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INTRAINDIVIDUAL VARIABILITY OF STRONTIUM AND LEAD STABLE ISOTOPES AND ELEMENT CONCENTRATIONS IN ARCHAEOLOGICAL SKELETONS FROM ROMAN STETTFELD (CA 150–300 CE), BADEN-WÜRTTEMBERG (GERMANY)

ABSTRACT: Strontium and lead stable isotopes, and element concentrations were measured in 12 uncremated skeletons from the Roman Stettfeld site (ca 150–300 CE; Baden-Württemberg, FRG). Samples were taken from three to five skeletal parts per individual that precipitated and/or accumulated trace elements at different ontogenetic stages, namely enamel of different permanent teeth, compact and trabecular bone, and new bone formations such as active periostitis or fracture callus. Six out of the 12 skeletons turned out to be immigrants to the site according to enamel and/or bone $^{87}Sr/^{86}Sr$ isotopic ratios. By use of enamel precipitation data, bone remodeling and strontium clearance rates from the skeleton, individual age at migration could be refined and showed that residence change took place during infancy or juvenile ages. One female skeleton exhibited negative turnover rates indicating a negative calcium balance. Together with a conspicuous sulcus praeauricularis and her young age-at-death, this is most plausibly interpreted as death shortly after pregnancy and birth. While the residence change of this female could have been due to exogamy, migration during childhood or juvenile age indicates movement of (family) groups of people. In Roman times, also slavery cannot be excluded. With regard to the geological variability at the site, catchment area of the immigrants should however have been small and was most probably restricted to the Black Forest and nearest surroundings. Rich ore deposits of the region, and the dependency of the Roman society from silver and lead, constituted a strong pull-factor that time. All but two individuals that had been identified as immigrants by $^{87}Sr/^{86}Sr$ exhibited skeletal lead stable isotopic ratios that are compatible with this region.

KEY WORDS: Strontium – Lead – Isotopes – Concentrations – Ontogeny – Mobility

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INTRODUCTION

Since many years, detecting and provenancing non-local individuals on a burial site by $^{87}Sr/^{86}Sr$ isotopic ratios in bioapatite has become routine for the solution of open questions concerning mobility and population dynamics in prehistory. Besides case studies, research efforts concentrate on the definition of local bioavailable $^{87}Sr/^{86}Sr$ ratios, and the definition of the most probable place of origin of the immigrants to a site including the assessment of appropriate cut-off values between local and non-local isotopic signatures (e.g. Frei, Frei 2011, Willmes *et al.* 2014, Crowley *et al.* 2017, Grimstead *et al.* 2017, Lengfelder *et al.* 2019). Most researchers prefer analyzing dental enamel over bone because the former is less susceptible towards diagenesis, although not inert (Sebald *et al.* 2018a, de Winter *et al.* 2019). In case dental enamel that has precipitated during childhood or juvenile age exhibits an isotopic signature that is different from the local one, the individual in question must have spent its childhood/adolescence somewhere else. However, almost nothing can be told about the individual age at migration. Some studies use the intraindividual difference between $^{87}Sr/^{86}Sr$ in dental enamel (or, in case of cremations: dentine) and compact bone to distinguish between early and late immigrants to a site (e.g. Alt *et al.* 2014, Sebald *et al.* 2018b). Dependent from both the individual age of residence change and age at death, the bone isotopic ratios will exhibit the local, a non-local, or a mixed isotopic signature according to the stage of adjustment to the local values in the course of tissue remodeling. Alternatively, enamel samples from teeth that precipitate at different ontogenetic ages are compared to get clues to possible residence changes during adolescence (e.g. Schweissing, Grupe 2003). However, it becomes more and more apparent that different $^{87}Sr/^{86}Sr$ isotopic ratios in different teeth of the same dentition can be the result of dietary change (Anders *et al.* 2019), a change of residence, or both.

Place of origin of immigrated individuals mostly cannot be defined by the geodependent $^{87}Sr/^{86}Sr$ isotopic signatures alone, because similar or even the same such ratios occur in many different places worldwide. Therefore, $^{87}Sr/^{86}Sr$ is frequently augmented by $\delta^{18}O_{phosphate}$ for a better estimation of the most probable place of origin. $\delta^{18}O$ is related to hydrological cycles and therefore not geodependent such as $^{87}Sr/^{86}Sr$, but rather a function of geographic and ecological parameters (Rozanski *et al.* 1993, Horita, Weselowski 1994, Gat 1996). Therefore, $\delta^{18}O_{phosphate}$ in bones and teeth is largely a function of the respective ratio in drinking water. While the isotopic signature of childhood is purposefully targeted by $^{87}Sr/^{86}Sr$ in dental enamel, this information can be significantly blurred in the case of $\delta^{18}O$ because of e.g. nursing effects. Moreover, Lightfoot and O'Connell (2016) have emphasized the very high interindividual variability of $\delta^{18}O$ even in local human populations. Therefore, the additional spatial information of this isotopic ratio is limited.

More recently, studies making use of lead stable isotopes for georeferencing human skeletons have grown in number (e.g. Font *et al.* 2015, Shaw *et al.* 2016, Price *et al.* 2017, Evans *et al.* 2018). What holds for strontium isotopes also holds for lead stable isotopes, including efforts aiming at an isotopic mapping (e.g. Durali-Müller *et al.* 2007, Reimann *et al.* 2012, Albarède *et al.* 2012). Just as strontium, lead substitutes for calcium lattice positions in the bioapatite. While strontium is mainly incorporated into the body with food and drinking water, contact and incorporation of lead into the body has changed over time. Today, lead is mostly taken up by inhalation of aerosols and dust particles (Kamenov, Gulson 2014), but before relevant measures for environment and health protection became mandatory, contaminated food and water were also an important lead source. This holds especially for Roman times, when a lead industry was already fully developed (Nriagu 1983, Lessler 1988, Hernberg 2000). Lead was mostly a byproduct of silver mining, but lead ores as such were also exploited. People had therefore contact with artifacts for daily use made of or containing substantial amounts of lead, such as cooking vessels, water pipes, tools, pigments, childrens' toys, and ornaments. The Romans were also famous for adding lead acetate as sweetener to beverages (Alfödi-Rosenbaum 1984, Lessler 1988). Georeferencing archaeological skeletons from Roman times with lead will therefore face the difficulty of mixed lead isotopic ratios in a consumer because of smelting and reworking artifacts made up of lead from different ores, and the unknown origin of lead that was added to food and drinks. Therefore, consumer lead stable isotopic ratios are not exclusively related to the place of residence, but also to the overall economy.

