12	8	<0.005	0.0114	0.07	0.00036	0.00051	0.00113	0.00319	0.0138	0.0199	0.0425
Corallian	53	52	<0.005		0.026							
Crag	18	18	<0.05		<0.05							
Fell Sandstone	3	3	<0.005		<0.005							
Granite	25	24	<0.05		0.28							
Igneous extrusive	13	12	<0.005		0.006							

Igneous intrusive	18	18	<0.05	<0.05								
Lincolnshire Limestone	19	19	<0.05	<0.05								
Lower Greensand	62	60	<0.005	0.006								
Lower Palaeozoic	104	93	<0.05	0.27								
Magnesian Limestone	48	46	<0.005	0.0309								
Millstone Grit	27	26	<0.005	0.008								
Old Red Sandstone	141	99	<0.02	0.0291	0.13	0.0157	0.0179	0.0229	0.025	0.0312	0.0397	0.05
Oolite	58	55	<0.005	0.11								
Ordovician	8	8	<0.05	<0.05								
Palaeogene	62	59	<0.005	0.08								
Permo-Triassic Sandstone	265	246	<0.005	1.85								
Precambrian	37	34	<0.005	0.02								
Sand & gravel	18	14	<0.005	0.013	0.029	0.003	0.00332	0.00434	0.00697	0.025	0.0262	0.0276
Silurian	40	40	<0.05	<0.05								
Tunbridge Wells Sand	6	3	<0.005	0.0085	0.014					0.013		
Wealden Group	2	0	0.016	0.047	0.078	0.016	0.016	0.016				

ncens: number of censored points (non-detects); P5-95: percentiles

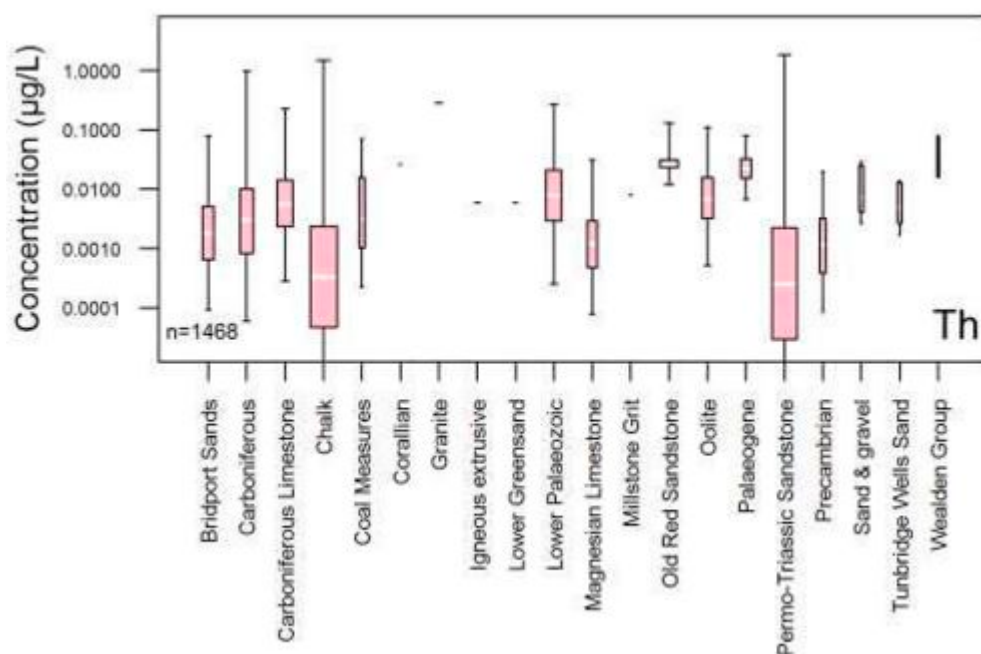


Figure 35 Boxplots showing thorium concentrations in groundwater from British aquifers; whiskers show the range of concentrations (box widths are proportional to the square root of the number of samples).

