THE AGE AND ORIGIN OF THE BATH THERMAL WATERS

New geochemical investigations as part of the Bath Spa Project

BRITISH GEOLOGICAL SURVEY

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Summary

The most comprehensive geochemical investigation to date has been carried out on the Bath thermal waters using samples obtained from the Stall Street borehole and the Cross Bath in August 2000. Prior to the current investigation the available evidence pointed to a Holocene age for recharge of the thermal water, but with an ill-defined lower limit. The use of the isotope ^{39}Ar in this study has now indicated that the bulk residence time of the thermal water exceeds 1000 years. Indications of a Holocene age were confirmed by the new results, and the thermal component now emerging is most probably some 5-10 thousand years old. However, stable isotopic evidence from confined groundwaters in the vicinity suggests that the thermal circulation may have been in operation since at least as far back as the late Pleistocene.

Solute and dissolved gas data show that the thermal water has evolved predominantly within the Carboniferous Limestone, with little or no evidence for contributions from the underlying Old Red Sandstone or overlying Coal Measures. The rise to the surface of the thermal water from a depth of ~3km is sufficiently slow to allow a temperature drop of some 20°C, pointing to the existence of a complex fracture network. Best estimates for Carboniferous Limestone porosities at depth imply a large (basin-wide) storage volume for the thermal water.

Earlier indications of the presence of a small proportion of modern water in the thermal discharge have been confirmed by noble gas and chlorofluorocarbon data to amount to some 5% by volume. This component now requires better characterisation in terms of fluctuations due to seasonal or other short-term factors, in order that its provenance can be better understood and any water quality implications fully appreciated.

1 Background to the Present Investigations

As part of the development of the new spa, the opportunity existed to update the database on the composition of the thermal springs and an improved understanding of their origin using state-of-the-art geochemical tools. Work was conducted which included detailed chemical analysis, radioactive and stable isotope studies on the water and the solutes, and investigation of dissolved gases. These investigations provide the scientific basis, alongside hydrogeological and geophysical studies, for explaining where the waters are likely to have come from, how old they are, as well as explaining many of their unique properties.

Scientific investigations of the Bath springs have been undertaken for around 175 years, although speculation on their origins have been made for over 300 years. No fewer than 10 analyses of the springs have been published (although with varying degrees of detail and accuracy) since 1823. Detailed studies of the springs have been carried out by BGS and co-workers for the past 25 years with two detailed studies similar in many ways to the present investigations. The most recent geochemical studies have suggested that the water could be around 6000 ± 2000 years; hydrogeological modelling has suggested that the age might be much lower than this, possibly as young as 500 years (Edmunds and Miles 1991).

Interpretation of the properties of the springs is made more difficult since there is only one point of emergence (although with bifurcation). Earlier studies have also suggested that there may be a degree of mixing of younger near-surface water with a much older and deeper source.

The objectives of the present investigations are therefore:

1.   To refine the estimates of age or ages of the thermal water and any extraneous component

2.   To estimate the maximum temperature of emergence and at depth and the likely maximum depth of circulation

3.   To determine the sources and amount of any modern groundwater contamination

4.   To determine, if possible the source area(s) and recharge conditions of the deeper sources as well as the reactions that have taken place during its circulation

5.   To explain several of the properties of the springs such as the degassing, the iron staining, the taste and odour, as well as other questions asked by the general public.

To achieve this a fully comprehensive suite of diagnostic indicators has been applied, representing the most complete look at the composition of the waters to date. The range of indicators and what they might show is summarised in Table 1. Several new techniques have been used, notably the dissolved gas compositions. The highlight of this investigation is the analysis of ^{39}Ar which should help resolve whether the water has any component with an age limit in excess of 1000 yr. Other dissolved gas components such as ^{85}Kr and CFCs (chlorofluorocarbons) can provide sensitive indications of recent additions to the main body of the water. The present studies thus provide an in-depth snapshot of the thermal waters in the year 2000, but no additional interpretation is made here of the springs in their regional context.

Arrangements were made for sampling of the inclined Stall Street (King's Spring) borehole and the Cross Bath spring on 11 August 2000. This involved cordoning off part of Stall Street above the borehole for a period of 24 hours to provide access for the gas sampling equipment. This attracted press attention (Annexe 1). In addition, it was necessary to arrange a sampling line above the Cross Bath spring chamber via a shallow pipe inserted rather precariously over the bath.

A comprehensive series of samples were taken in various containers as indicated in Table 1. *In situ* measurements were made for a number of parameters that were unstable or could not be measured in the laboratory such as T, SEC, pH and redox potential (Eh).

A highlight of the present work was the analysis of ^{39}Ar which was incorporated to help resolve whether water has any component with an age less than 1000 yr. The overall logistics for this sampling were considerable since approx 5 m^3 of water had to be processed to recover enough gas for analysis, taking most of one working day.

2 Analysis and Results for the 2000 Sampling

The range of chemical and isotopic measurements applied during the 2000 investigations may be considered under two categories – inert and reactive tracers. Inert tracers such as stable isotopes of water and chloride provide a record of input conditions whereas reactive tracers record the water rock interaction and hence a record of the reservoir and the reactions taking place.

The chemical and isotopic analyses were carried out in four laboratories: two in the UK (BGS Wallingford and Keyworth) and two in Switzerland (University of Bern and ETH Zurich). Radiocarbon analysis was conducted through a research grant application with the NERC Radiocarbon Laboratory (East Kilbride) who processed the water, converting the inorganic carbon in HCO$_3$ to graphite, the activity of which was then analysed by the University of Arizona. The main techniques used are also summarised in Table 1. Problems were met with the ICP-MS analysis at Wallingford and so samples were also sent to a commercial laboratory in Vancouver. Full details of the analytical methods used can be obtained by contacting one of us (WGD) at BGS in Wallingford.

Some of the areas for which the various measurements can be used for interpretation are summarised in Table 1. Full results of the 2000 sampling of the King's Spring borehole and the Cross Bath Spring are listed in Tables 2-4 and the main characteristics of the waters (their major and minor inorganic solutes, stable and radioisotope compositions, and dissolved gases) are briefly discussed under separate headings:

2.1   FIELD OBSERVATIONS

The discharge temperature of the King's Spring borehole is 44.8^{o}C (Table 2), slightly higher than the Cross Bath (44.5^{o}C), consistent with slight cooling of the latter as it travels further away from the discharge region.. The pH of both sources is similar. The redox potential of the King's Spring (-2 mv) indicates reducing conditions at depth and differs from the Cross Bath, which is less reducing and points to a possible difference in conditions as water travels from the deeper and shallower levels of emergence.

2.2   MAJOR ION CHARACTERISTICS

Seven major ions comprise the bulk of the dissolved solids in the Bath thermal waters (Table 2). The total mineral content of 2290 mg l^{-1} is dominated by sulphate and calcium. Sulphate comprises 1080 mg l^{-1} of the total. During its circulation the water has also acquired almost equivalent amounts of Na and Cl (218 and 344 mg l^{-1} respectively).

2.3   DISSOLVED ORGANIC CARBON

The content of organic carbon in the Cross Bath is relatively high and greater than that in the Stall Street borehole (Table 2). This may be a possible indicator of shallow groundwater influx.