This study is dedicated to a systematic analysis of both strontium and lead stable isotopic ratios in the bioapatite of archaeological skeletons, whereby samples were taken from each individual that represent different ontogenetic stages (enamel of different teeth, different bone samples). Not only isotopic ratios, but also the strontium and lead concentrations in the samples were measured. Taking both the age of enamel precipitation and the remodeling rate of bone into account, we tried to better differentiate primarily local from non-local individuals, and to estimate the individual age of residence change of the latter. We did not consider $\delta^{18}O$ for the reasons mentioned above.

MATERIAL

12 individual human skeletons from the Roman burial site at Stettfeld, Rosenberg field (Baden-Württemberg, FRG, ca 150–300 CE) were investigated (*Figure 1*). The Stettfeld site is located at the northern edge of the Black Forest in Baden-Württemberg, about 25 km north of the city of Karlsruhe, and less than 20 km from the eastern edge of the upper Rhine valley. The site has been excavated during several campaigns between 1978 and 1981. Due to modern overbuilding, it could not be excavated completely. About 370 individual skeletons were recovered, whereby about 90% of the skeletons were cremated and the remaining 10% uncremated. According to the archaeological and anthropological context, both the inhumations and cremations were performed by the same population (Alfödy-Thomas 1988, Wahl 1988, 1997). South-west Germany, and especially the Black Forest area is famous for the numerous ore deposits and its long history of lead and silver mining (Hildebrandt 2004). With regard to the dependency of the Roman society from both metals, the area should have constituted a rather strong "pull factor" for the people that time.

20 enamel and 22 bone samples were taken from 12 uncremated adult skeletons (*Table 1*). Skeletons were chosen according to the availability of skeletal samples that permit the monitoring of element uptake during different ontogenetic stages (according to AlQuahtani *et al.* 2010, Tsutaya, Yoneda 2013, Shagina *et al.* 2015):

Stage I: Enamel from a 1st permanent molar (first ca 3.5 years of life).

Stage II: Alternatively, enamel from a permanent incisor (first ca 4 years of life). The formation time of the crown of permanent incisors differs only little from those of first permanent molars, therefore, incisors were sampled from three individuals where no first permanent molar was available (grave nos 263, 300, 348B; see *table 1*).

Stage III: Enamel from a 3rd permament molar (about 7–13 years in the upper jaw, from about 8–14 years in the mandible).

Stage IV: Compact bone from a long limb bone (element uptake of up to 30 years prior to death).

Stage V: Cortical bone from a rib (element uptake of up to 15 years prior to death).

Stage VI: Recently formed bone such as active periostitis, osteophytes, or fracture callus.

METHODS

Stable isotope analysis

Laboratory processing of tooth and enamel samples, and mass spectrometry was performed according to Toncala *et al.* (2017a). Bone surfaces were manually removed by grinding, tooth enamel was manually separated from the dentin. Bone and enamel samples were washed ultrasonically (35 kHz) in deionized H_2O until the water remained clear, and air-dried. Bone samples were then defatted by a Soxhlet extraction for at least 5 hours with diethylether to remove humic substances that are known for cation capture. Next, each sample was devided into two aliquots, and one part was etched ultrasonically (35 kHz) for ≥ 5 minutes in concentrated HCOOH (98%, suprapure quality) for the measurement of $^{87}Sr/^{86}Sr$, the other for ≥ 10 minutes in concentrated HCl (37%, suprapure quality) for the measurement of Pb isotopes. This acid leaching is terminated when leachate and remaining sample have the same isotopic ratio within the measurement error. Next, the samples were washed ultrasonically in deionized water until a pH between 5 and 6 was reached, and air-dried. Ashing was performed in a muffle furnace at 800 °C for 12 hours. After cooling to room temperature, the samples were homogenized with a ZrO_2 ball mill to a powder as fine as possible to avoid phase heterogeneity.

For the measurement of $^{87}Sr/^{86}Sr$, a maximum of 50 mg homogenized sample was wet ashed in 1 mL concentrated HNO_3 (suprapure) in closed teflon cubes on a hot plate at 100 °C for 24 hours. Remaining liquid was evaporated at the same temperature overnight. After cooling to room temperature, 1 mL 8N HNO_3 (ultrapure) was added and the sample kept on a hot plate at 100 °C for 20 minutes. Column separation was performed with Sr SPEC resin. For the measurement of Pb stable isotopes, a maximum of 200 mg of the homogenized samples was solubilised in 1mL concentrated HNO_3 (suprapure) in closed teflon cubes.

Solubilized samples were evaporated for 2 hours under red light and ashed for 4 hours at 800 °C. Samples were then soaked with concentrated HBr and evaporated for 30 minutes under red light. The ash was solubilised ultrasonically in 0.45 mL 0.5N HBr for 1 minute and centrifuged at 13.400 rpm for 3 minutes. Column separation was performed by use of Dowex 1×8-anion exchange resin in two steps (column cleaning and conditioning, followed by a second cleaning and conditioning (= "clean-up")). Finally, 1 µL $HClO_4$ was added, and the sample evaporated under red light. Blanks contained << 1 ng Pb.

