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## **6. 'There's no place like home' - No isotopic evidence for mobility at the Early Bronze Age cemetery of Singen, Germany**

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#### *Abstract*

The Early Bronze Age necropolis of Singen (Hohentwiel) located near Lake Constance represents a population from a period of technological transition in south-western Germany. The site contains several graves with metal artefacts which originated in other parts of Central and Western Europe, and therefore these could be interpreted as being the graves of non-local individuals. The purpose of this study was to investigate this possibility through the application of isotopic analysis. The ratios of strontium and oxygen isotopes in human enamel reflect the geological origin of food and drinking water consumed during enamel formation in early life stages. Additionally, the ratio of sulphur isotopes from bone collagen reflects the origin of foods consumed during the last ten to twenty years of life of an adult individual. We used these three isotope systems to attempt to identify local and non-local individuals at the site. We found that the isotope ratios of Sr, O and S of the humans were relatively homogeneous and generally correspond to the isotope signature of the local geology, climate and environment. We conclude that the sampled population is of local origin and does not show patterns of individual mobility, even though there is evidence for long-distance trade and exchange of the metal artefacts at this site.

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#### **6.1. Introduction**

The Early Bronze Age site of Singen (Hohentwiel) is the largest known Early Bronze Age period (EBA A1) cemetery in southern Germany (Harding, 2000; Krause, 1988). The site is located in the valley next to the Hohentwiel volcano, a prominent landmark within the hilly landscape of the Hegau region, north-west of the Lake Constance. During excavation campaigns in the 1950s a total of 96 graves were discovered, which can be divided into four to five distinct zones (Fig.



**Figure 6.1:** Map of the archaeological site of Singen am Hohentwiel and the graves sampled (grave 101 lies outside the mapped area), modified after Krause (1988).

6.1) and have been interpreted as kin-related groups. Many graves contained elaborate stone settings, and in some cases the distribution of wedging stones indicated the presence of wooden coffins (Krause, 1988). Radiocarbon dates of seven human bone samples from the site range in age from approximately 2200 to 2000 cal BC (Krause, 1988; Kromer, 1988).The overall preservation of organic material at Singen is poor and only the remains of approximately 30 inhumations could be recovered, some of them only represented by bone and tooth fragments. However, in most cases age and sex could still be estimated. This anthropological evidence supports a sex differentiated burial practice (females oriented south; males oriented north) and sex-specific distribution of grave goods. Females tended to be buried with awls, pins, a neck ring or

bracelets, whereas males were buried with bracelets, pins and a dagger at their waist (Sprenger, 1995). The few exceptions to this pattern have brought up speculations whether females buried with daggers could be widows taking over male roles in the family or non-local individuals that entered the community by marriage (Harding, 2000). The well preserved and rich assemblage of metal artefacts at the cemetery has become a reference collection for bronze artefact typology for

the north alpine Bronze Age. The copper alloys found at Singen revealed high levels of the trace elements antimony, nickel, arsenic and silver, and predates the knowledge of alloying with tin. This 'Singen metal' or 'Singen copper' is widely distributed within the European '*Blechkreis*' and beyond, from the western Alps to the Baltic Sea and the Carpathian basin (Harding, 2000; Kienlin and Stöllner, 2009; Krause, 2003). Findings of contemporary flanged axes made of 'Singen copper' in Swiss sites and around the Lake Constance have led to the hypothesis that the copper was mined in the Alps and the Singen community played an important role in the transfer of this metal, maybe even controlled its' trading north of the lake (Kienlin and Stöllner, 2009; Krause, 1988). Other specific features at the site of Singen are the four bronze daggers, referred to as so called 'Atlantic daggers'. Their style and *pointiliè* decoration resembles the Armorico-British style (type A) from the Wessex Culture in Great Britain (Fig. 6.2). The same type of dagger is also well known as the Loucè and Rumèdon type in coastal Brittany, France (Krause, 1988). Chemical metal analysis strongly supports this assumption and reports high levels of tin, typical for the Atlantic region (Christoforidis and Pernika, 1988; Krause, 1988). Additionally the surface of all four Singen daggers was treated with arsenic bronze, a procedure commonly found in contemporary daggers from Brittany (Krause, 1988).



**Figure 6.2:** Topographic map of Central Europe. The site and map section used in Figures 6.3 and 6.4 are marked with a dashed box. The areas of the 'Atlantic' Early Bronze Age cultures are patterned (after Krause, 1988).

These various lines of evidence have led to the assumption that the Singen community had far reaching connections in southern Germany and possibly even reaching the Atlantic coast or eastern parts of Europe. However, it remained unknown to what extent these connections required actual physical mobility of members of the Singen community. Did the transfer of metal objects or metallurgical expertise require long range movement of group members? Or did trading networks include consolidation and exchange in form of exogamic marriage systems? The aim of this study was to investigate possible mobility or migration by applying biochemical analysis to the human remains of the Singen cemetery. By analysing different complementary isotope systems, we sought to gain novel information on whether individuals originated locally or derived from other geographical and geological regions. Moreover analysing different tissues can provide isotopic information on different stages in life history like childhood and adolescence (tooth enamel), as well as the last ten to twenty years before death (bone). Isotope analysis has been applied in various regions and time periods to reconstruct human mobility and diet (Ambrose, 1993; Bentley, 2007; Price, 1989; Richards et al., 2008; Richards et al., 2000). Many studies have shown the potential of stable isotope analysis of strontium and oxygen to reveal individual mobility during life history (Evans et al., 2006b), or to prove that groups of people migrated due to their lifeways (Price et al., 2004), due to force (Schroeder et al., 2009), or that only a certain portion of a population was mobile, e.g. due to exogamic traditions (Bentley, 2007).