2.4   MINOR AND TRACE ELEMENTS

This study provides the most comprehensive trace element analysis of the water carried out to date with 70 elements having been determined, 48 of which are present above their limits of detection (Table 3). These elements following initial rainfall inputs are largely derived through reactions with the geological formations through which the water has passed, thereby providing a fingerprint of the thermal water that can be used to restrict some of the interpretations of its origin. Similarities between the King's Spring borehole and Cross Bath also confirm the same primary source of the water.

2.5   STABLE ISOTOPES (WATER AND SOLUTES)

Stable isotope ratios ($\delta^{18}O$, δ^2H) provide an indication of the source of the water, especially whether it can be related to modern rainfall or not. Two other isotope ratios $\delta^{13}C$ and $^{87}Sr/^{86}Sr$ provide additional tracer information by giving an indication of lithologies encountered by the water and the extent of water-rock interaction. These data are provided in Table 4.

2.6   RADIO-ISOTOPES

Radiocarbon (^{14}C) and tritium (^3H) provide the traditional indicators of absolute age for the waters. In the present study these measurements have been made but difficulties with radiocarbon interpretation exist, since some may have been lost through reactions with the aquifer rocks rather than simply by decay. Relative differences in the two sources (Table 4) suggest that the Cross Bath (with higher radiocarbon) may have a component of more radioactive modern groundwater. In the present study however, the dissolved gas isotopes ^{85}Kr and ^{39}Ar provide additional indicators of the absolute age of the groundwaters (Table 4).

2.7   DISSOLVED GASES AND GAS ISOTOPES

Some gases dissolved in the groundwater from the atmosphere at the time of recharge retain their compositions and ratios to each other during groundwater circulation, while others may undergo chemical reactions. Gas compositions in the discharge may also be modified by addition from subsurface sources. The concentrations of gases in the thermal water reveal something of the balance of processes in the reservoir.

Of particular importance in the present investigation are the noble gases (Ar, Kr, Ne, Xe), which record the groundwater temperatures at the time of recharge, and CFCs, which can indicate if there is any modern contribution. The noble gases can also provide information via their isotopic content. The radioactive isotopes such as those of Ar and Kr provide quantitative residence time information, while those of helium are more qualitative but can nevertheless provide indications of contributions of very old water.

3 Comparison with Earlier Analyses

In this report a comparison is made (Table 5) between the current analysis and those made during two earlier detailed sampling campaigns in 1979 and 1986; comparisons with analyses prior to 1979 are found in Edmunds and Miles (1991). The results are given only for those elements where a direct comparison is relevant.

A decrease in the temperature of the groundwater derived from the Stall Street (King's Spring borehole was observed after its drilling in 1986, compared with the original King's Spring and this slightly lower temperature is confirmed. Some seasonal fluctuation, possibly related to flow,

may occur and monitoring information is required to resolve this. The recent temperature is shown in relation to historical temperature measurements in Figure 1.

The chemistry of the carbonate system (pH, HCO_3, Ca) has been constant over the 20-year period. Sulphate and chloride have been used as indicators of the compositional trends in the past and the present results (Figure 2) are compared with present data; both species are slightly higher than in previous analyses.

Other changes that may be significant are a slight increase in silica (indicating a slightly deeper circulation pathway, discussed below), higher F, Li, Sr – all trace elements which remain conservative in the system and may be interpreted in terms of a longer residence time. In contrast, the Br has decreased (relative to Cl) and the possible significance of this is discussed below.

Slightly more positive values for $\delta^{18}O$ and δ^2H are observed but these are within the analytical precision of the measurement and therefore probably not significant. Of more significance may be the decrease in ^{14}C activity (possibly indicating an increase in relative age or, more likely, change in mixing ratios).

4 Interpretation of the 2000 Data

4.1 AGE OF THE GROUNDWATER AND EXTENT OF MIXING WITH MODERN WATER

Two persistent problems in determining the groundwater age have been a) the difficulties in the modelling of a radiocarbon age due to reaction with the reservoir rocks which is likely at elevated temperatures, and b) the degree of mixing with any shallow water with a residence time of less than 1000 yr. In the present study, additional tracers (^{37}Ar, ^{39}Ar and ^{85}Kr) allow the mixing problem to be properly addressed before the age of the water is considered.

Being noble gases, ^{85}Kr and ^{39}Ar undergo no chemical reactions and are, in ideal cases, relatively easy to interpret as residence time indicators. Over the last 50 years ^{85}Kr has been released from nuclear fuel processing plants leading to steadily increasing concentrations in the atmosphere. With a half-life of 10.7 years, ^{85}Kr is suitable for the identification and dating of water components with residence times of decades. It has been shown that subsurface production of ^{85}Kr can be neglected (Loosli et al, 1999).

The atmospheric ^{39}Ar concentration originates mainly from the interaction of neutrons derived from cosmic ray activity with ^{40}Ar and does not vary over time (Loosli and Lehmann, 1991). With a half-life of 269 yr, groundwaters with ages up to 1000 yr can be identified. The usefulness of ^{39}Ar as a dating tool may be limited because ^{39}Ar atoms can be produced in the subsurface by the $^{39}K(n,p)^{39}Ar$ reaction. Subsurface neutron fluxes and production rates can be roughly estimated by theoretical calculations based on measured rock compositions and by comparison with other isotope concentrations that are produced in the subsurface. A suitable indicator is ^{37}Ar because (i) chemically it behaves identically to ^{39}Ar and (ii) it has a half-life of only 35 days which excludes an atmospheric origin in older groundwaters. ^{37}Ar is produced by the reaction of neutrons with ^{40}Ca.

Nuclide concentrations in water N_w are related to concentrations in the rock matrix N_R by:

$$N_w = N_R \cdot \frac{e}{p}$$

4

where e is the escape probability from the rock into the water and p is the rock porosity; e is mainly a function of the rock geometry and porosity and the spatial distribution of the target elements K and Ca.

Estimates of the nuclide production rates in the rock as well as escape probabilities are subject to considerable uncertainties. Therefore calculated activities can only give the order of magnitude of the expected measured values in the water.

The underground production rates of ^{37}Ar and ^{39}Ar were estimated from the chemical composition of the rocks in the reservoir and the cross section data for the energy of the neutrons (Andrews et al., 1991). It has been assumed that the water has equilibrated in the Carboniferous Limestone.The results of the calculations are compared with the measured gas concentrations in the water in Table 6.

Adopting equation (1) and assuming an escape probability and a porosity of the limestone (Andrews et al. 1992) of 1% for both, the numbers in the last two columns can be directly compared. The measured and calculated ^{37}Ar concentrations in water do approximately agree, confirming the correctness of the selected parameters used for the calculations. For the other two gases, however, there is a significant amount of atmospheric input denoting a modern component. In the case of ^{85}Kr, the *in situ* production is insignificant and the presence of young water is the only explanation. This is also supported by the presence of a small amount of tritium (1±2 TU) measured in earlier samples (Andrews et al. 1982) and confirmed again (0.035±0.637) in the present investigation.

Since there are data from two tracers, krypton and tritium, it is possible to calculate the age and the degree of mixing of the young component. The interpretation of the data applying a low-dispersivity (i.e. piston flow) model (Smethie et al, 1992), yields a residence time of 1-18 years and a mixing ratio of 1-5% for the young water component. This is similar to chemical estimates of a young component (5%) from earlier studies (Edmunds et al. 1991).

This result has implications for the ^{14}C interpretation. With a maximum residence time of 18 years for the young component, the input of ^{14}C is 135%. Applying the different correction models, the initial ^{14}C activity in the young component is reduced to 100% modern carbon. Considering a mixing ratio of 5%, the calculated ^{14}C activity in the mixture is 4 pmC (percent modern carbon), slightly in excess of the measured value. This must mean that the age of the main component cannot be recent.