$^{87}Sr/^{86}Sr$ was analysed with a TIMS Finnigan MAT 261.5 on single tungsten filaments. Standard reference material SRM 987 was used for quality control ($^{87}Sr/^{86}Sr$, n = 4: 0.710206 ± 0.00029; certified value 0.71034 ± 0.00026). Measurement error was ± 0.00001. Mass spectrometry of stable lead isotopes was performed with the same mass spectrometer, standard reference material SRM 982 served for quality control ($^{206}Pb/^{204}Pb$, n=5: 36.7294 ± 0.0129; certified value 36.7390 ± 0.0364; $^{206}Pb/^{207}Pb$, n=5: 2.14118 ± 0.00045; certified values 2.14101 ± 0.000929) (Toncala *et al.* 2017b).

Strontium and lead concentrations

Trace element concentrations were measured after microwave digestion of about 10 mg apatite powder in 1 mL HNO_3 by the DIN 38406 E6 method in an electrothermal atomic absorption spectrometer (GF-AAS). The selected analytical wavelenghts were 460.7 nm for Sr and 283.3 nm for Pb. Standard reference material SRM 1400 (bone ash) was used for quality control (Sr: 259 ±22.7 ppm, certified value 249 ± 7 ppm; Pb: 9.87 ± 1.42 ppm, certified value 9.07 ± 0.12 ppm).

Calculations

The probable range of local bioavailable $^{87}Sr/^{86}Sr$ isotopic ratios at the site was estimated by use of the Isoplot 3.70 software (Ludwig 2008) and comparison with published values from the region (Sjøgren *et al.* 2016). While tooth enamel is not remodeled after precipitation, the remodeling rate of bone is particularly slow and the biological half-life of incorporated trace elements very long accordingly. On average, only 3–5% of compact bone is remodeled per year (Martin *et al.* 1998) with the result that the biological half life of strontium in bone exceeds some years. Published values vary from 1100 days (Schenker *et al.* 1999) over 2000 days (Vaughan 1975) up to 2500 days (Papworth, Vennart 1973) in the adult. Therefore, trace element concentrations in compact bone accumulate over an individual's adult life time.

Because of the health hazard induced by the incorporation of the radioactive ^{90}Sr isotope, existing studies on the ^{90}Sr clearance rate from bone are also useful for an estimation of individual remodeling rates and strontium accumulation based on strontium concentrations in the skeleton. This clearance rate is both age- and sex-specific (Tolstykh *et al.* 1997). In this study, the data published by the International Commission on Radiological Protection (1973) and Shagina *et al.* (2015) were used, and also the respective estimations for collagen turnover (the latter is however faster than the mineral turnover) by Hedges *et al.* (2007).

In case of incorporation of Sr with a different $^{87}Sr/^{86}Sr$ ratio following a residence change, the individual age of migration of individuals with a non-local strontium isotopic signature can thus be estimated. Individuals that exhibit a non-local $^{87}Sr/^{86}Sr$ isotopic signature in enamel that precipitates during childhood must have been primarily non-local to the site of their recovery. Under the basic assumption that every year that such an individual had spent at the latter place, an age-related proportion of strontium with the new isotopic signature is incorporated into the bones, individual migration age was assessed by starting from its isotopic signature at death and a respective "backwards" calculation. For example, a collagen remodeling rate of about 6% shall be assumed (Hedges *et al.* 2007). In case of residence change, a maximum of about 6% of the incorporated Sr with the local signature will mix with 94% Sr with the isotopic signature of the place of origin every year, and so forth. Since the individual skeletal samples cover different ontogentic stages from early childhood to adulthood, the sex-specific average turnover rates of children (30% up to an age of 10 years in males, 20% in females; 13.7% and 6% between 10 and 19 years in males and females respectively; 3% and 4% between 20 and 29 years in males and females respectively; and 3% in adults of both sexes from 30 years onwards) were used according to Hedges *et al.* (2007). We are aware of the uncertainties that are especially due to the imprecise morphological age-at-death diagnosis, and other unknown parameters such as diet, health status, etc. However, a gross differentiation between migration during childhood/juvenile age and adulthood is possible. This information is crucial for assessing the most probable reasons for a detected individual residence change. Also, since collagen remodels a little faster than mineral, the earliest plausible age-range at migration is estimated this way.

Also, a "strontium accumulation rate" that is related to the bone formation rate was estimated from the measured strontium concentrations. For the subadult stage in an individuals' life, the percentage difference in the strontium concentrations between two enamel samples from the same individual was distributed over the approximate age difference between the enamel precipitation times. Taking burial no 114 as an example (*Table 1*), the difference in Sr concentrations between tooth 36 (enamel precipitates between ca 0–3.5 years) and tooth 38 (precipitation between ca 8–14 years) is 1.1% on average; with a minimum (0–14 years) of 0.5% and a maximum (3.5–8 years) of 1.6%. For the accumulation rate during adulthood, the percentage difference in Sr concentrations between the youngest tooth enamel and bone was calculated, what equals 14 years until age-at-death when a third molar was available. Only first permanent molars could be sampled from the skeletons of burials 26 and 240, therefore, the age range needed to be extended to 3.5 years until age-at-death what elevates the formation rates accordingly. Long-term skeletal distribution and retention of lead in mineralized tissues are similar to those of the alkaline earth elements (Leggett 1993).

RESULTS

Stable strontium and lead isotopic ratios and element concentrations per individual and sample are listed in *Table 1*. Estimated Sr accumulation rates as a measure for individual bone turnover rates for children and adults are listed in *Tables 2* and *3*. Percentage accumulation during childhood averages 5.74% Sr [ppm] per year with a range from 1.05–9.55%. In adults, average percentage accumulation per year is 5.9% with a variability form 1.28 to 14.21% Sr [ppm] per year. Finally, the changing accumulation rate in % per year during adulthood was assessed per decade and compared with published such values for humans (*Table 4*; burial 300 excluded, see discussion).