The stable strontium isotope  ${}^{87}Sr$  forms through radioactive decay of  ${}^{87}Rb$  in bedrock (Faure and Powell, 1972) and is measured in relation to the lighter isotope  ${}^{86}Sr$  ( ${}^{87}Sr/{}^{86}Sr$ ). The isotopic signature of a geological formation is determined by the age of the underlying rock, with older geological units having more radiogenic  ${}^{87}Sr/{}^{86}Sr$  ratios. The particular  ${}^{87}Sr/{}^{86}Sr$  signature of a geological area enters the biosphere by leaching and weathering and is finally absorbed by plants (Graustein, 1989). Animals feeding on these plants will incorporate the 'local'  $87\text{Sr}/86\text{Sr}$  signature in their bones and teeth because of the similar chemical properties of strontium and calcium (Ericson, 1985). Tooth enamel has shown to be the best substance for the analysis of  ${}^{87}Sr/{}^{86}Sr$  in archaeology as its compact structure is largely resistant to post-mortem diagenetic alteration and strontium uptake from the burial environment (Budd et al., 2000). The same applies to the analysis of stable oxygen isotopes. The ratio between the heavy and light isotope of oxygen ( $\delta^{18}$ O) is incorporated in the oxygen bonds of the enamel during formation and reflects the  $\delta^{18}$ O

in the local drinking water. Local  $\delta^{18}O$  values in water are determined by the local geography, climate and corresponding meteoric precipitation (Longinelli, 1984). The resulting local  $\delta^{18}O$ ratio relates to temperature, coastal proximity, as well as to latitude and altitude (Cuntz et al., 2002; Gat, 1980; Yurtsever, 1975), except if non-local drinking water is largely introduced by rivers or streams. Hence, the combination of  $\delta^{18}$ O and  $\delta^{87}$ Sr/ $\delta^{86}$ Sr signatures allows the reconstruction of human mobility and residential patterns (Bentley and Knipper, 2005). Tooth enamel is an ideal sampling tissue for this purpose, as it does not change its isotopic composition once it is formed (Humphrey et al., 2008). In humans the anterior teeth and the first molar are formed in the first years of life, whereas the premolars and second molars form in childhood, and third molars may not be completely formed until adolescence (Hillson, 1996; Reid and Dean, 2006). Therefore,  ${}^{87}Sr/{}^{86}Sr$  and  $\delta {}^{18}O$  in enamel reflect the residence of an individual during early life stages (childhood/adolescence) and does not carry information about the area of residence in adulthood.

Similar to strontium, sulphur isotopes in body tissues are also related to the isotopic signature of the local geology. Sulphur isotopes can be measured in bone collagen, which remodels more or less constantly during life (Hedges et al., 2007). Therefore, depending on the sampled bone, sulphur isotopes can be used to trace the provenance of the last years of an individual's life (Richards et al., 2001; Richards et al., 2003). Some bones however may reveal much slower turnover rates than others. Bone of the skull for example may not have completely remodelled after childhood (Geyh, 2001; Wild et al., 2000). Sulphur isotope ratios ( $\delta^{34}$ S) in bone collagen strongly correspond to dietary protein sources and give particular insight into whether terrestrial, freshwater or marine foods were preferentially consumed (Nehlich et al., 2010; Nehlich et al., submitted; Privat et al., 2007; Richards et al., 2003). Vika (2009) was able to identify immigrants to Bronze Age Thebes by using sulphur isotope analysis. She showed that humans and animals had the same sulphur isotopic signature, but one individual had a significantly more  $34$ S-depleted isotopic composition, which was concluded to result from non-local food sources, therefore this individual immigrated to ancient Thebes.

#### **6.2. Materials**

We sampled archaeological human bone, dentine and enamel for isotope analyses. Due to limited skeletal preservation at the site, only 29 individuals could be sampled. However, we could sample individuals from all of the four major grave groups in the cemetery (Fig. 6.1). Unfortunately, three out of four of the burials with the 'Atlantic' daggers (graves 60, 67, 76 and 84) did not contain preserved skeletons for analysis. In total we sampled bone from 29 burials, yet only 22 of these also contained teeth for sampling. In two subadult individuals (grave 6 and 66) we could sample one deciduous and one permanent tooth from the jaw, resulting in a subset of 24 tooth enamel samples in this study. All information on age and sex (Tab. 1) was taken from Gerhardt (1964) with some additions by J. Wahl (Krause, 1988). We were only able to measure the  $\delta^{18}$ O of nine enamel samples due to sampling, funding and measurement limitations. The aim of  $\delta^{18}$ O analysis was to find further evidence of local or non-local origin for those individuals which yielded the most heterogenic  ${}^{87}Sr/{}^{86}Sr$  signatures in our sample set, in comparison to a number of individuals with presumably very 'local'  ${}^{87}Sr/{}^{86}Sr$  signals. For measurement of  $\delta^{18}O$ we therefore selected those samples which showed the 'highest' (graves 65, 74 and 77) and 'lowest' (graves 55, 73 and 80)  ${}^{87}Sr/{}^{86}Sr$  values as well as samples which revealed 'intermediate/local'  ${}^{87}Sr/{}^{86}Sr$  values (graves 19, 57 and 70). Unfortunately, animal bones or teeth for comparison were not recovered at the site of Singen. For the analysis of  ${}^{87}Sr/{}^{86}Sr$  and  $\delta^{18}O$  we sampled tooth enamel because enamel reflects biogenic strontium and oxygen incorporated during tooth formation and is resistant to contaminations due to its dense structure (Budd et al., 2000; Hoppe et al., 2003). We also randomly sampled dentine of eight individuals as a proxy of local soluble strontium in the burial environment, as  ${}^{87}Sr/{}^{86}Sr$  values in dentine are likely affected by diagenetic uptake of soil derived strontium (Budd et al., 2000). However, stable isotope ratios of associated fauna are a valuable proxy for the bioavailability of strontium, oxygen and sulphur in a given environment (Bentley and Knipper, 2005; Craig et al., 2006; Price et al., 2002). Alternative sampling of animal bones from several other Bronze Age sites in the proximity of Singen was also not possible because material was not accessible in the depots of the State Office for Heritage Management and Archaeology, Konstanz, Germany. To assess the bioavailable strontium isotope ratios of the region between the Lake Constance and southern Black Forest, we collected a variety of modern snail shells and plants in June 2009 (Fig. 6.3).

Snails have a limited range of movement and are therefore an ideal candidate to detect the local variability in bioavailable strontium (Evans et al., 2010; Price et al., 2002). Strontium is deposited in the shell, as it substitutes for calcium, which is the main component of snail shell (Rosenthal et al., 1965). Plant strontium values reflect the mobile strontium in the local soil in different root depths as well as the strontium introduced by rainwater and atmospheric dust (Evans et al., 2009).