The confirmation of mixing also has implications for the ^{39}Ar, some of which may be derived from the young component. The ^{39}Ar concentration in modern water, according to the solubility at the recharge temperature ($4.813.10^{-4}$ cm^3 STP/g water at 10°C), is 11 atoms/cm^3 water. After mixing, the concentration of ^{39}Ar (5% of the young component) is estimated to be 0.4 atoms/cm^3 water. Consequently, 1.3 atoms per cm^3 water (of the total of 1.7 atoms) are from the old component. The range of *in situ* production is variable (0.3-1.0), due to uncertainties in a number of the parameters used (Andrews et al. 1991; Loosli et al. 1991). This means that the residual atmospheric ^{39}Ar that is relevant to dating is very close to the *in situ* production value and that the minimum groundwater age from this isotope must be 1000 years and most probably much older.

An independent constraint on the groundwater age comes from the dissolved noble gas concentrations (not reported). These indicate the temperature at which the groundwater was recharged at source. The mean ratio from duplicate measurements of the Stall Street borehole water is 10.3 ± 0.5 ^{o}C, identical within error to modern recharge. This must mean that the water entered the aquifer since the last ice age or, much less likely, during the last interglacial maximum (more than 100,000 yr ago). This would put a likely upper limit on the groundwater age of 12,000 yr, since volumetric constraints would preclude the older age hypothesis.

CFCs (chlorofluorocarbons) are extremely sensitive indicators of the presence of waters recharged in the second half of the 20th Century. The existence of three CFC species allows a

degree of cross-checking. In the Stall Street B/H, CFC-11, CFC-12 and CFC-113 give amounts of 'modern' water as 3, <1 and 4 % respectively, whereas at the Cross Bath the figures are 5, <1 and 7 %. The CFC-11 and CFC-113 values are similar both to each other and to those obtained from the noble gas isotopic and hydrochemical data (see above). On the other hand CFC-12, normally the most 'reliable' CFC, gives a below-detection value. However, little is yet known about how conservatively CFCs behave in thermal waters, and it may be that CFC-12 is more susceptible to thermal degradation than the other two species.

In the context of CFCs, the term 'modern water' implies recharge dating from no earlier than the mid-1990s, when the chlorofluorocarbons reached their atmospheric maxima. If the modern water component were rather older, i.e. nearer to the 18-year maximum suggested by the ^{3}H-^{85}Kr data, the measured CFC-11 and CFC-113 concentrations for the Stall Street B/H would convert to proportions of 4.3 and 10 % modern water respectively. Since these values do not agree particularly well with each other, the more recent age estimated for modern water is preferred. The combined CFC-radioisotope evidence is therefore best interpreted to indicate that the modern water component is in the lower part of the 1-18 year age range and the upper part of the 1-5 % proportion of the total water.

Unlike the noble gases and CFCs, the 'ordinary' gas content of the thermal water can give only qualitative indications of residence. Water commences recharge with its gas content in equilibrium with the atmosphere. Subsequent processes may add to or subtract from individual gases. By the time the thermal water is intercepted in Bath various changes have occurred, principally the total disappearance of O_2 and the elevation of CO_2 to around one-third of the gas phase by volume. These effects are typical of long-residence thermal waters in carbonate aquifers: O_2 has been consumed by water-rock interaction, while the steep rise in CO_2 is due to re-adjustment of the carbonate system.

Nitrogen on the other hand tends to behave inertly in thermal systems unless there is some extra source of N_2 gas such as denitrification. One check on this is to measure the N_2/Ar ratio of the water: in the case of the Stall Street borehole the value was 45.3, which is typical of groundwaters recharged at temperatures of around 10°C and which have been unaffected by denitrification.

Radon, with a half-life of only 3.8 days, cannot be regarded as a residence time indicator and is primarily used in calculations of fracture apertures, a subject beyond the scope of the present report. The present measurement for the Stall Street B/H is very similar to that previously obtained and discussed by Andrews (1991), though the Cross Bath value was significantly higher for reasons which are unclear.

4.2   THE MAXIMUM TEMPERATURE AND DEPTH OF CIRCULATION

The measured temperature of the borehole discharge has remained the same (within measurement error), since the last period of detailed study in 1986, at 44.8°C, although as noted earlier, there may be seasonal fluctuations, probably related to the few percent of mixing with a shallow cooler source. Water-rock reactions are temperature dependent and certain dissolved constituents may be used to calculate the maximum temperatures to which the groundwater has been subjected. The reaction kinetics of dissolution may be quite rapid, but once in solution the kinetics of re-precipitation are slow and thus the water retains a memory of the maximum temperature. Geothermometers are most reliable in high temperature geothermal systems and for the Bath system only silica is likely to be applicable (Edmunds and Miles 1991).

The groundwaters are supersaturated with quartz (Table 5), but are close to saturation with chalcedony, which is more likely to control silica solubility at the relatively low temperatures involved. For typical measured Si concentrations for the Stall Street borehole of 19.5-21.8 mg l^{-1} (corresponding to SiO_2 of 41.8-46.7 mg l^{-1}), temperatures of 63.1-68.5°C (chalcedony) and 93.6-98.6°C (quartz) are obtained. Assuming the lower temperature range as being more likely, a

minimum drop in temperature of more than 20°C is being exhibited by the thermal water between the main reservoir and its emergence at Bath. For the reasonably high output of 13 l s^{-1} this represents a significant thermal loss and suggests that the water is not being brought rapidly to the surface by a single major fracture as (for example) is suggested in the schematic cross-section of Andrews et al. (1982). It seems more likely that the water is travelling relatively slowly to the surface along a more complicated set of fractures while retaining its thermal signature.

Bath is in a low heat flow area of the UK with an estimated heat flow of about 45 mW/m^2, corresponding to a geothermal gradient of around 20°C km^{-1} (Downing and Gray 1986). With a surface temperature of 11°C, a maximum depth of circulation of 2.9-4.4 km is indicated; however since the chalcedony control is more likely in this low temperature environment, a maximum circulation depth of 3 km is considered more probable. This depth is consistent with the depth range of the Carboniferous Limestone (3.5 to 2.7 km) in the Pensford Basin, 12 km west of Bath (R. Gallois, pers. comm.). The silica concentrations are slightly higher in 2000 than in 1986 but, as with the change in radiocarbon activity noted earlier, this is likely to be the result of a small change in the amount of modern water mixing.

4.3   THE SOURCE OF THE WATER AND CIRCULATION PATHWAYS

The isotopic evidence provides indications for the source of the water, while the solute concentrations (major and trace elements) provide clues as to the types of rocks through which the water has passed, with qualitative indications also of residence times.

The stable isotope ratios (Figure 3) lie well within the range of modern waters measured in the region (Dennis et al, 1998) and distinct from palaeowater compositions (Darling et al, 1997). This supports the conclusion from the noble gas recharge temperatures that the groundwater is predominantly of Holocene age. Further support comes from deuterium data obtained from fluid inclusions from Mendip cave stalagmites dated at mid-Holocene age (Dennis et al, 1998): the values are identical within measurement error to those of the Bath thermal waters.

