$^{87}Sr/^{86}Sr$ isotope composition of bottled British mineral waters for environmental and forensic purposes

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Abstract

Mineral waters in Britain show a wide range of $^{87}Sr/^{86}Sr$ isotope compositions ranging between $^{87}Sr/^{86}Sr = 0.7059$ from Carboniferous volcanic rock sources in Perthshire, to $^{87}Sr/^{86}Sr = 0.7207$ in the Dalradian aquifer of Aberdeenshire. The $^{87}Sr/^{86}Sr$ composition of the waters shows a general correlation with the aquifer rocks, resulting in the waters from older rocks having a more radiogenic signature than those from younger rocks. This wide range of values means that the Sr isotope composition of mineral water has applications in a number of types of studies. In the modern commercial context, it provides a way of fingerprinting the various mineral waters and hence provides a method for recognising and reducing fraud. From an environmental perspective, it provides the first spatial distribution of bio-available $^{87}Sr/^{86}Sr$ in Britain that can be used in modern, historical and archaeological studies.

Keywords

Sr-isotopes; mineral water; Britain; environment; provenance; forensic; archaeology**.**

Introduction

Sr as an environmental tracer

Sr isotopes are well established as a method of fingerprinting biosphere signatures and are increasingly used as a tool for human migration studies in archaeology; recent examples include Price *et al*. (2004), Montgomery *et al.* (2005), Hodell *et al*. (2004) but so far little has been published on their use as a forensic tool. Barbaste *et al*. (2002) demonstrated the use of the technique in provenancing and finger printing wine; ivory was traced using $^{87}Sr/^{86}Sr$ isotopes (van der Merwe *et al*., 1990) and Negrel *et al*. (2004) used them to trace river catchments and monitor the effect of agricultural pollutants. More recently, Sr was one of the key isotope tools used in tracing modern humans (Beard *et al*., 2000) and the origins of a murder victim (Pye, 2004).

The rapid expansion of bottled mineral water over the last decade has lead to bottled water being produced and available all over Britain. Such waters are purchased primarily for the perceived " purity" of the product but, because of the ready availability of water, the potential for fraud is considerable. Isotope fingerprinting should be able to provide a clear and precise Sr isotope ratio to mineral springs that can be used to help define the product.

Sr has four naturally occurring isotopes, three of which (^{84}Sr, ^{86}Sr, ^{88}Sr) are stable but ^{87}Sr is derived from the decay of ^{87}Rb. This means that the $^{87}Sr/^{86}Sr$ ratio of a rock is dependent upon the Rb content and age of the rock. As a result, younger, low-Rb rocks such as basalt will have whole rock $^{87}Sr/^{86}Sr$ values around 0.706 whereas older and more Rb rich rocks such as granites tend to have much higher values.

The Sr isotope composition of water has been used in many studies of water rock interaction including aquifer sources, groundwater pathways and pollution studies (Wickman and Aberg, 1987; Aberg, 1995; Negrel *et al*., 2004). Water equilibrates with the host rock and reflects the isotope composition of the minerals able to exchange with water. In the case of carbonates, this is likely to reflect the bulk rock isotope composition as these rocks are essentially homogenous with respect to Sr but, in silicates, the Sr released during weathering is dominated by mineral-specific isotopic compositions which means that the composition of the water may be very different from the whole-rock value of the silicate host rock (Aberg, 1995; Blum *et al.*, 1993; Jacobson and Blum, 2002; Bau *et al.*, 2004). Derry *et al.* (2006) looked at the isotope variation between minerals within a quartz diorite from Puerto Rico and found the following variations in $^{87}Sr/^{86}Sr$ between minerals: biotite $^{87}Sr/^{86}Sr = 0.7827$, feldspar $^{87}Sr/^{86}Sr = 0.7036$ and amphibole $^{87}Sr/^{86}Sr$ 0.7058. This resulted in the pore fluid of the overlying soil varying between *c.* 0.711 and *c.* 0.705 depending upon the proportion of biotite to feldspar and amphibole within certain soil horizons. Britain has a very varied geological structure and might be expected to yield a wide range of Sr isotope compositions within its mineral waters. However very little data are presently available from British aquifers and groundwaters (Shand *et al.*, 2001; Spiro *et al.*, 2001; Bain and Bacon, 1994).