During field sampling, the different major geological formations in the region were located using geological mapping information (LGRB maps dGk25s: 7916 Villingen-Schwenningen-West, 8218 Gottmadingen, 8219 Singen (Hohentwiel), by the Landesamt für Geologie, Rohstoffe und Bergbau, Freiburg). In each geological unit we selected elevated forest patches where anthropogenic contaminations (e.g. fertilizers, traffic pollutions) are unlikely. At each sampling



**Figure 6.3:** Geological map of the study area in south-western Germany. The archaeological site of Singen is marked with a star. The black dots mark the sites of environmental sampling. Information on the mean  ${}^{87}Sr/{}^{86}Sr$ value and number of samples are reported for each sampling location:

Überlingen am Ried I-II (1-2), Gottmadingen (3), Hohentwiel I-III (4-6), Zimmerholz (7), Aulfingen (8-9), Espasingen (10), Hecheln (11), Pfaffenweiler (12), Magdalenenberg (13-14), Weilersbach (15-16), Tuningen (17- 18), Hochemmingen (19-20), Furtwangen (21-22), Triberg (23-24).

location (recorded using GPS) snail shells were collected alongside botanical samples from a deep rooting deciduous tree, a shallower-rooted shrub and a shallow rooted terrestrial herb. We did not sample modern environmental samples for oxygen and sulphur, because  $\delta^{18}O$  values may vary through time according to climate changes, and modern samples for  $\delta^{34}S$  are likely affected by anthropogenic sulphur pollutants (Krouse et al., 1991).

#### **6.3. Methods**

Strontium was extracted and purified from tooth enamel and dentine as well as plants and snail shells following the ion exchange method outlined by Deniel and Pin (2001) at the clean laboratory and MC-ICP-MS facility at the Max Planck Institute for Evolutionary Anthropology in Leipzig, Germany (Richards et al., 2008). First, tooth samples were manually cleaned with a dental drill to remove superficial contaminations. Then, after cutting a chip of the tooth crown, the enamel was mechanically separated from attached dentine. The opposite procedure was applied to dentine samples, where the attached enamel was removed. The pieces of enamel and dentine were then cleaned by rinsing and in an ultrasonic bath with deionised water. Samples were transferred to the clean laboratory, rinsed in ultrapure acetone and dried overnight. Subsequently ~10-20mg of enamel or dentine was weighed into clean Teflon beakers and digested in 1ml of 14.3M HNO<sub>3</sub> on a hotplate (120 $^{\circ}$ C). Snail shells were cleaned by repeated rinsing (and ultrasonic cleaning) with deionised water to remove attached sediments. Then each sample (1-4g) of snail shell or plant leaves was combusted at  $800^{\circ}$ C in clean ceramic beakers for 12 hours. The remaining ash was transferred to the clean laboratory where 10-50mg of ash was weighed into clean Teflon beakers and digested in 1-2ml of  $14.3M HNO<sub>3</sub>$  on a hotplate (120 $^{\circ}$ C).

The dissolved samples of enamel, dentine and ash were evaporated to dryness and were combined with 1ml of 3M HNO3 before being loaded on clean, pre-conditioned 2ml columns containing cleaned Sr-spec<sup>TM</sup> resin (EiChrom, Darien, IL, USA). Samples were reloaded three times to maximize the amount of strontium attached to the resin. After several washes with 3M HNO<sub>3</sub>, the strontium was eluted from the resin with ultrapure deionised water into clean Teflon beakers and dried down on a hot plate. The remaining samples, again re-dissolved in  $3\%$  HNO<sub>3</sub>, were then ready for the measurement parallel to the standards SRM\_987 and SRM\_1486, as well

as one beaker blank per run, in a Thermo Fisher Neptune<sup>TM</sup> MC-ICP-MS instrument (Thermo Fisher Scientific Inc., Dreieich, Germany).

For the analysis of  $\delta^{18}O$ , we selected nine individuals. We extracted PO<sub>4</sub> radicals out of enamel bioapatite by applying the modified silver phosphate precipitation method (Dettmann et al., 2001; O'Neil et al., 1994). First 10-15 mg of tooth enamel was cut from the tooth crown, manually cleaned with a dental drill and then ground to fine powder. The sample was then dissolved in 1ml 2M HF. After 24 hours, the samples were centrifuged and the solution containing the phosphate was transferred into a new tube where  $300\mu$  of NH<sub>4</sub>OH was added to buffer the HF. Several drops of BTB (Bromothymol blue) was previously added to check the pH ( $\le$ 7). When the sample was neutral,  $\sim$ 700 $\mu$ l 2M AgNO3 was added. Subsequently the silver phosphate crystals precipitated corresponding to the decrease in  $pH$ , while  $NH<sub>3</sub>$  was discharged from the solution. The resulting residue, consisting of Ag3PO4 crystals of light yellow colour, was centrifuged and rinsed with deionised water four times. The residue was then dried down in a freeze dryer. The measurement of the  $Ag_3PO_4$  samples in duplicates was conducted in the Department for Hydrology at the Helmholtz Centre for Environmental Research - UFZ, Halle, Germany. After weighing  $\sim 700 \mu g$  Ag<sub>3</sub>PO<sub>4</sub> into silver capsules,  $\sim 0.5$ mg of graphite was added (Vennemann et al., 2002). The capsules were then combusted to CO in a HekaTech hightemperature combustion oven with helium carrier gas at 1450 °C. The CO was lead via a Thermo Finnigan ConFlow III into a Thermo Finnigan DeltaXLplus IRMS (Thermo-Finnigan®, Bremen, Germany) for isotope analysis. Measurement precision was controlled using two duplicates of the commonly accepted NBS 120c standard, as well as external (Durham horse enamel) and internal laboratory standards.

To analyse sulphur isotope ratios, we extracted collagen from 29 human bone samples. The collagen extraction followed a modified Longin method (Brown et al., 1988; Collins and Galley, 1998; Longin, 1971). Bone samples were cleaned by air abrasion and then demineralized in 0.5M HCl for several weeks at 4°C, with acid changes every few days. Demineralized samples were then rinsed three times with de-ionized water and gelatinized at 70°C in a pH3 solution for 48 hours. The insoluble fraction was first filtered with a  $5\mu$ m EZEE<sup>©</sup> filter, and then again filtered using Amicon<sup>©</sup> ultra filters (>30kDa). The purified solution was frozen and freeze dried for 48 hours. Finally, 10mg of dried collagen sample was weighed into tin capsules. The measurement was performed in duplicates in a HekaTech EuroVector coupled to a Delta V plus

mass spectrometer (Thermo-Finnigan®, Bremen, Germany) at the Max Planck Institute for Evolutionary Anthropology in Leipzig, Germany.