Hydrogeochemical evidence from waters confined by the Mesozoic fill to the west of the city centre suggests that the sediments have been invaded by thermal outflows either from the King's Spring area or possibly from another unidentified point of emergence

(Edmunds and Miles 1991; Darling and Edmunds, 2001). However, the stable isotope values of these waters are depleted and therefore it appears that at least a proportion must have been introduced in the late Pleistocene. If this is indeed the case, the implication is that the thermal circulation has been in existence significantly longer than the bulk of the water currently passing through it.

The Br/Cl ratio is a diagnostic indicator of the source of Cl (Edmunds 1996) and it was concluded in the 1986 study (Edmunds and Miles 1991) that since this ratio was slightly enriched in Br relative to sea water, the Cl was derived from organic rich formation waters rather than an evaporite source. This is consistent with an evolution in the Carboniferous Limestone sequence. The slight increase in Cl since 1986 (see Figure 4) is matched by a slightly lower Br/Cl ratio, which is consistent with small amounts of mixing from higher Cl waters in either the Carboniferous or overlying strata (Darling and Edmunds, 2001).

Strong evidence for a prolonged groundwater evolution in the Carboniferous Limestone (or indeed any marine limestone) is provided by the $\delta^{13}C$ value. As discussed in Edmunds and Miles (1991), the value of −1.54‰ (identical in both studies) indicates that isotopic equilibrium is approached with the rock – near to 0‰ - as compared with a young groundwater from limestone, which would have a value of near to -13‰, reflecting the initial reactions in the soil. This process is likely to have taken thousands rather than hundreds of years although reactions would have accelerated during residence at the higher temperatures. Strontium isotope evidence is more

equivocal in that an unambiguous source of Sr cannot be identified. The measured $^{87}Sr/^{86}Sr$ range of 0.710551-0.71075 suggests that much of the Sr content of the water may have been acquired during infiltration through Triassic strata. However, this would not rule out a long residence in the Carboniferous Limestone by the thermal water.

The relatively low Mg/Ca ratio (see Figure 5) taken from Edmunds and Miles (1991) also suggests that the groundwater has evolved in a relatively pure limestone formation. If this were a Mesozoic Limestone or a dolomitic limestone then it is likely that an Mg/Ca ratio after prolonged water rock interaction would be roughly equimolar (1:1). The ratio is slightly Mg-enriched compared with younger waters in the Carboniferous Limestone, as would be expected following incongruent solution where traces of Mg are added to the groundwater over time. The similarity with the Hotwells (Bristol) thermal water, which issues from the Carboniferous Limestone, is also apparent. There seems no doubt therefore that the Carboniferous Limestone is the host rock for the thermal water evolution.

The reducing conditions in the thermal waters restrict the mobility of most trace metals in the groundwater, with the exception of iron (Fe^{2+}). Very few metals exceed 5 μg l^{-1}. Arsenic and selenium are exceptions with concentrations of 7.1 and 8.5 μg l^{-1} respectively; the reduced form, arsenic $^{(III)}$ (5.9 μg l^{-1}) predominates. The concentration of dissolved Fe (0.77 mg l^{-1}) is similar to previously measured concentrations. As the groundwaters ascend in the aquifer and mix in the near-surface with traces of oxygenated water (<0.7 mg l^{-1} recorded on emergence in the present samples), a sharp rise in Eh takes place, causing precipitation of some of the Fe in the final stages of the ascent and emergence.

Based on the conclusions of Andrews (1991), the required karst storage for a thermal water age of 10,000 yr would be about 8 % of the Carboniferous Limestone volume within a 30° segment from Bath to the Mendip Hills (the traditionally accepted source of the Bath thermal water). Even allowing for a degree of karstic porosity over and above the 1% porosity allocated to the Carboniferous Limestone largely on account of fracturing, 8% is a highly improbable value. This implies that if the thermal water is of early Holocene age, storage must involve the Carboniferous Limestone throughout the Bath-Bristol basin rather than simply a relatively small segment.

Estimates of the saturated volume of the Carboniferous Limestone from which the thermal springs could potentially draw their water has a maximum possible volume of 480 km^3 although this figure is likely to be somewhat overestimated (R. Gallois pers. comm.). The present-day flows from the springs (60 m^3/hr or 525600 m^3/yr) translate into 5265 km^3 for the maximum likely residence time of 10,000 yr if one assumes steady state, and would thus be consistent with a porosity of around 1%. However the evidence for such high porosity values remains tenuous and constrains the upper limit of residence at 10-12,000 years (in line with the other geochemical evidence). Somewhat younger ages would allow lower porosities to be included in the model, although the absolute limit would be 0.1 % (as constrained by the argon data).

4.4   Contributions at Depth from Formations other than the Carboniferous Limestone

The possibility exists that the thermal water could contain small contributions from the overlying Coal Measures or the underlying Old Red Sandstone. There is no direct evidence of the former. Methane is present only at very low concentrations in the thermal waters (Table 4). At Stall Street ethane was just detectable. The low CH_4 concentration and the high CH_4/C_2H_6 ratio of ~2000 are typical of bacterially-produced reduced carbon gases found in waters of comparable redox status in low-organic-content formations. There is no evidence from these results that the thermal waters have been exposed to Coal Measures or other rocks with an elevated organic content; higher concentrations of CH_4, C_2H_6 and C_3H_8 would be expected in such circumstances.

There is better evidence for a minor contribution from the Old Red Sandstone. Although the N_2/Ar ratio measured for the Stall Street water suggested no perturbation of the dissolved N_2

load (see above), the uncertainties associated with the measurement could mask small additions of Ar in the subsurface. This is derived from the radioactive breakdown of ^{40}K to ^{40}Ar. The extent of such additions can be revealed by measuring the ratio $^{40}Ar/^{36}Ar$. As part of the present investigations a ratio of 297.0 was measured which exceeds the atmospheric solubility equilibrium value of 295.5. Owing to the low potassium concentration in the Carboniferous Limestone it is unlikely that the radiogenic production within this formation is solely responsible for this excess of ^{40}Ar. The time to produce the observed excess would be over 20 million years assuming an escape of 10 % for argon, which is most probably an overestimate. The only explanation for this excess is an external source - whether a very old water component from the underlying Old Red sandstone or a diffusive influx of argon gas. This finding is in agreement with the anomalously high ^{4}He concentrations and low $^{3}He/^{4}He$ ratio found in the thermal waters at Bath (Table 4). It is considered therefore that upward leakage of gases from underlying formations is necessary to explain these features.

6   Conclusions

Analyses of water, solutes, isotopes and dissolved gases carried out in 2000 provide the most comprehensive interpretation to date of the origins, age and circulation history of the Bath Thermal Springs. It is shown using ^{3}H, ^{85}Kr and CFCs that the discharge contains a proportion of up to 5% modern water less than two decades old, and probably derived from Mesozoic strata some 10-20 metres below the point of emergence, entrained in the permeable funnel of the spring chamber (Kellaway 1991). This modern water contains traces of oxygen and is responsible for the precipitation of iron, colouring the waters on emergence. The modern water was intercepted formerly by the spring under natural discharge conditions, but apparently slightly more of this water has been intercepted since the drilling of the Stall Street (King's Spring) borehole.

Confirmation of mixing with modern water provides an improved explanation of the residence time and the source of the deeper, thermal component of the spring. The age of this water must be in excess of 1000 years, as indicated by the ^{39}Ar dating. However it must be less than 12,000 years old on the basis of its dissolved noble gas and stable isotope contents. Qualitative evidence (for example enriched ^{13}C and a likely negligible, residual ^{14}C) suggests the water to be nearer the upper age limit, and a range of 6-10 kyr is proposed.