The waters in this study

Only mineral waters with clear details of provenance were included in this study. The labels on the bottles clearly stated that the content was bottled at source and an address was given. The locality of the sample is based on the post-code of the bottling address. Many of these samples are recognised “Natural Mineral Waters” as defined by The Food Standards Agency. The requirements of a “Natural Mineral Water” are that: 1) it must come from a specified groundwater source, protected from all kinds of pollution; 2) it must be officially recognised after a qualifying period of two years; 3) it must be untreated and bottled at source; 4) the label must carry the proper description “Natural Mineral Water”, which cannot be used for any other types of bottled water; and finally, 5) the label must show the name of the recognised source and mineral content values. At the time of writing, ninety-nine waters from Britain were certified as “Natural Mineral Waters” by the Food Standards Agency.

Many wells that are now exploited on a commercial basis, such as St Anne’s Well in Buxton, Derbyshire, have a long documented history of use and it is highly likely they were also a source of drinking water in prehistoric times. Some were believed to be endowed with healing properties (e.g. Burnett 1925; Whelan and Taylor 1989) while others would have simply been a source of clean water. This means that the analysis of water from these wells not only gives us a useful forensic tool but it is also of use in environmental studies as it provides a direct measurement of a dietary component of ancient people that is free of modern contaminants.

Method

The seal on the bottled water was broken in the clean laboratory and 20 mls of each sample was measured into pre-cleaned Teflon beakers. Each sample was mixed with ^{84}Sr tracer solution, acidified using 1ml of 6M Teflon distilled HCl, and evaporated to dryness. Sr was collected using conventional Dowex© resin ion exchange methods.

Sr isotope composition and concentrations were determined using a 262 Finnigan MAT multi-collector mass spectrometer using TaF activator on single rhenium filaments (after the method of Birck, 1986). All samples were run to an internal precision of ± 0.000007 (1SE) or better. The international standard for ^{87}Sr/^{86}Sr, NBS987, gave values of 0.710207 ± 0.000030 (n=8, 2σ). All Sr ratios have been corrected to an accepted value for the standard of 0.710250. Sr blanks are *c.* 40 pg. The data and sample information are presented in Table 1.

Results

The results are presented on Figure 1 and the locations of the samples are given superimposed on the surface geology of Britain on Figure 2. The majority of the boreholes and wells from which these waters are drawn are between 50 and 200 m depth so that the water is drawn from aquifer rocks at or near the surface. From Figure 1 it can be seen that, in general, there is a trend for increasing ^{87}Sr/^{86}Sr signatures in the waters, with increasing stratigraphic age of the aquifer host rock. The results are discussed below in relation to the geology of their host aquifer. The absolute times given for each system are rounded to the nearest whole number and taken from Gradstein *et al.* 2004.

Cretaceous 145-66 Ma

The Chalk was deposited during Late Cretaceous times and records ^{87}Sr/^{86}Sr ratios between 0.7075 and 0.7078 (McArthur *et al*., 2001). “Hildon” water, from Hampshire, taps a chalk aquifer and has an ^{87}Sr/^{86}Sr ratio 0.70771, which shows it has equilibrated with the aquifer host rocks.

Jurassic 200-146 Ma

Carbonates formed between 178-160 Ma record low ^{87}Sr/^{86}Sr ratios between 0.7068 –0.7073 (McArthur *et al*., 2001). “Cotswold Spring Water” has a value of 0.70846, which is one of the least radiogenic waters but is, nevertheless, higher than its host aquifer carbonate value. This suggests that this water derives its Sr from an additional source, or sources, and may be influenced by other silicate bearing rocks, or silicate impurities within the limestone.

Triassic and Permian 292-200 Ma

The “Aqua Pure” and “Willow” water from Cumbria and “Swithland” from Leicestershire are all sourced from Triassic sandstones and provide an estimate for Triassic aquifers with values of 0.70963, 0.70959 and 0.71050 respectively. The Permian sourced water "Hadrian", from the Magnesian Limestone, has a similar value of 0.71026.

Carboniferous 359-200 Ma

Limestone

Three waters from Carboniferous Limestone aquifers were sampled: "Brecon Carreg" from south Wales; "Buxton" from Derbyshire; and "Findley's" from East Lothian. The East Lothian and Derbyshire waters were very similar in composition, giving 0.71037 and 0.71018 respectively but the Welsh water was considerably lower at 0.70926. All these waters are more radiogenic that the seawater values during the Lower Carboniferous, which ranged in Sr isotope composition between 0.7075 and 0.7082 (McArthur *et al*., 2001). The results suggest that Derbyshire and East Lothian waters have a greater contribution from silicates sources than the Welsh aquifer.