#### **6.4. Results**

#### *6.4.1. Strontium*

The repeated <sup>87</sup>Sr<sup>86</sup>Sr measurement of the standard SRM 987 resulted in an average value of  $0.710268 \pm 0.000026$  ( $2\sigma$ , n = 24) and was subsequently corrected to the accepted value of  $0.710240 \pm 0.00004$  (Johnson et al., 1990; Terakado et al., 1988). Total procedural blanks, one for each batch of 13 samples, were considered negligible. The  ${}^{87}Sr/{}^{86}Sr$  measured in 24 enamel samples ranged from 0.70740 to 0.70940 with a mean of 0.70838  $\pm$  0.00044 (1 $\sigma$ ). This mean  $87\text{Sr}$ <sup>86</sup>Sr value for enamel is almost identical with the mean value of  $87\text{Sr}$ <sup>86</sup>Sr measured in the dentine samples (0.70827  $\pm$  0.00028, 1 $\sigma$ , n=8), which likely reflect the soil <sup>87</sup>Sr<sup>/86</sup>Sr signature. Finally, the deciduous and permanent molars of two infants revealed  ${}^{87}Sr/{}^{86}Sr$  ratio pairs that were almost identical (grave 6: 0.70833 and 0.70836; grave 66: 0.70877 and 0.70853). For further details see Table 1. Environmental samples were obtained from 24 sampling sites in 11 geological units, reaching from the Swiss-German border south of the site of Singen, to approximately 70km north-west in the southern Black Forest (Fig. 6.3). The mean values as well as more details on sampling sites for each geological unit are presented in Table 2.

#### *6.4.2. Oxygen*

Oxygen isotope ratios, reported relative to the relative to the international standard VSMOW (Vienna Standard Mean Ocean Water). The measurement error calculated from the standard materials was less than 0.6‰. The  $\delta^{18}$ O values measured in the NBS 120c standards were 21.4 ± 0.3‰ and  $21.8 \pm 0.1\%$  (1 $\sigma$ ), which is in agreement to a value of  $21.7 \pm 0.5\%$  reported for NBS 120c from other laboratories (summarized in Chenery et al. (2010). The duplicate measurement of an external lab standard (Durham horse enamel) yielded values of  $14.7 \pm 0.5\%$  and  $14.6 \pm 1.6\%$ 0.6‰ (1 $\sigma$ ). The average reproducibility of the analysis of human enamel was better than 0.30  $\pm$ 0.2‰ (1 $\sigma$ ). The  $\delta^{18}$ O ratios measured in the nine human enamel samples ranged from 13.8‰ to 16.6‰ with a mean of 15.5‰ ( $\pm$  0.8, 1 $\sigma$ , n=9).

**Table 6.1:** Individual data (age, sex, bone/tooth sampled), collagen quality criteria (S wt%, C:S, N:S, %collagen) and isotope data  $(\delta^{34}S, {^{87}Sr}^{86}Sr)_{\text{enamel}}$ ,  ${^{87}Sr}^{86}Sr$  <sub>dentine</sub>,  $\delta^{18}O$ ) for each grave. The radiocarbon dates are reported after Krause (1988), the grave marked with an asterisks contained an 'Atlantic' dagger. infans=1-13 years, infans II=6-13 **8 7Sr/8 6Sr 8 7Sr/8 6Sr 1 8O** 

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**Table 6.2:** Details on the environmental sampling in the Hegau and Black Forest region. The sampling sites are named after the neighbouring village. For each sampling location the type of sample, species and GPS coordinates are reported next to a description of the geological conditions and the results of the <sup>87</sup>Sr/<sup>86</sup>Sr analysis.





**Table 6.2:** continued.

#### *6.4.3. Sulphur*

We extracted sufficient amounts of collagen ( $>9$ mg) for sulphur isotope analysis out of 23 human bone samples. Six bone samples had insufficient collagen yield for sulphur isotope analysis. The total amounts of extracted collagen (% collagen), atomic ratios (C:S, N:S) and measures of the sulphur weight% (S wt%) are presented in Table 1. All 23 samples meet the recommended quality criteria for collagen (Ambrose, 1990; DeNiro, 1985). The ratios of C:S and N:S meet the recommended values of  $600 \pm 300$  and  $200 \pm 100$  respectively, and the weight % of sulphur in the collagen ranges between 0.15 and 0.35 (Nehlich and Richards, 2009). The analytical error, calculated from repeated analysis of internal and international standards, was less than  $\pm$  0.6‰ for  $\delta^{34}$ S. Sulphur isotope ratios, scaled against the standard V-CDT, measured in human collagen ranged from  $-0.6\%$  to  $+3.4\%$  (mean  $+1.2 \pm 0.9\%$ , 1 $\sigma$ ).

#### **6.5. Discussion**

#### *6.5.4. Environmental samples*

Most of the environmental background samples had  ${}^{87}Sr/{}^{86}Sr$  values that mainly reflected the expected  ${}^{87}Sr/{}^{86}Sr$  isotope values of the underlying bedrock, while a number of environmental samples had  $87\text{Sr}/86\text{Sr}$  ratios that were outside of the expected values. The Hegau region (sample locations 1-11) west of the Lake Constance is the result of the formation and modification of the Alps. The region is dominated by alluvial sediments (1-2), molasses (7) and moraines from different glacial periods (3, 10-11). Except for the volcanic tuffs (4-6), these sediments revealed consistent <sup>87</sup>Sr/<sup>86</sup>Sr signatures with an average of 0.70919 ( $\pm$  0.00098, 1 $\sigma$ , n=22), reflecting relatively young geological formations.