From geothermometry the water is likely to have reached a maximum temperature of 69°C, indicating a probable maximum circulation depth of around 3 km. The ascent of the water to the surface is sufficiently indirect that a temperature loss of more than 20°C is sustained. There is overwhelming evidence that the water has evolved within the Carboniferous Limestone formation, although the chemistry alone cannot pinpoint the geometry of the recharge area or circulation route. Volumetric calculations imply both a large storage volume and circulation pathway if best estimates for porosities of the limestone at depth are used.

Residual thermal water is recognised in the surrounding area to a distance of a few km, and from its isotopic composition it is probable that recharge and discharge of thermal water has been taking place at least since the cooler climates of the Late Pleistocene.

7   Recommendations and Outstanding Issues

The present report provides a snapshot of the thermal waters for August 2000. It should be stressed that all previous studies have indicated that there are temporal variations in the springs and the results should therefore be regarded against this background.

There is a strong indication from the recent work that a small proportion of modern water is entering the discharge. It should be possible to resolve this by careful monitoring of the spring (Stall Street borehole) for both chemistry (including specific electrical conductance) and temperature. The recently initiated high resolution monitoring of the King's Spring and other points of discharge should help to detect any short and long term oscillations and point towards the nature of any mixing with a shallow component. It is also recommended that weekly measurements of the Stall Street borehole discharge be carried out during a one year period at the same time as the present sampling for microbiology. The minimum recommended parameters for detection of mixing with shallow waters are:

(ToC, pH, SEC); mineral analysis (Cl, SO_4, HCO_3, Na, K, Mg, Ca), Fe, DOC (Dissolved organic carbon).

In addition the variations in Eh as measured in the past could provide an additional check on the oscillations in the redox conditions. Small changes in pH may also be detectable due to pressure changes during ascent (and may also slightly influence the redox conditions). A one-off full analysis for non-biodegradable microorganic components should also be made (if not already carried out) since the presence of CFC's indicate that there may be other qualitative indicators of modern water, especially if derived from shallow urban drainage.

It is also recommended that at least monthly samples be taken for CFCs, which are judged to provide the most reliable and quantitative measurements of the proportion of recent inflow.

Some possibility exists that thermal water is discharging elsewhere beneath cover rocks in the Avon valley, although volumetric estimates now make this a less likely theory than previously thought.

The chemical and isotopic data cannot resolve the exact recharge area(s) of the thermal water, nor its underground pathway or circulation network. Further discussion is now needed to resolve the most likely hydrogeological model based on all the evidence now available. A more thorough consideration of the present data in conjunction with analogues of thermal waters elsewhere in the UK and Europe where circulation occurs in the Carboniferous Limestone also needs to be made.

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Tables and Figures

Table 1   Geochemical indicators and their possible use applied to the Bath thermal waters. Sampling details and measurements carried out by the different laboratories are given.

Indicator	Use	Lab	Sampling details and notes
FIELD			
Temperature	In situ properties	Field	Wellhead
Field pH			Wellhead
Dissolved Oxygen (DO)			Wellhead (anaerobic)
Redox Potential (Eh)			Wellhead (anaerobic)
SEC			Wellhead
Alkalinity (HCO_3)			Field titration
INORGANIC			
Major anions (Cl, NO_3),	Fingerprinting and water-rock interaction	WL	Nalgene bottle 60 ml, F/UA
Major cations (Na, K, Ca, Mg, SO_4)			Nalgene bottle 60 ml, F/A
Minor ions (Br, F, I, NH_4, NO_2)			Nalgene bottle 60 ml, F/UA
Trace elements (ICP-AES) Sr, Ba, Li, Si, B, Fe, Mn, Zn, P, V	Si geothermometry, fingerprinting and water-rock interaction	WL	Nalgene bottle 60 ml, F/A
Trace elements (ICP-MS) Li, Be, B, Al, Cr, Mn, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Rb, Y, Mo, Ag, Cd, Sb, Cs, Ba, La,Tl, Pb, Bi, Th, U, REE (Rare Earth elements)	Fingerprinting, source rocks and water-rock interaction; residence time	WL	Nalgene bottle 1 litre, F/A
As^{III}/As^{V}	Redox, water-rock interaction	WL	Nalgene bottle 60 ml, F/A
ORGANIC			
DOC	Natural vs contaminant source of C	WL	Glass bottle 50 ml, Al seal
TOC		WL	Glass bottle 50 ml, Al seal
STABLE AND RADIOISOTOPES			
$\delta^{18}O$, $\delta^{2}H$	Origins and recharge characteristics of the water	WL	Glass bottle 30 ml
$\delta^{13}C$	Source of inorganic carbon; correction of ^{14}C	WL	Glass bottle 50 ml
^{14}C activity	Groundwater age (5730 yr half-life)	RCL	Amber glass bottle 2x500ml
$\delta^{87}Sr/\delta^{86}Sr$	Source of Sr	KW	Nalgene bottle 1 litre
DISSOLVED GASES (AND ISOTOPES)			
CH_4, N_2, O_2, N_2/Ar	Redox processes, provenance of water	WL	Stainless steel 50 ml 'bomb'
CFCs	Fingerprint of modern water/age	WL	Glass 500 ml (anaerobic)
$^{39}Ar/^{37}Ar$	Groundwater age (269 yr half-life)	B/Z	Steel cylinders: gas extracted from 5 m^3 water
$^{3}He/^{4}He$	Crustal component and high precision groundwater age (^{3}H-^{3}He)	B/Z	Clamped copper tubes
Kr. Ne, Xe	Recharge temperature	B/Z	Clamped copper tubes
^{3}H (^{3}He)	Modern component (12.43 yr half-life)	B/Z	Clamped copper tubes
^{85}Kr	Modern component (10.8 yr half-life)	B/Z	Clamped copper tubes
^{222}Rn	Radioactivity of source rocks	KW	Amber glass bottle 250 ml

B/Z – University of Bern/ETH Zurich; KW – BGS Keyworth; RCL – NERC Radiocarbon laboratory; WL – BGS Wallingford.

Table 2 **Site data, field and laboratory measurements of major and minor species for the August 2000 sampling of the Stall Street borehole and the Cross Bath**

Site	Date	T	pH	Eh	SEC	Diss. O_2	Ca	Mg	Na	K	Cl	SO_4	HCO_3	NO_3-N	NO_2-N	NH_4-N	P	TOC	DOC	F	Br	I	Si
		°C		mv	µS cm^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}
Stall Street B/H	11/8/00	44.8	6.71	-2	2560	<0.1	385	50.9	218	19.9	344	1080	187	< 0.5	< 0.003	0.35	< 0.2	1.1	4.3	2.28	1.86	0.04	21.78
Cross Bath	11/8/00	44.5	6.70	158	2830	<0.1	407	52.5	221	20.4	335	1120	149	< 0.5	< 0.003	0.37	< 0.2	5.6	19.2	2.23	1.75	0.04	22.48

Table 3 **Trace element data for the August 2000 sampling of the Stall Street borehole and Cross Bath. All results expressed as µg l^{-1}.**