Millstone Grit and Culm Measures

Eight waters have been measured from boreholes and springs in Carboniferous sandstones. Three samples come from the Culm of Devon: "Tarka Spring"; "Exeter"; and "Devon Hills". The first two come from the same spring and give the same $^{87}Sr/^{86}Sr$ value of 0.71250 and "Devon Hills" gives a similar value of 0.71257. These Devon waters giving *c.* 0.7125 can be distinguished from the more northerly Millstone Grit hosted waters which are slightly less radiogenic. Cheshire and Derbyshire, represented by "Brittannia" and "Grindleford", give 0.71192 and 0.71194 respectively; Northumberland and West Yorkshire represented by "Abbey Well" and "Pennine Spring" respectively, give 0.71142 and 0.71126; and the lowest value comes from the "Harrogate Spa" water in North Yorkshire with 0.71103. The Carboniferous is deposited in a number of basins across England and Wales which have differing sediment sources (Chisholm *et al*., 1996; Leng *et al*., 1999) and these results suggest that the basins may be identifiable in terms of their leachable components that exchange to give the mineral waters their distinguishable values.

Volcanic rocks

The lowest $^{87}Sr/^{86}Sr$ ratio so far found in a mineral water comes from Perthshire from the "Caledonian" water which taps an aquifer in Carboniferous volcanic rocks. Basic rocks with low Rb/Sr ratios and mantle origin tend to have low $^{87}Sr/^{86}Sr$ ratios and despite being of Carboniferous age have generated very little radiogenic Sr. Hence, the exchangeable Sr from the volcanic rocks give this mineral water an unradiogenic $^{87}Sr/^{86}Sr$ vale of 0.70587.

Devonian 416-359 Ma

Sandstones

The "Edgemoor" (Devon) and "Styles" (Somerset) waters have very similar values of 0.71202 and 0.71210 whereas the Scottish "Strathmore" has a comparatively low value of 0.70972. This suggests that the Devonian basins of England and Scotland have significantly different detritus sources and that the exchangeable Sr within the detrital minerals originates from different sources.

Volcanic rocks

"Highland Spring" is sourced in Devonian Volcanic rocks and, like the nearby "Caledonian" water, has a very low $^{87}Sr/^{86}Sr$ ratio of 0.70659. The result is close to values recorded in river water (0.7080 ± 0.00005 SD, n=12) that drains the Devonian andesites in the Sourhope valley near Kelso in the Scottish borders (Bain and Bacon, 1994).

Silurian 444-416 Ma

All five Silurian based waters in this study come from Wales. Four are from the Churchstoke ("Springbourne", "Montgomery Spring" and "Fairbourne") and Knighton ("Radnor Hills") areas of east central Wales in Powys and one (“Ty Nant”) from Cardiganshire in west central Wales near Aberystwyth. With the exception of the “Radnor Hills” water the results group tightly, with ^{87}Sr/^{86}Sr of 0.71169, 0.71198, 0.71172 and 0.71210 respectively. The values are close to the initial ratio of Silurian mudrocks as determined during Rb-Sr whole rock dating (Evans, 1989; 1996). The “Radnor Hills” water has a more radiogenic signature of ^{87}Sr/^{86}Sr = 0.71393.

Ordovician. 488-444 Ma

One Ordovician hosted water was sampled: “Lakeland” from Cumbria. It gave a ^{87}Sr/^{86}Sr value of 0.71385. The value is similar to the ^{87}Sr/^{86}Sr initial ratio of Welsh Ordovician mudrocks (Evans, 1989) and close to Welsh river water composition from the Plynlimon area that drain Ordovician rocks and give values up to 0.714 (Shand *et al.*, 2001)

Precambrian of England and Wales > 542 Ma

“Malvern” water is taken from the springs that rise in the Precambrian outcrops of the Malvern Hills in Worcestershire. It has a radiogenic signature of 0.71329, which is in keeping with the old age of its source rocks. However, “Church Stretton” mineral water is also sourced in Precambrian rocks of the Longmynd but it has a much less radiogenic signature than “Malvern Water”. This is probably because the Longmynd is derived from the erosion of similar-aged volcanic rocks such as the Uriconian which is known to be relatively unradiogenic (Thorogood, 1990). Consequently, the resulting ^{87}Sr/^{86}Sr value of 0.71033 is not unusual despite the considerable age (>600Ma) of the host rock.