The alluvial sediments surrounding the site of Singen were sampled at Überlingen am Ried (1-2) and revealed values of 0.70983 ( $\pm$  0.00085, 1 $\sigma$ , n=5), similar to what was measured in biological samples from alluvial sediments by Price and colleagues (2003). However, it is important to note that the sample location of Überlingen am Ried is dominated by Holocene sediments superposed on the Pleistocene gravels. This stratigraphy likely caused slightly more radiogenic values in the deep rooting trees of this subset (1c and 1d), whereas the snails (1a and 2a) seem to have sourced less radiogenic  ${}^{87}Sr/{}^{86}Sr$ , which is more representative for the more recent overlying sediments at Überlingen am Ried. The present day city of Singen itself is located in the alluvial sediments

(Fig. 6.3 and 6.4), while the burial site of Singen itself is likely partly covered by smaller extensions of the Würm moraine (represented by the sample location 10) and therefore also has been influenced by strontium from the moraine. The Würm moraine itself had a slightly lower mean <sup>87</sup>Sr<sup>/86</sup>Sr signature of 0.70892 ( $\pm$  0.00022, 1 $\sigma$ , n=4) which matches well with the <sup>87</sup>Sr<sup>/86</sup>Sr ratios we measured in the human dentine  $(0.70827 \pm 0.00028, 1\sigma, n=8)$ .



**Figure 6.4:** Geological map of the study area in south-western Germany with the according range of  $87\text{Sr}^{86}\text{Sr}$  ratios measured in snail shells, herbs/shrubs and trees, as indicated by the according symbols.

The lowlands of the Hegau are interrupted by very small scale volcanic outcrops, e.g. the Hohentwiel volcano (4-6) west of the city of Singen. Samples from this volcano revealed the lowest <sup>87</sup>Sr/<sup>86</sup>Sr values measured in this study. Here, the three different sample locations at the volcano demonstrate a range of  ${}^{87}Sr/{}^{86}Sr$  values at one single geological site: Probably due to weathering and leaching of the rock, the 'lowest' values were measured in biosphere samples from the foot of the volcano (4a-e) with a mean of 0.70628 ( $\pm$  0.00067 1 $\sigma$ , n=5). Slightly higher values (mean  $0.70767 \pm 0.00038$ ,  $1\sigma$ , n=4) were measured at the volcano's flank (5) and top (6), which are dominated by phonolite rocks.

A strip of Late Jurassic limestone bordering the Hegau to the west revealed a mean  ${}^{87}Sr/{}^{86}Sr$ value of 0.70773 ( $\pm$  0.00052, 1σ, n=13) for the sample location Aulfingen (8-9). At Aulfingen we see only a small variation in  ${}^{87}Sr/{}^{86}Sr$  among the 13 different samples, indicating that we likely sampled biological materials representative of the underlying geology. Even though the sampled forest patch at Aulfingen may also contain small outcrops of molasse, the values measured here are similar to what is reported for Jurassic deposits elsewhere (Horn et al., 1985; Price et al., 2004). Further north/northwest of the Hegau region the lithostratigraphy of the landscape changes to the middle Jurassic *Braunjura* and *Keuper* strata (17-20) of the Neckar valley, which had a mean  ${}^{87}Sr/{}^{86}Sr$  value of 0.70944 ( $\pm$  0.00090 1 $\sigma$ , n=13). One maple tree (18d) in the *Braunjura* at Tuningen revealed unusual radiogenic values of 0.71143, which is unusual and we have no explanation for this, as the geology in this area is very homogeneous. If the result from this tree is excluded, the  ${}^{87}Sr/{}^{86}Sr$  values measured in the remaining 12 samples range from 0.70830 to 0.71059, resembling what was reported by Bentley and colleagues (Horn pers. comm. in Bentley et al., 2003). Further westwards are the foothills of the Black Forest with the characteristic *Muschelkalk* and *Buntsandstein* deposits. In the uniform *Muschelkalk* area at Weilersbach (15-16) we found very variable  ${}^{87}Sr/{}^{86}Sr$  values, ranging from 0.70873 to 0.71175. Strontium isotope data reported for the *Muschelkalk* in southwest Germany range broadly between 0.708 and 0.709 (Price et al., 2003). The data from Weilersbach I (15) suggests that data measured in the plants and the two large snails (*Helix pomatia*) are representative for the *Muschelkalk*, whereas the more radiogenic  ${}^{87}Sr/{}^{86}Sr$  value of 0.71175 measured in the whitelipped snail (15a) is likely due to Sr contamination, as the snail could possibly have been feeding on fertilised agricultural fields, which are located several hundred meters away from the sampling spot. At Weilersbach II (16) however, the highest value of 0.71055 is measured in a wild strawberry (16c), while the four remaining samples of snails and deeper rooting plants reveal a lower mean value of  $0.70921 \pm 0.00042$  (1 $\sigma$ ). It is possible that the strawberry was growing close to the forest path (which are often paved with non-local sediments) and may therefore not be representative. If these two samples (15a and 16c) are excluded, the range is 0.70873 to 0.70973 and the mean is 0.70914 (± 0.00038, 1σ, n=9) for the *Muschelkalk* at Weilersbach, which meets what was reported in previous studies (Price et al., 2003). The *Buntsandstein,* a red sandstone of the Black Forest, was sampled in two locations (12 and 13-14) which revealed distinct  ${}^{87}Sr/{}^{86}Sr$  signatures. For Pfaffenweiler (12), a snail and a tree were