The Isoplot 3.7 software (σ=1.82) identified a clear peak in the distribution of $^{87}Sr/^{86}Sr$ isotopic ratios of all samples between 0.70901 and 0.70975 with a mean of 0.70938 ± 0.0002 (1 sd). (*Figure 2*). According to Burton & Price (2013), a variability of ± 0.00015 is typical for local individuals, therefore, we set $^{87}Sr/^{86}Sr$ = 0.70938 as the local value for the site. This value was entered into the calculations for the most plausible individual age range at migration. According to this and the variation of isotopic ratios by ontogenetic age visualized by *Figures 3a, b*, the individuals from burials 26, 114, 225, 300, 348A and 348B were primarily not local to Stettfeld. In the old adult male from burial no 225, $^{87}Sr/^{86}Sr$ isotopic ratios of the mineralized tissues changed twice during his life (*Figure 3b*).

This graphical illustration of the intraindividual change of stable strontium isotopic signatures not only

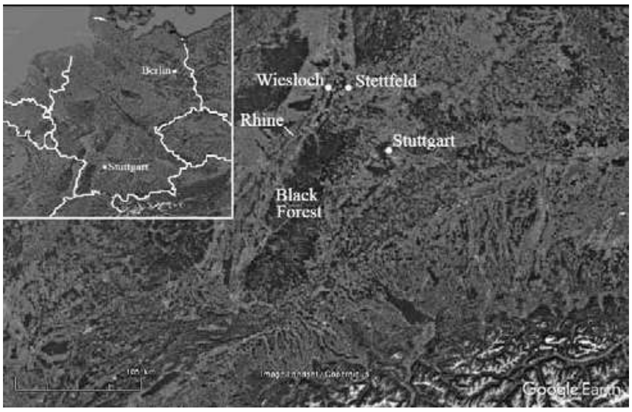

FIGURE 1: Location of the Stettfeld site and the Black Forest in Baden-Württemberg, FRG. Map Germany: 50°55'49.97" N, 10°23'20.66" E, Google Earth, 2018, date of access: January 15, 2019; map Black Forest: 48°29'56.51" N, 9°09'40.14" E, Google Earth, 2018, date of access: January 15, 2019.

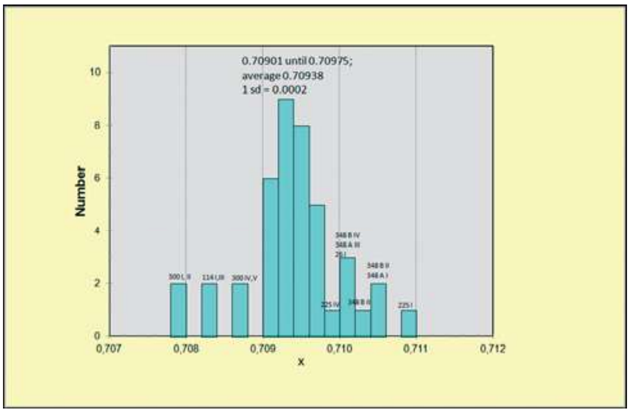

FIGURE 2: Probability plot of the distribution of $^{87}Sr/^{86}Sr$ isotopic ratios (x) over all samples, generated by Isoplot. Burial numbers and ontogenetic stage (Roman numbers) are indicated.

TABLE 1: Investigated individuals, bone/tooth samples, radiocarbon dates, and analytical results. SE [%] = double standard error in percent. Age-at-death and sex determination by Wahl (1988). Radiocarbon dates (± 1) according to Knötzele 2018.

burial	*age at death (years)*	*sex*	*Bone/ enamel sample*	*Sr µg/g*	*Pb µg/g*	*$^{87}Sr/^{86}Sr$*	*2SE [%]*	*$^{208}Pb/^{204}Pb$*	*2SE [%]*	*$^{207}Pb/^{204}Pb$*	*2SE [%]*	*$^{206}Pb/^{204}Pb$*	*2SE [%]*	*$^{208}Pb/^{207}Pb$*	*2SE [%]*	*$^{206}Pb/^{207}Pb$*	*2SE [%]*
19	60–70	male	**left femur**	343	17.8	0.70929	0.0027	38.4091	0.2002	15.6276	0.1503	18.4163	0.1005	2.4578	0.0500	1.1809	0.0500
			16	196	10.9	0.70946	0.0023	38.3758	0.2009	15.6222	0.1506	18.3567	0.1006	2.4565	0.0503	1.1750	0.0501
			48	174	8.72	0.70961	0.0019	38.4044	0.2001	15.6275	0.1501	18.3862	0.1002	2.4575	0.0500	1.1765	0.0500
26 (135-214 CE)	55–60	probably male	**left femur**	386	2.89	0.70921	0.004	38.3760	0.2002	15.6102	0.1502	18.4755	0.1002	2.4584	0.0501	1.1835	0.0500
			left fibula hyperosto sis	368	4.9	0.70936	0.0022	38.5648	0.2003	15.6304	0.1503	18.6755	0.1003	2.4673	0.0500	1.1948	0.0500
			46	94,4	1.91	0.71011	0.0022	38.2431	0.2007	15.6168	0.1505	18.3102	0.1006	2.4489	0.0502	1.1725	0.0500
114	40–50	male	**left tibia**	408	18.2	0.70957	0.002	38.4702	0.2008	15.6361	0.1508	18.4680	0.1011	2.4603	0.0501	1.1811	0.0501
			left fibula callus	395	20.5	0.70957	0.0022	38.4157	0.2001	15.6190	0.1501	18.4502	0.1002	2.4596	0.0500	1.1813	0.0500
			36	224	4.05	0.70834	0.0043	38.3557	0.2003	15.6126	0.1503	18.4136	0.1005	2.4567	0.0501	1.1794	0.0500
			38	240	0.4	0.70839	0.0024	38.4523	0.2024	15.6424	0.1527	18.4871	0.1036	2.4582	0.0504	1.1843	0.0503
225	50–60	probably male	**right humerus**	378	2	0.70995	0.0025	38.3727	0.2001	15.6110	0.1502	18.5239	0.1002	2.4581	0.0500	1.1866	0.0500
			right rib	304	3.13	0.70966	0.0025	38.4686	0.2005	15.6301	0.1503	18.5225	0.1003	2.4612	0.0502	1.1851	0.0501
			26	136	1.21	0.71091	0.0024	38.4715	0.2011	15.6369	0.1511	18.4682	0.1016	2.4601	0.0502	1.1810	0.0501
			28	123	0.7	0.70945	0.0036	38.3512	0.2003	15.6189	0.1503	18.4810	0.1004	2.4554	0.0501	1.1833	0.0500
240	50–60	male	**ribght tibia periostitis**	287	6.49	0.70944	0.0017	38.6129	0.2001	15.6436	0.1501	18.6708	0.1001	2.4683	0.0500	1.1935	0.0500
			right femur	362	7.56	0.70925	0.0017	38.5071	0.2002	15.6376	0.1502	18.4627	0.1003	2.4625	0.0501	1.1806	0.0500
			46	95	1.22	0.70939	0.0024	38.4805	0.2010	15.6332	0.1510	18.5646	0.1014	2.4615	0.0501	1.1875	0.0501
263	25–30	female	**left tibia**	699	4.04	0.70901	0.0022	38.4077	0.2006	15.6160	0.1504	18.4973	0.1005	2.4594	0.0503	1.1845	0.0500
			11	193	3.99	0.70953	0.0017	38.5055	0.2019	15.6372	0.1519	18.5930	0.1024	2.4623	0.0504	1.1890	0.0502
			38	228	2.03	0.7094	0.0019	38.4938	0.2005	15.6446	0.1505	18.5170	0.1006	2.4605	0.0501	1.1836	0.0501
264	30–35	female	**right femur**	457	3.41	0.70919	0.0035	38.3976	0.2002	15.6239	0.1503	18.4463	0.1004	2.4577	0.0501	1.1806	0.0500
			right rib	440	3.96	0.70913	0.0028	38.4560	0.2004	15.6245	0.1503	18.5243	0.1004	2.4613	0.0502	1.1856	0.0500
			48	307	0.7	0.70961	0.0023	38.5229	0.2007	15.6418	0.1508	18.5546	0.1012	2.4628	0.0501	1.1887	0.0501
			36	186	1.73	0.70943	0.0025	38.5438	0.2012	15.6460	0.1509	18.5976	0.1012	2.4635	0.0504	1.1911	0.0501