Precambrian of Scotland >542 Ma

In contrast to the Welsh/English basement, Scotland has a long geological history going back *c.* 2300 million years and this starts to be seen in some of the most radiogenic waters found in this study. Dalradian migmatites that form the aquifer rock for “Deeside” water give the water a $^{87}Sr/^{86}Sr$ composition of 0.71463 and the Dalradian Glenfiddich Pelite Formation sequence endow the “Speyside Glenlivet” water with the most radiogenic signature in this study of 0.72065. Values close to this (average = 0.7184, n=16) have been reported from streams draining Cambrian metamorphic rocks and Dalradian schists (Bain and Bacon, 1994) and such values are comparable with waters reported from areas of geologically old terrains such as Scandinavia (Aberg, 1995), central Australia (Harrington and Herczeg, 2003), and Brittany and the Massif Central, France (Negrel *et al*., 2000).

Discussion

The implications for fingerprinting mineral waters

The results from this study show that the spring and mineral waters from Britain have a wide range of $^{87}Sr/^{86}Sr$ isotope compositions (0.7059 – 0.72), which indicates that this method provides a good approach to fingerprinting many bottled waters. Although not all the waters have a unique $^{87}Sr/^{86}Sr$ isotope signature there is a general overall trend showing that the younger rocks give lower, less radiogenic $^{87}Sr/^{86}Sr$ ratios whereas the water from older rocks give the high values (Figure 2) and this is consistent with the higher levels of ^{87}Sr due to the longer decay time.

The exception to this trend comes from aquifers in basic volcanic rocks, such as “Highland Spring” and “Caledonian” water, where the rubidium-poor nature of the host rocks means that even the older, Palaeozoic, horizons yield low $^{87}Sr/^{86}Sr$ ratios that fall outside the general trend of data. The Carboniferous rocks also show a systematic difference in $^{87}Sr/^{86}Sr$ values between the limestone and Gritstone hosted waters, the latter being more radiogenic. In order to extend this technique as a commercial fingerprinting methodology it is essential that the variation within wells through the year is monitored to provide the range of values associated with a particular site. The identical value from the “Exeter” and “Tarka Springs” which were bought at different times but come from the same well suggests that such variations may be negligible particularly for sources that tap older aquifer waters, but rapid recharge or over use of supply may lead to some variation through time.

The dietary contribution of water

The Sr incorporated into human body tissues mainly derives from ingested Sr that is absorbed in the digestive tract (ATSDR, 2004). However, not all dietary components will contribute equally to body fluids due to complex synergisms and antagonisms with other dietary components such as calcium, fibre and protein, which ensure that there is no simple direct transfer of Sr from the diet to the skeleton. (Elias, 1980; Elias *et al*., 1982; Burton and Wright, 1995; Burton *et al.*, 2003). Consequently, the Sr isotope composition of tooth enamel, which is increasingly used as a migration tool in archaeological studies, will be a weighted average of the ^{87}Sr/^{86}Sr composition of Sr absorbed through the gut and circulating in body fluids during the mineralization phase of the tooth during childhood.

The daily intake of Sr is approximately 3.3 mg per day of which 2 mg/day is drinking water and 1.3 mg per day is food (ATSDR, 2004). This is based on the assumption that an adult human drinks 2 L of water a day with a Sr concentration of 1 mg/L, and modern dietary uptake of Sr is 1.3 ppm per day (Ysart *et al*., 1999). The data suggest that the assumption that water has a concentration of 1 mg/L could be an overestimate as an average concentration of 0.2 ppm is recorded from the waters of this study. However, similar overall Sr intakes have been calculated by Varo *et al*. (1982), 1.9mg/day; Beliles (1994), 2 mg/day; and Shiraishi *et al.* (1989), 2.3 mg/day (for adults. The contribution from water to children’s diet is likely to be higher that that of adults as they consume more fluids (ATSDR 2004). Such figures show that the Sr isotope composition of water is an important component in determining the isotope composition of ingested Sr, although the uptake of Sr through the gut is controlled by a number of factors, which may mean that the uptake ratios from these dietary components differ from their intake ratios. This makes it important to understand the contribution and composition of water in a diet both for archaeological and forensic studies. Water is unlikely to have been a commodity that was transported and traded long distances in the past. In modern times this situation has changed and bottled water from around the world is available in Britain. However, the quantity of bottled water we drink is generally small in comparison to our overall water (*c*.2 litres/day, Ysart *et al*., 1999) intake and so, despite the modern "supermarket" diet phenomenon, the Sr contribution to modern diets from water is likely to be dominated by local sources.