Site	Ag	Al	As_T	As_3	Au	B	Ba	Be	Bi	Cd	Ce	Co	Cr	Cs	Cu	Dy	Er	Eu	Fe	Ga	Gd	Ge	Hf	Hg	Ho	In	Ir	La	Li	Lu	Mn	Mo
Stall Street B/H	<0.05	3	7.1	5.92	<0.05	516	322	0.15	<0.05	0.13	0.06	<0.02	<5	3.33	1.1	0.01	0.01	<0.01	774	<0.05	0.03	0.72	<0.02	<0.1	<0.01	<0.01	<0.05	0.05	0.39	<0.01	456	1
Cross Bath	<0.05	1	7.1	5.55	<0.05	520	309	0.09	<0.05	0.09	0.06	<0.02	<5	3.32	1.2	0.01	0.01	<0.01	722	<0.05	0.03	0.76	<0.02	<0.1	<0.01	<0.01	<0.05	0.05	0.41	<0.01	483	1

Site	Nb	Nd	Ni	Os	Pb	Pd	Pr	Pt	Rb	Re	Rh	Ru	Sb	Sc	Se	Sm	Sn	Sr	Ta	Tb	Te	Th	Ti	Tl	Tm	U	V	W	Y	Yb	Zn	Zr
Stall Street B/H	0.01	0.08	<2	<0.05	<2	<0.2	0.01	<0.001	22.6	<0.001	<0.001	<0.05	<0.05	5.41	8.5	<0.05	0.07	6601	<0.05	<0.01	0.14	<0.05	<10	0.1	<0.001	<0.05	2	<0.1	0.12	0.01	44	<0.5
Cross Bath	0.01	0.07	<2	<0.05	<2	<0.2	0.01	<0.001	22.0	<0.001	<0.001	<0.05	<0.05	5.31	8.8	<0.05	0.08	6395	<0.05	<0.01	0.09	<0.05	<10	0.11	<0.001	<0.05	1	<0.1	0.12	0.01	65	<0.5

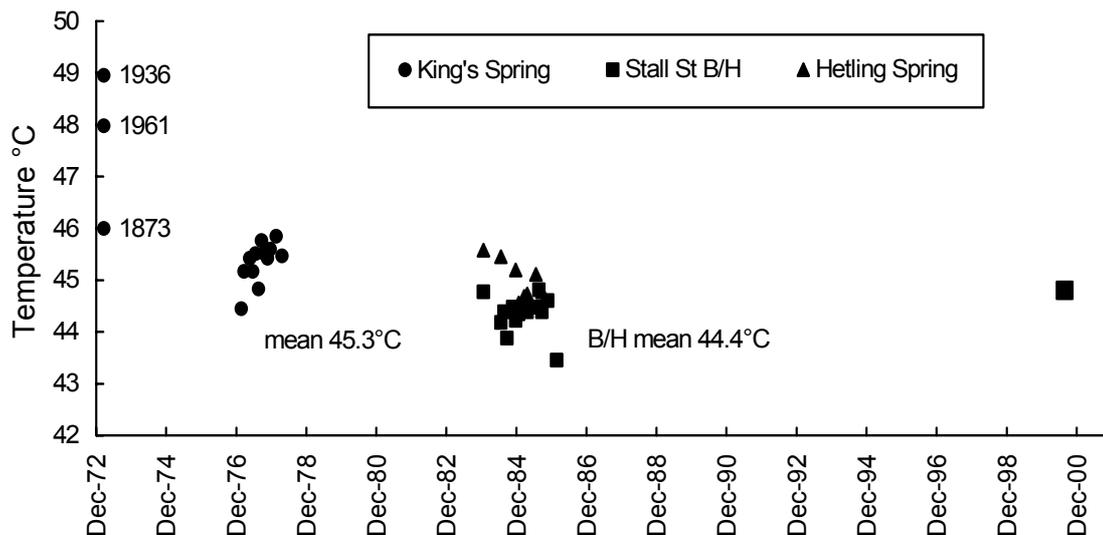**Table 4** **Isotope and gas data for the August 2000 sampling of the Stall Street borehole and Cross Bath.**

Site	δ^2H	$\delta^{18}O$	$\delta^{13}C$-DIC	^3H	^{14}C	$^{87}Sr/^{86}Sr$	N_2	O_2	Ar	CO_2	CH_4	N_2/Ar	CFC-11	CFC-12	CFC-113	^{39}Ar	^{37}Ar	^{85}Kr	^4He	$^3He/^4He$	Ne	^{222}Rn
	‰	‰	‰	TU	pmC	ratio	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	molar	pmol l^{-1}	pmol l^{-1}	pmol l^{-1}	pmA	dpm l^{-1} Ar	dpm l^{-1} Kr	cm^3 l^{-1} STP	ratio	cm^3 l^{-1} STP	Bq l^{-1}
Stall St B/H	-45.0	-7.14	-1.54	< 0.7	2.16	0.710551	23.5	< 0.7	0.52	36.3	0.053	45.3	0.17	< 0.01	0.02	15	0.14	600	3.59E-02	3.36E-08	3.29E-04	92.9
Cross Bath	-45.6	-7.21	-1.86		3.27	0.710550	29.3	< 0.7	0.65	46.9	0.071		0.26	<0.001	0.04							137.4

Table 5   **Comparison of the current analysis for the Stall Street borehole with earlier data for Stall Street and the King's Spring**

	Unit	Stall St B/H	Stall St B/H	Kings Spring
Year	-	2000	1986	1979
T	^{o}C	44.8	44.4	45.3
pH	-	6.71	6.70	6.65
Eh	mV	-2	variable	variable
Dissolved Oxygen	mg l^{-1}	<0.1	<0.1	<0.2
Ca	mg l^{-1}	385	390	382
Mg	mg l^{-1}	50.9	58	53
Na	mg l^{-1}	218	228	183
K	mg l^{-1}	19.9	18.1	17.4
Cl	mg l^{-1}	344	335	287
SO$_4$	mg l^{-1}	1080	1030	1032
HCO$_3$	mg l^{-1}	187	187	192
NO$_3$-N	mg l^{-1}	< 0.5	<0.05	<1
F	mg l^{-1}	2.28	2.0	2.08
Br	mg l^{-1}	1.863	2.0	2.02
I	mg l^{-1}	0.04	0.042	0.043
Si	mg l^{-1}	21.78	19.5	20.6
B	μg l^{-1}	516	590	590
Ba	μg l^{-1}	32	30	24
Fe	μg l^{-1}	774	1000	880
Li	μg l^{-1}	0.394	230	242
Mn	μg l^{-1}	45.6	50	68
Sr	μg l^{-1}	6601	6300	5920
δ^2H	‰	-45.0	-47	-47
$\delta^{18}O$	‰	-7.14	-7.4	-7.4
$\delta^{13}C$-DIC	‰	-1.54		-1.5
A^{14}C	pmC	2.16		4.5
$^{87}Sr/^{86}Sr$	ratio	0.710551		0.71075

**Table 6   Measured Ar and Kr concentrations and calculated values for *in-situ* production**

| Isotope	Measured values			Calculated values
	Isotope activity	Gas concentration cm^3 STP/cm^3 water	Isotope concentration atoms/cm^3 water	Isotope concentration atoms/cm^3 rock
^{39}Ar	15 ± 2.4 % modern Ar	$(4.81 \pm 0.03)\ 10^{-4}$ total argon	1.65 ± 0.28	Range $0.3 - 1.0$
^{37}Ar	0.14 ± 0.01 dpm/L Ar	$(4.81 \pm 0.03)\ 10^{-4}$ total argon	$(4.9 \pm 0.2)\ 10^{-3}$	$\sim 5\ 10^{-3}$
^{85}Kr	0.6 ± 0.1 dpm/cc Kr	$(1.07 \pm 0.01)\ 10^{-7}$ total krypton	0.52 ± 0.09	$\sim 1\ 10^{-4}$