measured, resulting in a mean of 0.71095 ( $\pm$  0.00084, 1 $\sigma$ , n=2). Thus, the values measured for the archaeological site of Magdalenenberg (13) and the adjoining 'Laible' forest patch (14) are much more radiogenic. Magdalenenberg is located on a small accumulation of *Buntsandstein*, which had pushed through the *Muschelkalk* bedrock and formed a small hill. Contamination of the soil by leached fertilizers appears unlikely, as the sampling loci are elevated relative to the surrounding fields. The average  ${}^{87}Sr/{}^{86}Sr$  signature measured for Magdalenenberg is 0.71345 ( $\pm$ 0.00141,  $1\sigma$ ,  $n=6$ ). The range of values measured at Pfaffenweiler and Magdalenenberg compares well to what is reported for *Buntsandstein* bedrock (Bentley et al., 2003; Horwath, 2000; Price et al., 2003) but exceeds the values measured in the biosphere (Price et al., 2003). Much older geological units with even higher  ${}^{87}Sr/{}^{86}Sr$  values for the biosphere appear approximately 40km northwest of the site of Singen in the more radiogenic gneiss (mean  $0.71453 \pm 0.00313$  1 $\sigma$ , n=8) and granite bedrocks  $(0.71525 \pm 0.00293$  1 $\sigma$ , n=9) of the Black Forest, with values similar to what was reported for the bedrocks' isotope signatures (Baumann and Hofmann, 1988; Price et al., 2003). At the sampling location Furtwangen, a region strongly dominated by gneiss bedrock, we sampled in two locations, one in higher altitudes (21), and one in the foothills next to a small stream (22). The higher altitude sample location had much more radiogenic  ${}^{87}Sr/{}^{86}Sr$  signatures ranging from 0.71502 to 0.71877 and with a mean of 0.71715 ( $\pm$ 0.00186,  $1\sigma$ , n=4), which can be expected from older metamorphic rocks (Baumann and Hofmann, 1988). In the foothills of the gneiss however, we measured much lower  ${}^{87}Sr/{}^{86}Sr$ values (mean  $0.71192$ ,  $\pm 0.001071$ , n=4), which are less representative for gneiss. We suggest that we sampled a very small scale geological outcrop of a much younger geological stratum, which is likely caused by the small stream and is not marked on the geological maps. In the granite mountainous region above the waterfall of Triberg (23-24) we gained the highest  $87\text{Sr}/86\text{Sr}$  value of our biosphere sampling. This value was measured in the leaves of deep rooting tree (24d with 0.72190), indicating that much of the  ${}^{87}Sr/{}^{86}Sr$  utilized by the tree derived almost directly from the underlying granite rock. More shallow rooting plants on the other hand revealed much lower values, suggesting a more mixed sourcing for  ${}^{87}Sr/{}^{86}Sr$ .

The critical aspect of the application of strontium isotope analysis of archaeological remains is to determine which isotope signature is local and which is not (Bentley et al., 2004; Budd et al., 2004). In this study, we mapped the landscape surrounding the archaeological study site to reveal

the local and the more remote  ${}^{87}Sr/{}^{86}Sr$  signatures. We found a quite homogeneous isotopic pattern for the site and the surrounding area of approximately 20km, if the small scale volcanic tuffs are excluded (Fig. 6.3, 6.4 and 6.5). Only the Hohentwiel volcano featured less radiogenic  $87\text{Sr}$ <sup>86</sup>Sr values (0.70570 to 0.70822), yet we can exclude that growing crops on the steep volcanic slopes was relevant for the local subsistence, despite the volcanic tuffs seem to have been relevant as pastures for livestock in the Neolithic (see below). The  ${}^{87}Sr/{}^{86}Sr$  signature of the landscape becomes significantly more radiogenic 40km northwest of the site of Singen in the *Buntsandstein* and the older rocks of the gneiss and granite.

#### *6.5.5. Strontium isotope signatures in human enamel*

Although the cemetery of Singen was used over a time period of more than 200 years, the values we measured in the human enamel appear quite homogenous. We suggest that the overall variation in  ${}^{87}Sr/{}^{86}Sr$  of 0.02000 within the 24 enamel samples can be explained by the consumption of plants and animals from local geologies close to the site. Additionally, none of the Singen individuals featured  ${}^{87}Sr/{}^{86}Sr$  ratios higher than 0.70940, and hence none showed clear evidence for distant residence during the period of tooth formation (Fig. 6.5). All individuals incorporated an isotopic signature similar to what is present at the cemetery and its surroundings, leading to the conclusion that the location of residence and the source of everyday diet were closely linked to the Hegau region and the western shores of the Lake Constance, respectively. Given the strong correlation between the enamel and dentine  ${}^{87}Sr/{}^{86}Sr$  ratios measured in eight individuals, mean values of 0.70830 ( $\pm$  0.00036, 1 $\sigma$ ) and 0.70827 ( $\pm$  0.00028, 1σ), one could suggest that the area of residence was located in the proximity of the cemetery, or at least on very similar alluvial sediments and Pleistocene moraines. Thus, our finding accords to the discovery of pottery fragments near the cemetery site, which have been suggested to indicate the presence of a related settlement (Krause 2001).

Additional insights into the life histories of two infant individuals (graves 6 and 66) were possible, although their poor skeletal preservation did not allow a precise age estimate. From the deciduous molars we found that the residence of their mothers was local during pregnancy and first few months of breastfeeding. From the  ${}^{87}Sr/{}^{86}Sr$  ratios measured in the crowns of two nonerupted permanent molars, we can reconstruct evidence that the children's residence remained the same over the following years (Humphrey et al., 2008; Reid and Dean, 2006).

Further data for the determination of the local bioavailable  ${}^{87}Sr/{}^{86}Sr$  comes from archaeological fauna from prehistoric sites in direct proximity to the Singen cemetery. Bentley and Knipper (2005) analysed pig enamel samples from the Neolithic sites Hilzingen and Singen-Offwiese



**Figure 6.5:** Strontium isotope ratios measured in human enamel (black diamonds), human dentine (gray squares) and Neolithic pigs (back cross, data reported by Bentley & Knipper, 2005). The grey bars represent the bioavailable  ${}^{87}Sr/86Sr$  ratios measured for the geological units in this study within a ~25km radius around the site of Singen.

(<4km distance). The mean  ${}^{87}Sr/{}^{86}Sr$  value of all three pigs is 0.708 (Bentley and Knipper, 2005), which would resemble the human enamel data from Singen. However, one of the Hilzingen pigs had a more radiogenic of  ${}^{87}Sr/{}^{86}Sr$  value of 0.711 which could not have been formed from consuming foods from the local geology around the site of Singen. According to our biosphere data, a possible source of this higher Sr isotope signal could be the Riss moraine northwest of the Singen site or the *Keuper*, *Braunjura* or *Muschelkalk* layers to the west (Fig. 6.5). A second pig from Hilzingen had a much lower  ${}^{87}Sr/{}^{86}Sr$  value of 0.706 which may reflect feeding on the slopes of the Hohentwiel volcano or the neighbouring volcanic tuffs (<2km distance). A similar pattern is found in the pig from Singen-Offwiese which had a  ${}^{87}Sr/{}^{86}Sr$  signature of 0.707 (Bentley and Knipper, 2005) and may also have derived from a region of volcanic tuffs (Fig. 6.5). From our modern environmental samples we infer that, due to the potential of pigs to feed

in more remote or forested pastures, archaeological pig data might not always give a good proxy for humans, in terms of the local bioavailability of strontium.