Conclusion

Mineral waters in Britain display a wide range of ^{87}Sr/^{86}Sr ratios that broadly reflect the age and lithology of their host aquifers. Such variation suggests that ^{87}Sr/^{86}Sr isotope signatures could potentially be used to fingerprint and distinguish between the mineral waters as a tool against fraud.

As water is a vital component of the diet that is frequently sourced locally, the spatially distributed, geologically-related data, presented here, provide a geographic map of dietary ^{87}Sr/^{86}Sr values. Moreover, given the poor survival rate of archaeological bone in the acidic soils hosted by many silicate and basaltic rocks, these results help to define possible places of origin, unaffected by modern contamination, for archaeological individuals who are not consistent with the carbonate rocks in which they are predominantly found. Mineral water data has the potential to contribute to our understanding of population ^{87}Sr/^{86}Sr signatures and hence contribute to the understanding of ^{87}Sr/^{86}Sr isotopes as a tool for ancient and modern migration studies.

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Tables

Table 1. The Sr concentration and ^{87}Sr/^{86}Sr isotope composition for bottled waters from Britain with the county of origin, the aquifer lithology and location given as longitude and latitude.

Sample	Sr ppm	87Sr/86Sr	commercial name	County of source	Geology of aquifer	Latitude	Longitude
JMW 39	0.27	0.707711	Hilldon	Hampshire	Cretaceous Chalk	51.07	-1.543
JMW 37	0.62	0.708459	Cotswold spring	Gloucestershire	Jurassic limestone	51.50	-2.367
JMW 15	0.02	0.710052	Aqua-Pura	Cumbria	Triassic sandstone	54.77	-2.772
JMW 35	0.47	0.709631	Swithland	Leicestershire	Triassic sandstone	52.71	-1.188
JMW 16	0.17	0.709586	Willow	Cumbria	Triassic sandstone	54.17	-2.969
JMW 17	0.16	0.710255	Hadrian	Tyne and Wear	Permian sandstone/dolomite	54.92	-1.418
JMW 06	0.09	0.709260	Brecon Carreg	Powys	Carboniferous limestone	51.84	-3.948
JMW 01	0.58	0.710184	Buxton	Darbyshire	Carboniferous limestone	53.26	-1.915
JMW 33	0.12	0.710368	Findleys	East Lothian	Carboniferous limestone	55.97	-2.573
JMW 10	0.04	0.712504	Tarka Springs	Devon	Carboniferous Millstone grit	50.93	-4.187
JMW 28	0.09	0.712566	Devon Hills	Devon	Carboniferous Millstone grit	50.85	-3.885
JMW 29	0.04	0.712497	Exeter	Devon	Carboniferous Millstone grit	50.93	-4.187
JMW 24	0.06	0.711923	Brittania	Cheshire	Carboniferous Millstone grit	53.22	-2.028
JMW 25	0.04	0.711943	Grindleford	Darbyshire	Carboniferous Millstone grit	53.31	-1.625
JMW 11	0.20	0.711026	Harrogate Spa	Yorkshire	Carboniferous Millstone grit	53.99	-1.564
JMW 38	0.57	0.711424	Abbey Well	Northumberland	Carboniferous Millstone grit	55.16	-1.670
JMW 32	0.47	0.711462	Pennine Spring	Yorkshire	Carboniferous Millstone grit	53.66	-1.787
JMW 04	0.09	0.705868	Caledonian	Perthshire	Carboniferous Volcanic rocks	55.97	-4.203
JMW 21	0.06	0.712024	Edgemoor	Devon	Devonian Sandstone	51.16	-3.940
JMW 03	0.44	0.709725	Strathmore	Angus	Devonian Sandstone	56.62	-3.002
JMW 19	0.06	0.712101	Styles	Somerset	Devonian Sandstone	51.15	-3.410
JMW 14	0.22	0.706595	Highland Spring	Perthshire	Devonian volcanic rocks	56.26	-3.778
JMW 08	0.40	0.711685	Springbourne	Powys	Silurian mudrocks	52.54	-3.074