TABLE 1: Continued.

burial	*age at death (years)*	*sex*	*Bone/ enamel sample*	*Sr µg/g*	*Pb µg/g*	$^{87}Sr/^{86}Sr$	*2SE [%]*	$^{208}Pb/^{204}Pb$	*2SE [%]*	$^{207}Pb/^{204}Pb$	*2SE [%]*	$^{206}Pb/^{204}Pb$	*2SE [%]*	$^{208}Pb/^{207}Pb$	*2SE [%]*	$^{206}Pb/^{207}Pb$	*2SE [%]*
300 (225-322 CE)	25–30	female	**right femur**	595	6.1	0.70865	0.0023	38.6807	0.2005	15.6551	0.1503	18.6478	0.1003	2.4708	0.0502	1.1912	0.0501
			left rib	375	7.58	0.70875	0.003	38.7052	0.2005	15.6587	0.1504	18.6786	0.1005	2.4718	0.0501	1.1929	0.0500
			28	674	0.3	0.70782	0.002										
			32	602	3.44	0.70792	0.0023	38.8033	0.2015	15.6713	0.1509	18.7097	0.1009	2.4761	0.0509	1.1964	0.0501
309 (139-235 CE)	30–35	male	**left femur**	n.a.	n.a.	0.70927	0.0037	38.3686	0.2023	15.6157	0.1523	18.4765	0.1032	2.4571	0.0504	1.1831	0.0502
			right rib	373	3.84	0.70913	0.0024	38.6053	0.2024	15.6528	0.1518	18.6525	0.1012	2.4664	0.0506	1.1916	0.0507
			18	112	0.5	0.70972	0.0019	38.6933	0.2025	15.6903	0.1528	18.6465	0.1037	2.4661	0.0504	1.1909	0.0503
382	60–65	male	**right femur**	518	3.1	0.70906	0.0021	38.4475	0.2005	15.6213	0.1504	18.6869	0.1005	2.4613	0.0501	1.1962	0.0501
			right rib	547	2.78	0.70909	0.0024	38.5268	0.2004	15.6338	0.1503	18.6254	0.1004	2.4643	0.0502	1.1913	0.0501
			28	115	0.3	0.70975	0.0023	38.4891	0.2028	15.6403	0.1533	18.5221	0.1044	2.4609	0.0505	1.1867	0.0502
348 A	25–30	male	**right humerus**	552	4.11	0.70931	0.0022	38.4328	0.2026	15.6373	0.1517	18.4297	0.1015	2.4579	0.0510	1.1786	0.0504
			left rib	593	10.7	0.70923	0.0023	38.6686	0.2002	15.6623	0.1501	18.6080	0.1002	2.4688	0.0502	1.1881	0.0500
			26	183	0.8	0.7105	0.0028	38.5267	0.2005	15.6418	0.1505	18.5360	0.1007	2.4631	0.0502	1.1875	0.0500
			28	183	0.7	0.7101	0.003	38.5091	0.2006	15.6359	0.1504	18.5292	0.1006	2.4629	0.0506	1.1875	0.0501
348 B	40–60	female	**left femur**	357	5.47	0.70947	0.0018	38.2968	0.2010	15.6050	0.1509	18.4116	0.1011	2.4541	0.0503	1.1798	0.0500
			left rib periostitis	221	7.16	0.71004	0.0024	38.7422	0.2003	15.6582	0.1502	18.8302	0.1002	2.4742	0.0502	1.2026	0.0500
			18	229	0.3	0.71026	0.0017	38.5651	0.2017	15.6606	0.1514	18.6999	0.1019	2.4626	0.0507	1.1965	0.0502
			32	197	0.4	0.71043	0.002	38.4397	0.2022	15.6365	0.1521	18.5008	0.1030	2.4583	0.0510	1.1856	0.0506

TABLE 2: Estimated Sr accumulation rates (% per year) during childhood.