Figure 1 **Temperature of the King's Spring and Cross Bath in relation to historical temperature measurements. Note the change in monitoring from the King's Spring to the Stall Street (King's Spring) borehole in 1986**
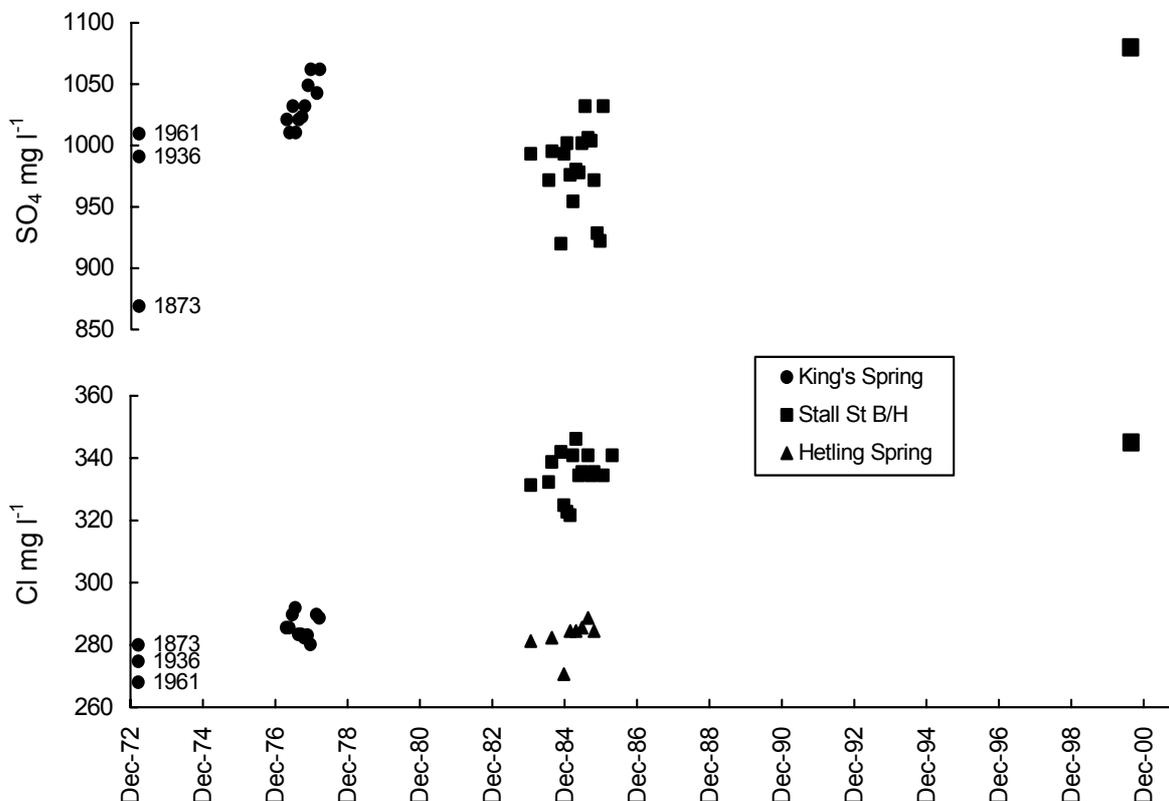
Figure 2 **Sulphate and chloride concentrations from the present study compared with historical measurements**
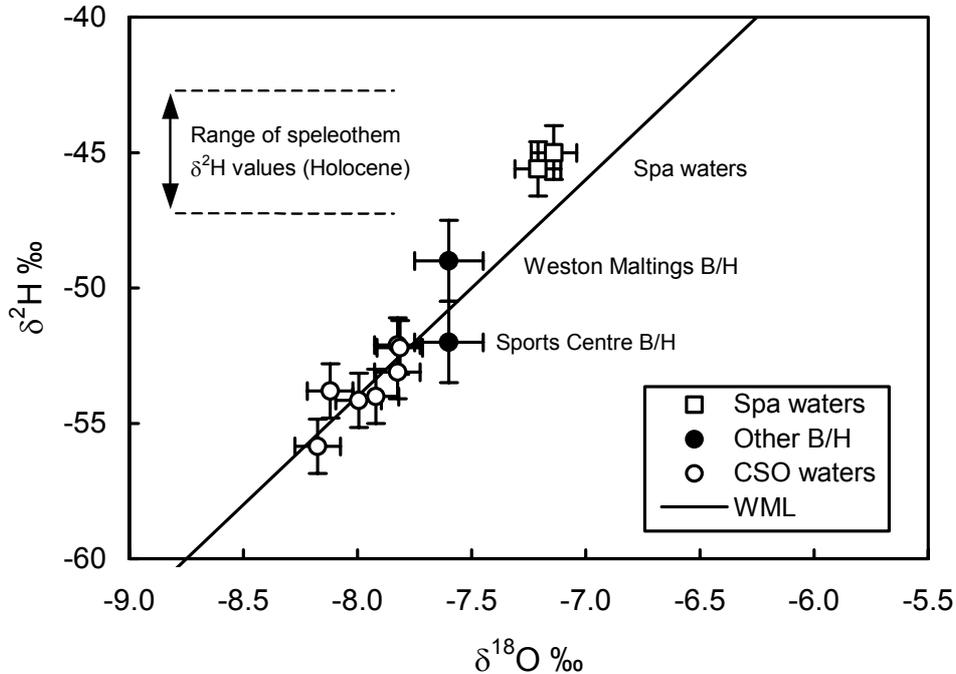
Figure 3 **Stable isotope diagram comparing present analyses with (i) Holocene speleothem δ²H values (Dennis et al., 1998), (ii) presumed Pleistocene palaeowaters from the CSO investigation boreholes in western Bath (Darling and Edmunds, 2001), and (iii) other boreholes apparently producing a mixture of old and modern waters. WML – world meteoric line.**
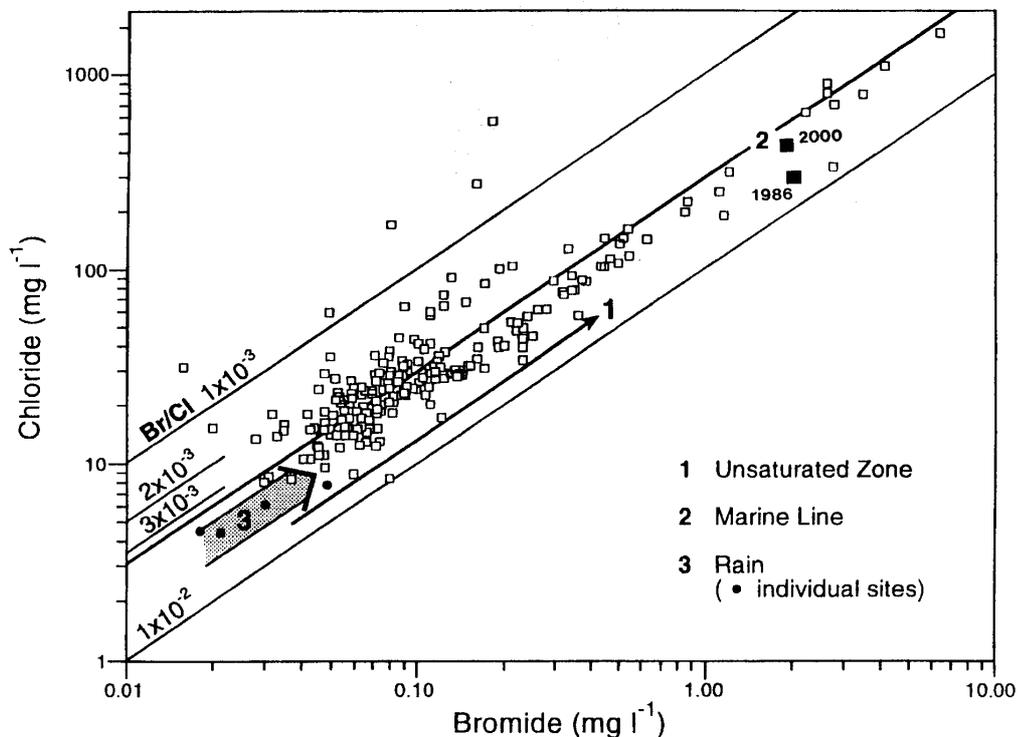
Figure 4 **Bromide/chloride relationships in British groundwaters, surface and rainwaters showing the relative compositions of the thermal water at Bath in 1986 and 2000. Bromide enrichment implies greater contribution of Cl from and organic-rich source and depletion in Br indicates a greater evaporite component.**
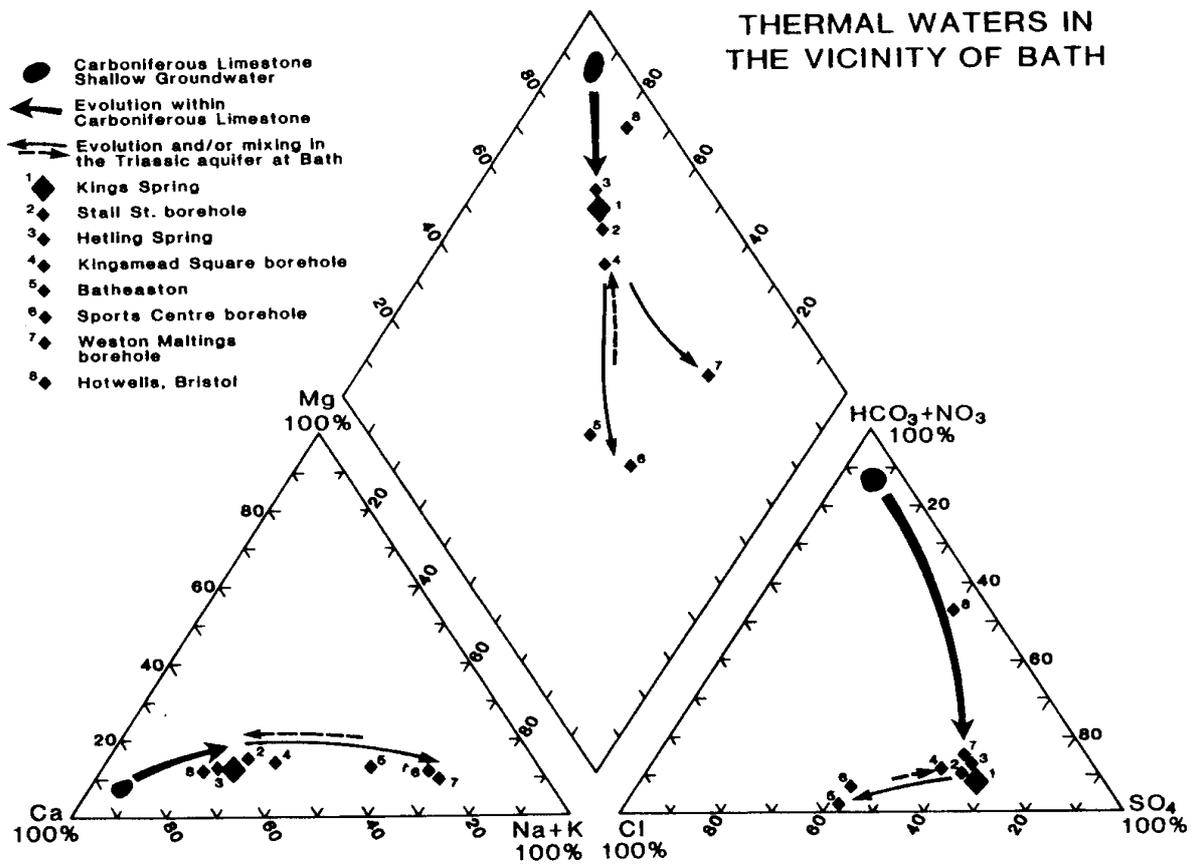
Figure 5   Trilinear plot for the major cations (taken from Edmunds and Miles 1991).