JMW 13	0.28	0.711917	Montgomery Spring	Powys	Silurian mudrocks	52.54	-3.074
JMW 20	0.47	0.711781	Fairbourne Spring	Powys	Silurian mudrocks	52.54	-3.074
JMW 26	0.10	0.712099	Ty Nant	Cardiganshire	Silurian mudrocks	52.25	-4.108
JMW 05	0.14	0.713932	Radnor Hill	Powys	Siluran mudrocks	52.35	-2.962
JMW 27	0.004	0.713851	Lakeland	Cumbria	Ordovician mudrocks	54.60	-3.096
JMW 07	0.04	0.713288	Malvern	Worcestershire	Precambrian volcanics	52.08	-2.381
JMW 34	0.08	0.710328	Church Stretton	Shropshire	Precambrian sedimentary rocks	52.55	-2.805
JMW 18	0.01	0.714510	Deeside	Aberdeenshire	Precambrian migmatites	57.06	-3.000
JMW 23	0.03	0.720647	Speyside Glenlivit	Highlands	Precambrian pelites	57.27	-3.255

Figures

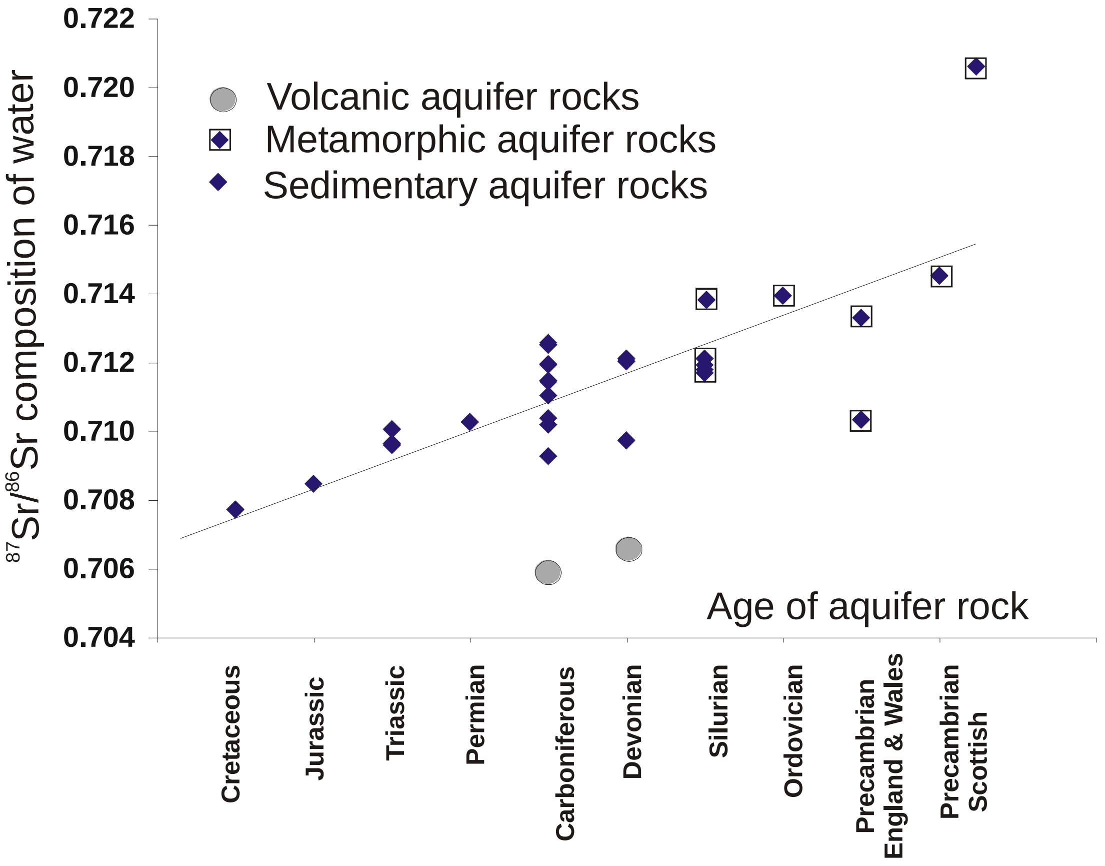

Figure 1. The ^{87}Sr/^{86}Sr isotope composition of bottled waters plotted against the geological stratigraphy of the aquifer source rock. The errors on the isotope data (y-axis) are within the plotted symbols. The x- axis is not to scale as the geological systems are of differing length (see text).