	Sr accumulation rate during childhood		
	% Sr µg/g per year		
burial	maximum	minimum	average
19	-0.80	-2.49	-1.65
114	1.59	0.51	1.05
225	-0.74	-2.73	-1.73
263	4.53	1.30	2.91
264	14.46	4.65	9.55
300	11.96	0.92	6.44
348 B	16.24	1.25	8.75

differentiates non-local from local individuals but also shows that not only enamel samples, but also some bone samples plot outside the estimated local range. While three out of four newly formed bone samples exhibit local $^{87}Sr/^{86}Sr$ ratios, the fourth one does not. Since the estimated bone turnover and Sr acculumulation rates are compatible with modern such values (see discussion), and an average local $^{87}Sr/^{86}Sr$ isotopic ratio was evaluated for the Stettfeld site, age at migration of the non-local individuals could be estimated as well. The resulting ages and age-ranges are: ca 13 years (male 26), ca 10–11 years (male 114), ca 4–8 years and 13–22 years (male 225), ca 12–19 years (female 300), ca 6 years (male 348A), and 6–25 years (female 348B).

With regard to the lead stable isotopes, again not only enamel but also bone isotopic ratios exhibit a considerable inter- and intraindividual variability. Ströbele *et al.* (2012, 2015) have published a compilation of measurements of lead stable isotopes of mineralizations in the Black Forest that are distinguished by their age, the source rocks, and tectonic settings. The ore deposit closest to the Stettfeld site at a distance of about 20 km is Wiesloch (*Figure 1*), a Mississippi-Valley type deposit (Ströbele *et al.* 2015). As shown by *Figure 4*, lead stable isotopic ratios of nearly all archaeological samples are consistent with the local geological signatures, whereby most data cluster in the Wiesloch and the "south-west German Triassic" group, galena samples that could have either been mined in close proximity to the

TABLE 3: Estimated Sr accumulation rates (% per year) during adulthood.

		Sr accumulation during adulthood ***% Sr µg/g per year***		
burial no	bone	maximum	minimum	average
19	femur	2.11	1.57	1.84
26	femur	5.99	5.15	5.57
	newly formed	5.63	4.83	5.23
114	tibia	2.69	1.67	2.18
	newly formed	2.48	1.54	2.01
225	rib	3.98	2.78	3.38
	humerus	5.60	3.91	4.76
240	femur	6.04	4.68	5.36
	newly formed	4.35	3.37	3.86
263	tibia	18.78	9.34	14.08
264	femur	3.05	1.81	2.43
	rib	2.71	1.60	2.16
300	femur	-0.98	-0.51	-0.74
	rib	-3.70	-1.93	-2.81
309	rib	13.71	8.32	11.02
382	femur	7.46	6.04	6.75
	rib	7.99	6.48	7.23
348 A	rib	18.67	9.74	14.21
	humerus	16.80	8.77	12.79
348 B	femur	1.51	1.05	1.28
	newly formed	-0.13	-0.07	-0.10

TABLE 4: Bone formation rate per decade in adults (% per year) (except burial 300).

age group	all bones	ribs	Int. Comm. on Radiol. Prot. (1973)
20–30 years	13.8	14.2	3–10.4
30–40 years	5.2	6.6	1.2–2.4
40–50 years	2.1	–	2.4–5.0
50–60 years	4.7	3.4	2.4–4.8
60–70 years	5.3	7.2	2.7–5.3

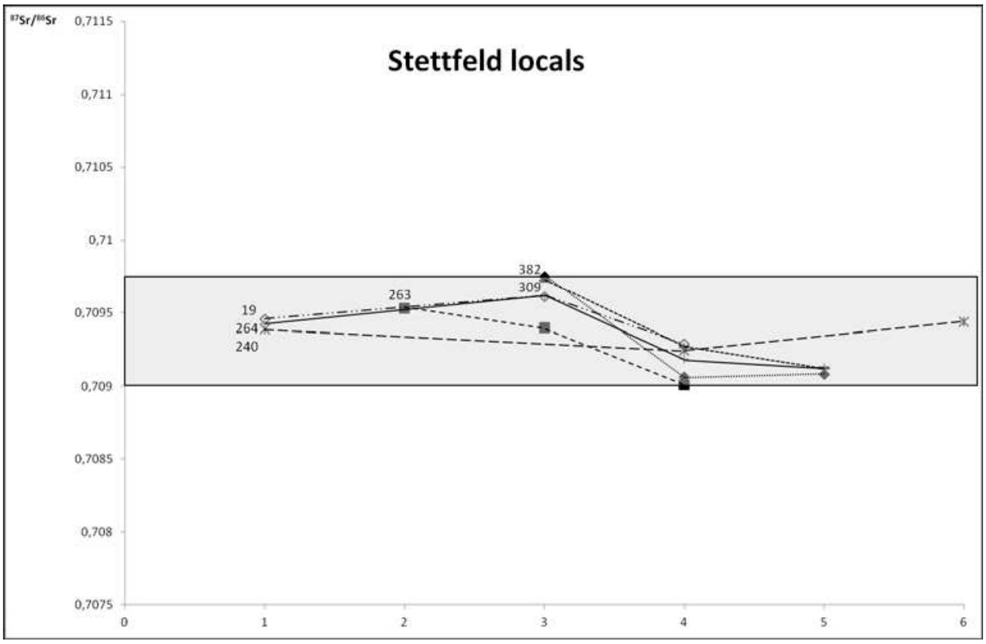

FIGURE 3a: Intraindividual change in $^{87}Sr/^{86}Sr$ isotopic ratios of local individuals in comparison with the most probable local range according to the Isoplot calculation. 1–6: developmental stages.