Uranium

Occurrence and controls

Natural uranium comprises three radioactive isotopes: ^{234}U , ^{235}U and ^{238}U , by weight constituting respectively about 0.0054%, 0.72% and 99.27%. Natural waters often display secular equilibrium between these isotopes but can depart from these mass proportions as a result of natural nuclear and chemical processes, leading to either enrichment or depletion of ^{234}U relative to ^{238}U . The theoretical mass ratio of $^{234}\text{U}/^{238}\text{U}$ (0.0000554) has been observed to vary from 0.00003 to 0.0014 in natural waters, with activity ratios in the range 0.5 to 40 relative to the secular equilibrium ratio of unity (Ivanovich and Harmon, 1982^[29]). High $^{234}\text{U}/^{238}\text{U}$ activity ratios (greater than unity) have been linked to preferential leaching of ^{234}U from minerals as a result of alpha decay causing recoil of ^{234}U

and ^{234}Th (Andrews and Kay, 1982^[30]), in turn causing damage to the crystal lattice (Gascoyne, 1992^[31]). High $^{234}\text{U}/^{238}\text{U}$ ratios have also been found in reducing aquifers close to redox boundaries, linked to redox transformations of uranium (Andrews and Kay, 1982^[30]; Bonotto and Andrews, 2000^[32]).

Uranium occurs in nature in two main oxidation states: the hexavalent form, U(VI), and the tetravalent form, U(IV). Uranium forms a major constituent of minerals such as uraninite, coffinite and autunite. These can be significant localised sources of uranium in some groundwaters, especially in ore mineralised areas and some granitic terrains. In Britain, occurrences of uranium mineralisation are relatively rare and usually below economic grade. Pitchblende-bearing mineral veins occur alongside Cu-Sn and Pb-Zn-Ni mineralisation around the Cornubian granite of south-west England. In Scotland, U mineralisation occurs in U-Pb mineral veins as well as phosphatic and carbonaceous horizons of the middle Devonian (Old Red Sandstone) of Caithness and Orkney. It also occurs in veins associated with granitic rocks (Helmsdale granite of Ousdale; Criffel-Dalbeattie granodiorite). Other occurrences in England & Wales are mainly associated with black shale deposits.

Uranium is also closely associated with the more common iron oxides, phosphates, clays and organic matter, and these minerals can be important sources, as well as sinks, of uranium. Iron oxides and clays are rock-forming minerals and are particularly important in iron-rich and argillaceous sediments and metasediments. Phosphate minerals, especially apatite and hydroxyapatite, are accessories in granitic and sedimentary rocks. The concentrations of uranium in rock-forming silicate minerals such as quartz and feldspar and carbonate minerals are usually low.

Iron oxides, including goethite, haematite, ferrihydrite and magnetite, have a strong affinity for uranium (Bianconi and Kögler, 1992^[33]; Casas et al., 1994^[34]; Duff et al., 2002^[35]; Scott et al., 2005^[36]). This is either attached via surface adsorption of the U(VI)O₂²⁺ uranyl ion, or incorporation of U(VI) within the structure of haematite and other iron oxides (Duff et al., 2002^[35]).

Additional anthropogenic sources of uranium in the environment can derive from industry (e.g. ceramics, chemicals, photographic and lighting industries) and agriculture. Uranium can be present (abundance around 20–200 mg/kg) in phosphate fertilisers (Zielinski et al., 2000^[37]) and may therefore be introduced to soils by agricultural activities. Enriched uranium (enriched in ^{235}U) is used in nuclear power plants and nuclear weapons, and its by-product, depleted uranium (relatively enriched in ^{238}U), has been used in military operations and the aircraft industry.

The mobility of U in water is controlled substantially by pH, redox status and concentrations of other coexisting solutes. Uranium is a redox-sensitive heavy metal that occurs in water principally under oxidic conditions in its hexavalent (U(VI)) state. It is usually complexed in solution, especially with carbonate ligands (Kohler et al., 2004^[38]; Villalobos et al., 2001^[39]). At alkaline pH, the uranyl ion forms stable complexes with carbonate ions, notably UO₂(CO₃)₂²⁻ and at higher pH, UO₂(CO₃)₃⁴⁻. Uranium complexes less significantly with phosphate, fluoride or sulphate, depending on their respective concentrations and ambient pH (Drever, 1997^[40]; Sandino and Bruno, 1992^[41]). Chloride and sulphate may become important ligands in saline waters (Porcelli and Swarzenski, 2003^[42]); U(VI) also forms stable complexes with dissolved organic carbon (Arey et al., 1999^[43]; Cothorn and Lappenbusch, 1983^[44]; Higgs et al., 1993^[45]).