Appendix 1 Report of the recent sampling from the *Bath Chronicle*, 4 August 2000

Spa dream finally gets the green light

By Paul Wiltshire and Phil Chamberlain

BATH'S spa dream will finally begin to take shape this month.

Work is due to begin on August 29 on the £19m scheme to restore the city's best-known natural feature.

Politicians yesterday gave the go-ahead for council officials to sign a contract with building firm Mowlem for the £11.2m construction work.

But the question of who will operate the long-awaited facility once it opens its doors in 2002 remains unresolved.

Bath and North East Somerset councillors meeting behind closed doors yesterday approved the signing of legal documents with Mowlem, which will trigger a 20-month building contract.

This is another hurdle cleared by the Millennium Commission-funded project, which has been beset by rising costs and delays.

Councillors were also updated on the situation regarding the choice of operator for the attraction.

Dutch firm Thermae is now no longer the priority choice, after missing two deadlines by which it was supposed to come up with £5m to the council. The authority is exploring the possibility of using other firms to run the spa, although Thermae remains in the frame.

The company has written to project officers with a new financial offer and this letter is now being analysed by B&NES legal advisers.

Project director Paul Simons said the council was "at perfect liberty to negotiate with whoever we choose".

If Thermae's latest offer is regarded as legally watertight and a good deal by the lawyers, it could still be accepted. A decision on whether to do that will have to be taken by the full council, which has recently voted to underwrite the project to ensure it does go ahead.

Because no meeting of all the councillors on B&NES is scheduled to take place until October 12, a special one may have to be slotted in.

Meanwhile, sensitive equipment was installed in the centre of Bath yesterday in the latest effort to date and source the city's natural springs. Scientists from Britain and Switzerland worked from 8am to 10pm, filtering five tonnes of water and compiling a raft of data.

That will now go back to Switzerland and the results should be ready in the next six to 12 months.

Prof Mike Edmunds, from the British Geological Survey, was helping to lead the work which he described as getting a water fingerprint. By analysing the various elements within the water the scientists should be able to chart where it has been and its age.

Prof Edmunds said: "These investigations will provide the scientific basis, alongside hydro-geological studies, to explain where the waters are likely to have come from and how old they are."

Scientific investigations of the hot springs has been going on for some 175 years and they have been the object of fascination for philosophers and physicians for many years.

It is hoped that the results will help resolve the age of the water as well as questions including the depth at which the waters circulate, the likely source area and whether it is mixed with other, shallower sources.

■ **TESTING TIME:** Purtschert Roland in action with the water testing machine at the Roman Baths

PICTURE: Jon White