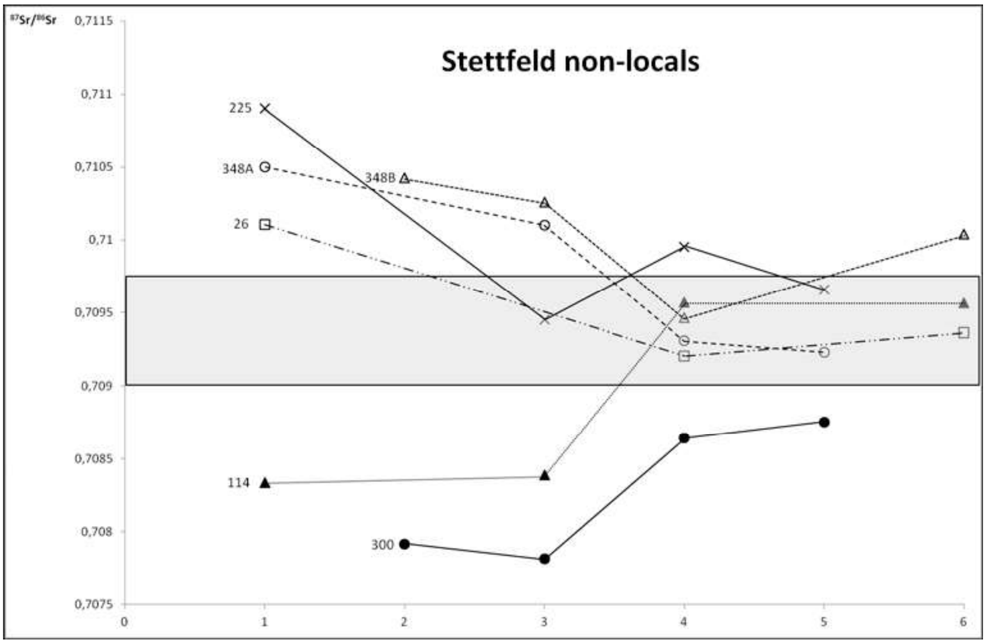

FIGURE 3b: Intraindividual change in $^{87}Sr/^{86}Sr$ isotopic ratios of non-local individuals in comparison with the most probable local range according to the Isoplot calculation. 1–6: developmental stages.

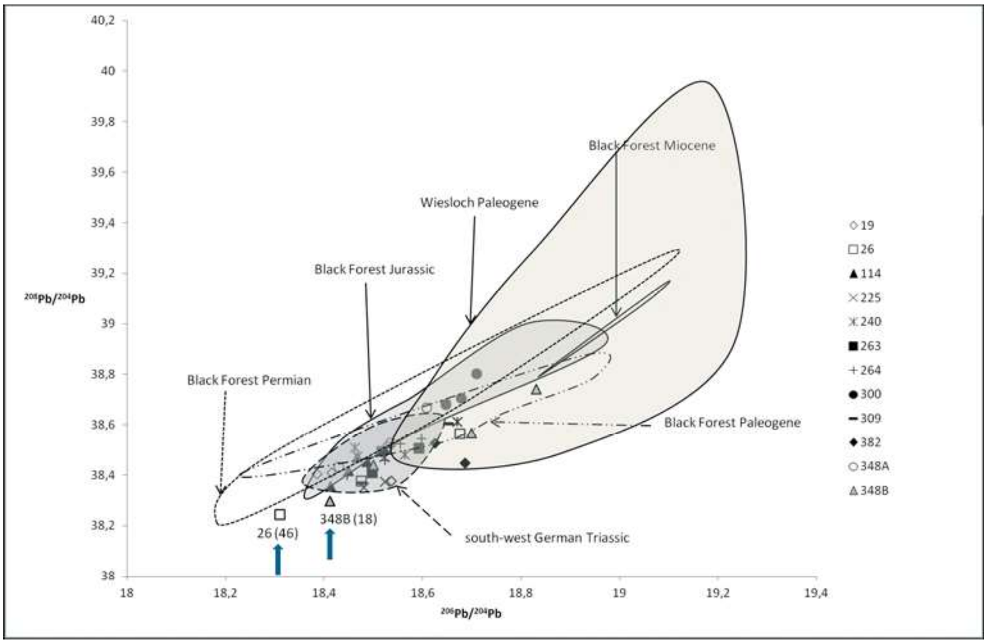

FIGURE 4: Comparison of lead stable isotopes in archaeological samples with geological such values in the Black Forest and Wiesloch region (Ströbele *et al.* 2012).

Stettfeld site, or up to 100 km further to the south (Ströbele *et al.* 2012). It is therefore hardly possible to match any skeletal lead stable isotopic ratio with a particular mining site, let alone that the Roman lead industry will have led to anthropogenic isotopic mixtures. The only two exceptions are the non-local individuals 26 (tooth 46), and 348B (tooth 18), suggesting that they had spent their childhood/juvenile age elsewhere. This is compatible with their non-local strontium isotopic signatures.

DISCUSSION

By relating $^{87}Sr/^{86}Sr$ isotopic ratios and strontium concentrations in mineralized tissues that represent different ontogenetic stages of the individual tested, we tried to get clues to the most probable age of residence change. The investigation was augmented by the measurement of lead stable isotopic ratios and lead concentrations of the samples. Analyses of Roman artifacts revealed that in the southwest of Germany, the majority of Pb originated from local ore deposits up to the 2nd century CE. It was not before the following 3rd and 4th centuries CE that supraregional Pb import gained in importance (Durali-Mueller *et al.* 2007).