Despite the relative mobility of U(VI) in the aqueous environment, the concentrations of U under oxidising conditions can be limited by adsorption onto phosphate minerals and iron oxides, as well as organic matter and clay minerals (Ivanovich, 1994^[46]). These minerals can therefore act as important sinks as well as potential sources. As many of the uranyl-carbonate complexes are anionic, sorption

is generally less strong at alkaline pH (>7). Sorption of U onto minerals is enhanced under pH-neutral conditions (Langmuir, 1978^[47]; Prikryl et al., 2001^[48]). Pabalan and Turner (1997)^[49] observed maximum sorption of U(VI) onto montmorillonite at pH 6.0–6.5.

Under anoxic conditions, U is reduced to its tetravalent U(IV) state and its concentration in water is low as a result of stabilisation of the sparingly soluble uraninite (UO₂), or adsorption to mineral surfaces such as iron oxides and clays. Equilibrium with uraninite limits uranium concentrations to less than 0.06 µg/L (Gascoyne, 1992^[31]; Langmuir, 1978^[47]). The reduction of U(VI) is facilitated by microbial activity (Davis et al., 2006^[50]; Fox et al., 2006^[51]; Lovley et al., 1991^[52]).

Concentrations in groundwater

Observed baseline concentrations of uranium in British groundwater span some four orders of magnitude, from <0.002 to 67 µg/L in the BGS databases. The observed maximum equates to 1.7 Bq/L assuming secular equilibrium. Median values are almost invariably 1 µg/L or less (up to around 0.026 Bq/L, same assumption; Table 4, Figure 36). Highest concentrations are observed in groundwater from red-bed sandstone aquifers (Old Red Sandstone; Permo-Triassic Sandstone). These high values occur in oxic groundwater conditions, the sources of uranium likely being iron oxides and/or phosphate minerals (Read et al., 1993^[53]) in the sandstone matrices. A similarly large range of concentrations was found in Permo-Triassic Sandstone groundwater from Merseyside. Ivanovich et al. (1992)^[54] reported concentrations of 0.3–13.6 µg/L (234U/238U activity ratios of 1.6–13.8). Cuttell et al. (1988)^[5] recorded 238U concentrations up to 42 µg/L in this aquifer. Fresh oxic groundwaters had 234U/238U activity ratios of around 1–5, as did fresh estuarine and saline groundwaters; flushed brackish groundwaters by contrast had activity ratios up to around 14.8, potentially linked to ion-exchange and desorption of U from iron oxides. Talbot et al. (2000)^[8] reported concentrations of uranium up to 11.6 µg/L in groundwater from Devon, the highest concentrations being from the Dartmoor Granite. Such high concentrations for the granite of south-west England were not recorded in the BGS database (Table 4). Relatively high U concentrations are also observed in the Carboniferous Limestone and igneous rock types, likely linked to the presence of authigenic phosphate minerals and uranium mineralisation. Lowest baseline concentrations are observed in the Lincolnshire Limestone (Jurassic) and Palaeogene. These groupings contain a high proportion of anoxic groundwaters from confined aquifers.

Table 4 Statistical summary data for uranium (U) in groundwater from various aquifers in Great Britain (BGS databases)

Stratigraphy	n	ncens	Min	mean	Max	P5	P10	P25	P50	P75	P90	P95
Ashdown & Hastings Beds	8	3	<0.002	0.0135	0.061				0.003	0.011		
Bridport Sands	52	7	<0.05	0.608	5.47			0.16	0.5	0.808	0.97	1.06
Carboniferous	78	8	<0.02	0.862	9.96	0.003	0.009	0.03	0.19	0.727	3.16	4.74
Carboniferous Limestone	100	7	<0.02	1.10	7.84	0.005	0.05	0.3	0.69	1.57	2.61	3.44
Chalk	446	50	<0.002	0.368	7.63	0.002	0.018	0.11	0.23	0.41	0.77	1.29
Coal Measures	12	2	<0.002	0.132	0.417			0.005	0.05	0.249	0.335	
Corallian	53	8	<0.02	0.43	7.46	0.006	0.006	0.07	0.21	0.4	0.89	1.22
Crag	18	5	<0.02	0.123	0.37				0.04	0.24	0.33	
Fell Sandstone	3	0	0.052	0.168	0.4	0.052	0.052	0.052	0.053			
Granite	25	0	0.02	0.983	3.57	0.04	0.2	0.29	0.55	0.88	2.97	3.56
Igneous extrusive	13	1	<0.2	1.42	13.3	0.02	0.03	0.04	0.23	0.84	1.94	
Igneous intrusive	18	1	<0.02	0.933	12.0		0.02	0.04	0.14	0.46	1.07	
Lincolnshire Limestone	19	13	<0.02	0.0753	0.4	0.0002	0.00033	0.0012	0.00635	0.03	0.374	0.391
Lower Greensand	99	70	<0.02	0.0688	0.81	0.0013	0.00213	0.0053	0.0172	0.0442	0.181	0.363
Lower Palaeozoic	115	56	<0.02	0.162	7.93	0.017	0.017	0.017	0.02	0.07	0.23	0.76