Modern isotope data for comparison with the human enamel data in this study derives from the Lake Constance itself. Radiocarbon dated sediment cores from the lake revealed very homogeneous  ${}^{87}Sr/{}^{86}Sr$  values for the Holocene. For the period of the Bronze Age the values measured in crustacean shell material ranged between 0.786 and 0.788 (Kober et al., 2007). Although strontium uptake from drinking water is negligible, these values give a good proxy of the lake basin geology, which is tertiary molasse. Modern  ${}^{87}Sr/{}^{86}Sr$  data from molluscs collected in Lake Constance had values of 0.7085 and 0.7084 (Buhl et al., 1991), which are similar to the mean <sup>87</sup>Sr/<sup>86</sup>Sr values for the Singen people (0.70838  $\pm$  0.00044). Nevertheless, we find similar  $87\text{Sr}/86\text{Sr}$  signatures in other parts of Europe, e.g. in Britain (Chenery et al., 2010), the Carpathian Basin (Giblin, 2009), other areas of southern Germany (Price et al., 2006; Schweissing and Grupe, 2003) and in the Swiss region between Lake Constance and Lake Zurich (Tütken et al., 2008). Therefore, although unlikely it is possible that if people had migrated from these other



**Figure 6.6:** The <sup>87</sup>Sr/<sup>86</sup>Sr ratios plotted against the  $\delta^{18}$ O values measured in human enamel phosphate (n=9). The dashed box indicated the predicted local  $\delta^{18}$ O range (mean value  $\pm 1\sigma$ ), and the  $\pm 0.6\%$  represents the measurement error (1 $\sigma$ ). The data from the three local Neolithic pigs (x) are taken from Bentley & Knipper (2005).

areas to Singen, they could have similar  ${}^{87}Sr/{}^{86}Sr$  values to the local values, and therefore mistakenly be identified as local. Therefore, we also measured isotope ratios of other chemical elements to further confirm our conclusions that the humans we sampled from Singen are indeed local.

#### *6.5.6. Oxygen isotope ratios in human enamel*

Oxygen isotope ratios of enamel are related to the oxygen isotope composition of drinking water. Lake Constance serves as the major drinking water reservoir in the region and is a good proxy for the local oxygen isotope composition of the Hegau region, including its lakes, streams and groundwater. If the Early Bronze Age population from Singen lived in the Hegau area west of Lake Constance, as their strontium isotope ratios suggest, their oxygen isotope ratios should match the local water and meteoric precipitation oxygen isotope values. We found quite consistent  $\delta^{18}$ O values in the human enamel from Singen (mean 15.5 ± 0.8‰, 1 $\sigma$ , n=9) with two outliers (Fig. 6.6). One individual from Grave 74 had a  $\delta^{18}$ O value of 13.8‰ which is outside the average and 1σ range of our data set. This data point should be considered with caution, because the measurement error of this sample was high  $(0.96\%$ ,  $1\sigma$ , a-measurement: 13.11‰, bmeasurement: 14.47‰), suggesting the sample was either contaminated, or an error occurred during measurement. The second sample falling slightly out of the  $1\sigma$  range is a canine from grave 55, with a  $\delta^{18}$ O value of 16.6‰. This tooth, and the canines from the graves 74 and 73, might be reflecting breastfeeding oxygen isotope values, rather than drinking water values. As the canine forms on average between the age of 1.5 and 5 or 6 years (Reid and Dean, 2006) the consumption of <sup>18</sup>O enriched mother's milk during this developmental stage may influence the tooth's isotopic composition (Wright and Schwarcz, 1998). However, the  $\delta^{18}O$  value measured in the canine of grave 73 (15.3‰) falls close to the mean value, indicating no influence from mothers' milk. We can conclude then that the outliers in our small oxygen data set are more likely the cause of technical issues, than representing humans with slightly different drinking water sources. The approximate  $\delta^{18}O$  value of the drinking water can be assessed by calculating the fractionation between  $\delta^{18}O_{water}$  and  $\delta^{18}O_{phosphate}$ . We followed the calculation by Levinson et al. (1987) with a correction of the enamel phosphate value for the NBS 120c standard by -1.4, as outlined and recommended by Chenery et al. (2010). The corrected  $\delta^{18}$ O values for the humans

from the site of Singen result in a mean predicted  $\delta^{18}O_{\text{drinking water}}$  of -11.5  $\pm$  1.7‰, 1σ. This prediction matches with the modern water from Lake Constance itself. Water samples of the lake revealed  $\delta^{18}$ O values of -12.1‰ and -11.9‰ in spring, and -13.4‰ and -12.2‰ in fall, demonstrating the influence of annual temperature variation on water  $\delta^{18}O$  (Buhl et al., 1991). Also, the modern annual precipitation data for Germany shows remarkable similarity between the predicted  $\delta^{18}O_{\text{drinkino water}}$  for the site of Singen and the  $\delta^{18}O$  values in meteoric water in the southern part of Germany, ranging from -10.6 to -11.2‰ (Tütken et al., 2004). Archaeological human  $\delta^{18}$ O data with similar signatures are reported 35km south of Singen, in the Canton Zurich. With a mean of  $14.7 \pm 0.5\%$  (n=4), these human enamel samples are regarded as typical for the southern German and Swiss region (Tütken et al., 2008). More local archaeological  $\delta^{18}O$ data is available for the Neolithic pig enamel from Hilzingen (16.6‰ and 15.4‰) and Singen-Offwiese (14.2‰ and 13.6‰) (after Bentley and Knipper 2005, corrected after Iacumin et al. 1996). The range found in pig teeth is remarkably similar to the range we measured in the Bronze Age human teeth, including the previously discussed outliers (13.8‰ to 16.6‰). Bentley and Knipper (2005) reconstructed the local meteoric water of the Hegau using these samples and obtained mean annual meteoric water values (SMOW) of -12.2‰ for Singen-Offwiese and - 9.7‰ for Hilzingen.