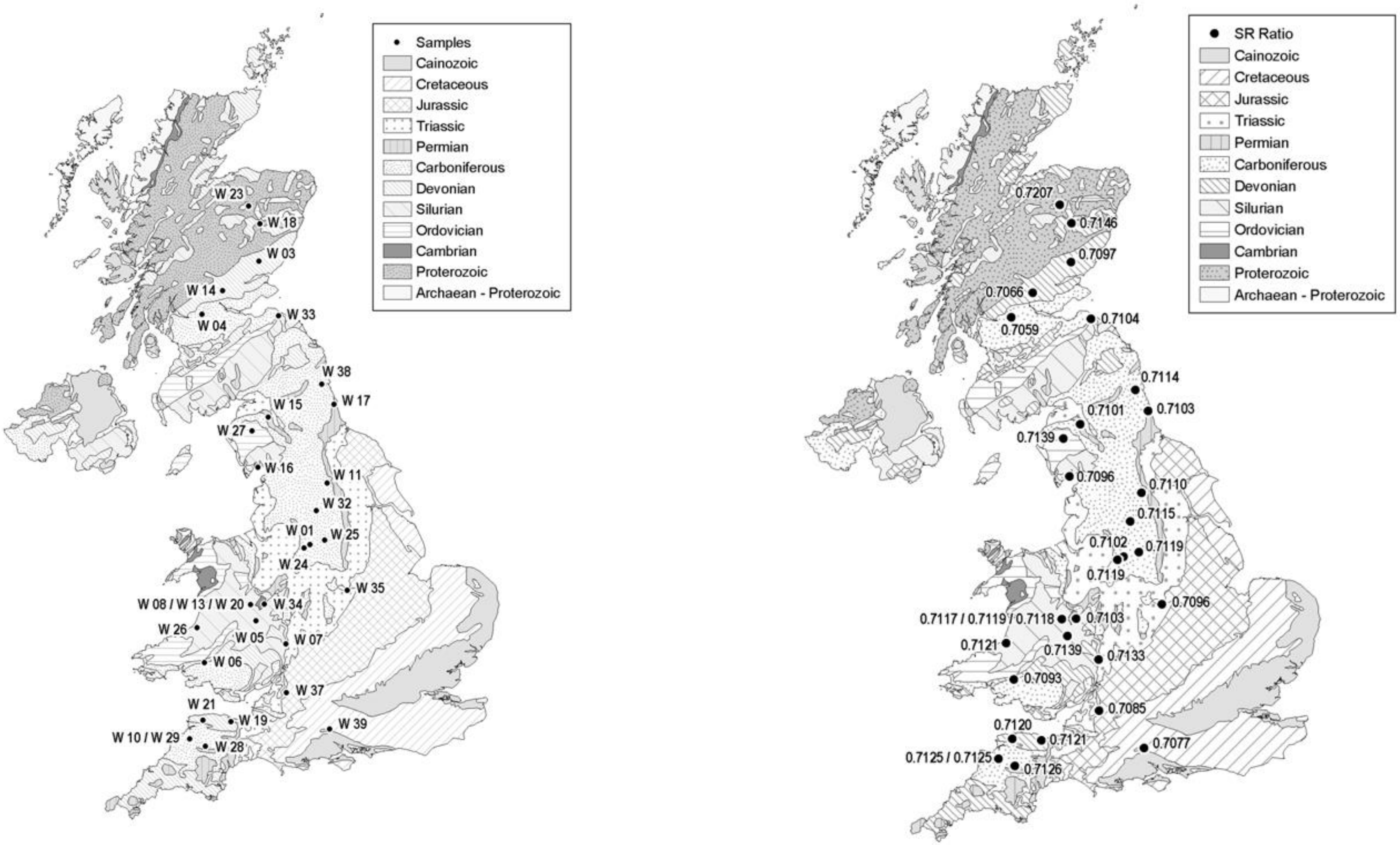

Figure 2a and b. The geographic distribution of a) bottled water samples and b) ^{87}Sr/^{86}Sr ratios from mineral waters shown relative to the main geological subdivision of the UK.