Magnesian Limestone	48	0	0.0211	1.65	8.08	0.373	0.833	1.13	1.53	1.86	2.45	2.59	
Millstone Grit	27	9	<0.02	0.191	1.87	0.008	0.008	0.008	0.04	0.29	0.49	0.69	
Old Red Sandstone	141	6	<0.005	2.71	48.0	0.033	0.11	0.29	1.01	2.29	5.36	8.4	
Oolite	58	10	<0.02	0.506	3.7			0.17	0.29	0.51	1.19	2.3	
Ordovician	8	1	<0.02	0.286	1.3			0.03	0.04	0.13			
Palaeogene	62	28	<0.005	0.306	4.65			0.005	0.02	0.09	0.495	2.38	
Permo-Triassic Sandstone	327	11	<0.02	1.94	67.2	0.04	0.18	0.392	0.9	1.93	4.38	6.86	
Precambrian	37	10	<0.02	0.806	6.19				0.09	0.9	3.03	4.98	
Sand & gravel	18	1	<0.005	1.29	6.11			0.009	0.027	0.342	1.07		
Silurian	40	5	<0.02	0.729	4.14			0.03	0.17	0.4	1.14	1.66	2.15
Tunbridge Wells Sand	6	0	0.013	0.0408	0.155	0.013	0.013	0.013	0.014	0.026			
Wealden Group	2	0	0.037	0.05	0.063	0.037	0.037	0.037					

ncens: number of censored points (non-detects); P5-95: percentiles

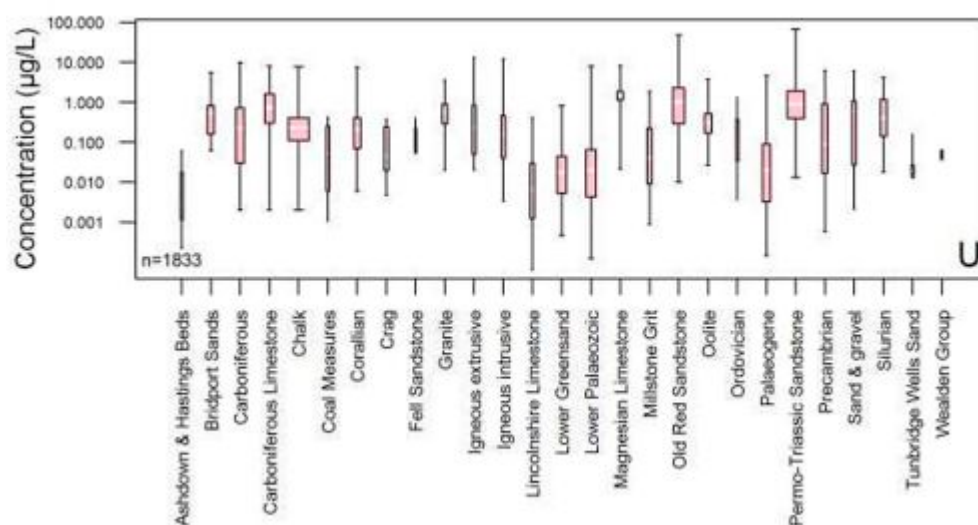


Figure 36 Boxplots showing uranium concentrations in groundwater from British aquifers; whiskers show the range of concentrations (box widths are proportional to the square root of the number of samples).

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