These various data from archaeological and modern samples indicated that the  $\delta^{18}O$  values measured in the Singen population resembles the local drinking waters, including lakes, streams and local rainfall. We found no evidence for a suggested coastal influence that was inferred from the 'Atlantic' style of metal artefacts. Drinking water in southern Britain has an oxygen isotope value of -5 to -7‰, which resulted in enamel  $\delta^{18}$ O values of ~16-19‰ for prehistoric British populations (Chenery et al., 2010; Eckardt et al., 2009; Evans et al., 2006b). Though there is little data available for coastal France,  $\delta^{18}O$  values of drinking water from northern Bordeaux (~50km from the Atlantic coastline) of -8.1‰ resulted in a  $\delta^{18}O$  value of 18.3‰ for human enamel (Daux et al., 2008). Other clearly coastal data derives from two Neolithic sites from the Netherlands with  $\delta^{18}$ O values between ~17 and 18‰ for local individuals (Smits et al., 2010). A range of historic human tooth samples from southern Lorraine (western France) revealed  $δ<sup>18</sup>O$ values between ~16-18‰ (Daux et al., 2005). Currently, there is no archaeological or environmental  $\delta^{18}$ O data available for Hungary or the Carpathian Basin for a comparison with the Singen data. Nevertheless, Evans et al. (2006a), also searching for points of comparison with



present day Hungary, suggested the  $\delta^{18}$ O values of drinking water to be lower than -9.0‰. Therefore, we conclude that the measured oxygen isotope values of the humans from Singen, despite the limited dataset, support the strontium isotope evidence for the individuals being local to the Singen region.

### *6.5.7. Sulphur isotope ratios in human bone*

The humans bone collagen sulphur isotope values are very similar to each other, with a mean  $\delta^{34}$ S value of 1.2  $\pm$  0.9‰ (Fig. 6.7.) which is typical for a terrestrial diet in temperate Europe (Nehlich et al., 2010). The sulphur isotope data supports what we found in the oxygen isotopes; there is no evidence for a marine influence or slightly elevated  $\delta^{34}$ S ratios due to sea spray effects (Craig et al., 2006). The mean  $\delta^{34}$ S value of 1.2  $\pm$  0.9‰ shows a clear terrestrial signal (Krouse and Levinson, 1984) and suggests no measurable input of freshwater fish protein in human diets from Lake Constance, since the average modern  $\delta^{34}S$  value of the water sulphates from the Lake Constance is 7.6‰ (Hoeppner et al., 1981). The range of the  $\delta^{34}S$  values is clustered tightly (Nehlich et al., 2010; Vika, 2009) and suggests a very homogenous composition of the diet. These results are similar to the carbon and nitrogen isotope data of these individuals, which demonstrate that the individuals from Singen had terrestrial omnivorous diets (K. Kupke, personal information). Unfortunately, there is no human or animal  $\delta^{34}S$  data available from this region for comparison. The closest archaeological site with sulphur isotopic results is the Late Bronze Age necropolis of Neckarsulm (Nehlich and Wahl, 2011). The humans from Neckarsulm averaged at  $2.2 \pm 1.1\%$ , no immigrants or individuals with non-local sulphur isotopic compositions could be found. For rough estimations of the possible available local sulphur isotopic signature geochemical information might be helpful (Nehlich et al., submitted). Sulphur

isotope ratios of the south-eastern Black Forest range from -3.4‰ to +9.8‰ (Gehlen et al., 1962). Evaporites from the Northern Alps range in their sulphur isotope values from 11.3‰ to 32.0‰ (Niedermayr et al., 1989). These geochemical signatures are not particularly helpful in this case because they are too far away from the cemetery site. Nevertheless, the results of the archaeological tissues fall within the range of the modern, local, geochemical data and suggest a probable local origin from the area around the Hohentwiel and the western shores of Lake Constance.

#### **6.6. Conclusion**

Following these various lines of evidence, the isotope evidence has shown that the sampled individuals from Singen can be considered to be a local population. Even the single male buried with an Atlantic dagger in his grave (grave 67) had local  ${}^{87}Sr/{}^{86}Sr$  values of 0.70834 in both his enamel and dentine, implying that area of childhood and burial are likely the same. We also found no proof for exogamy, at least for the five sampled female individuals in this study. But if this group was largely local, how did they acquire foreign metal objects? We cannot rule out that exchange networks did require mobility by the Singen people. However, long distant traveling did not occur during childhood, and did not include long-term stays in coastal areas with significant inputs of marine diets. Were foreign metal objects traded into the Hegau by other related populations? If so, this likely required participation in exchanging networks which have been described for the Later Bronze Age in Europe (Wells, 2008). One could argue that if the Singen people can be considered as local elite, it is likely that goods were imported to Singen and exchanged for Singen copper. Evidence from copper mining sites in the Balkans indicates that there was a strong preference for exotic metal even close to the own mines (Chapman, 2008). However, the hypothesis considering the Singen community as some kind of social elite due to its control over the copper ores in the western Alps has been challenged. Kienlin and Stöllner (2009) argue that according to the archaeological and ethnological record, there is no need for social stratification in the development of mining and metallurgy. Additionally, the cemetery of Singen itself does not show strong indications for social ranking. Hence, we would rather expect to deal with a small-scale community. Also, we see no variation in  $\delta^{34}S$  that could indicate freshwater fish consumption (Nehlich et al., 2010), known to indicate social

stratification in other historic periods (Richards et al., 1998), although Lake Constance would have been suitable for local fishing.

By applying complementary isotope systems we found evidence that the Singen people we studied were all of local origin, and there was no evidence for migrants from the Atlantic regions of France or Great Britain. However, we cannot exclude the possibility that the Singen people were connected with the region between Lake Constance and Lake Zurich, as the region has very similar strontium and oxygen isotope values. The closest regional copper ores are located approximately 80km south-west from this region in the mountainous area of Grisony (*Graubünden*) and the Montafon valley, which could have been utilized for copper mining industries (Krause, 2009). Also, a connection to the Carpathian Basin cannot be ruled out due to the lack of isotopic background information from this region. Finally, the main limitation of this study derives from the fact that only 25 out of 96 graves were sampled for mobility patterns. We cannot completely exclude the possibility that some individuals, e.g. the three non-sampled males with 'Atlantic' daggers, were of foreign provenance. Due to the insufficient skeletal preservation, this question remains unresolved.

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