Strontium concentrations and the calculated accumulation rates during childhood and adult age are variable but meet expectations (e.g. Specht *et al.* 2017). Strontium is incorporated via food and drinking water. Since strontium is always associated with calcium in nature, different dietary preferences are most probably responsible for the observed variation (Specht *et al.* 2017). Accumulation rates exceeding 10% were observed in only three individuals, namely in the female no 263, and the two males with the grave numbers 309 and 348A. A few samples with negative such accumulation rates were excluded from further interpretation, because negative accumulation rates imply calcium loss from the skeleton. This concerns two individuals (burials no 19 and 225) during their childhood, the young adult female with the burial no 300 during her adult life, and the newly formed bone (a sample from an active periostitis) from the old adult female number 348B (*Tables 2 and 3*). Negative strontium accumulation rates during childhood are best explained by survived childhood stress and disease. Diagenesis can largely be ruled out (see below) with the probable exception of the very small sample taken from the active periostitis that was characterized by immature bone with a high porosity by nature. We also exclude an artifact caused by e.g. leaching strontium from the sample in the course of sample processing, not only because formic acid is not capable of dissolving intact apatite. Incorporated strontium occupies calcium lattice positions in the bioapatite, and the acid treatment removes diagenetic strontium that is necessarily superficially bound to the apatite crystal since solid state transitions are not possible at soil temperature (Reynard, Balter 2014). The most plausible explanation is indeed a temporary negative calcium balance during ontogeny since the accumulation rate is assessed by integrating several years during ontogeny. Therefore, temporary negative calcium balances can still show up in the average. Also, the negative accumulation rates calculated for the childhood are rather low. The only individual with negative accumulation rates in both bones (compact femoral bone and rib corticalis) during adulthood is the young female from burial no 300. The negative rate in the faster remodeling rib (-2.8%) is much more pronounced than in the femur (-0.74%). Due to the association of strontium with calcium in the diet, this observation also implies that she had been in a negative calcium balance before she died. Since the skeleton did not exhibit osteologial symptoms of e.g. rickets, this negative strontium (and necessarily also calcium) balance is most plausibly explained by pregnancy and birth shortly before her death. This is supported by the differing accumulation rates in rib and femur, and in particular by a pronounced *sulcus praeauricularis* (Wahl 1988).

Also the calculated bone formation rates of the skeletal finds from Stettfeld (*Table 4*) are well comparable with modern such values (Marshall *et al.* 1972, International Commission on Radiological Protection 1973) and the average bone turnover rate in adults. Higher such values are estimated only for the young adults, whereby also modern values are elevated in this age group. Also, Shagina *et al.* (2015) report a ^{90}Sr clearance rate up to 18% in young adults of both sexes.

Although the measurement data (isotopic ratios and element concentrations) are physiologically plausible, both tooth enamel and bone have been investigated. Some researchers are reluctant to measure trace elements and isotopic ratios in bone apatite because of its higher susceptibility towards diagenetic alterations (e.g. Trickett *et al.* 2003, Bentley 2006 and references therein). Both lead and strontium in the burial environment are bone-seeking elements and tend to contaminate archaeological skeletons. The peri- and endosteal parts of a sample are most affected by soil/bone interactions and were removed mechanically. However, bone is a porous substance. Therefore, an acid wash in an appropriate acid that takes the different solubilities and solubility products of Ca-, Sr- and Pb-salts into account is capable of removing both the outer and inner surfaces of the sample, and getting rid of newly formed salts. Bone mineral is a carbonate-hydro-apatite (Greiner *et al.* 2019) and it had been shown previously that the solubility product of a calcium-deficient hydroxyapatite is lower than in stoichiometric hydroxyapatite (Liu *et al.* 1999, Bufler 2004). Heavily contaminated bone (and also enamel, see de Winter *et al.* 2019) therefore tends to rapidly disintegrate during the acid pretreatment and the sample quickly loses weight. To all experience, such samples are unsuitable for trace element or apatite stable isotope analyses, and a stage where leachate and remaining sample exhibit the same element concentration and isotopic signature will hardly if ever be reached. The removal of contaminating Pb necessitates stronger acids. Some researchers claim that archaeological strata are hardly affected by anthropogenic lead because Pb is largely retained in the topsoils (United Nations 2010, Bacon *et al.* 1996, Budd *et al.* 2000), but this does not necessarily hold true for ancient mining regions. It is, however, possible to relate $^{207}Pb/^{206}Pb$ to $^{208}Pb/^{206}Pb$, following common practice in environmental science and permitting a comparison with the European Standard Lead Pollution regression line (ESLP; Haack et al. 2002, 2003). Both regressions differ significantly from each other: $^{208}Pb/^{206}Pb = 1.1719 \times {}^{207}Pb/^{206}Pb + 1.0907$ (ESLP), and $^{208}Pb/^{206}Pb = 1.6059 \times {}^{207}Pb/^{206}Pb + 0.7204$ (Stettfeld samples). We therefore conclude that also the measured lead stable isotopes are in fact biological rather than diagenetic signals. This is supported by the fact that in this study, not only enamel isotopic ratios, but also bone isotopic ratios plot outside the estimated local range for $^{87}Sr/^{86}Sr$ and exhibit a likewise considerable variability for lead isotopic ratios.

In addition to stable isotope analyses, also the measured element concentrations give clues to whether the samples may still be contaminated and the laboratory processing unsuccessful accordingly. The concentration of strontium in bone and teeth is strongly diet dependent. For a direct comparison of physiological data with concentrations in archaeological bones and teeth, a conversion of the data into the unit µg/g (ash) is often necessary. A review of reported modern concentrations of strontium and lead in the human skeleton and dentition including this necessary conversion into the common dimension µg/g (ash) has been performed by Neußer (2017; Appendix). In cortical bone, Sr concentrations vary from less than 100 ppm up to >500 ppm with no apparent consistent difference between compact and cortical bone. In permanent teeth, Sr concentrations in enamel and dentin vary between about 100 and >300 ppm. Modern lead concentrations in bone and teeth vary between about 5 until 80 ppm. Pb concentrations in the Stettfeld skeletons average only 4.6 ppm, with a range from 0.3 to 20.5 ppm. They are thus consistent with physiological concentrations and do not indicate *post mortem* contamination. We conclude that also the lead stable isotopic ratios in both tissues are not biased by diagenesis what was already supported by the deviation from the ESLP.

In a few specimens only, Sr concentrations exceed 600 ppm what holds for both bone and enamel. Sample surfaces had been removed by grinding and etching, whereby the etching steps remove another about 200 µm. Trace element concentration profiles of archaeological bones are typically U-shaped with higher concentrations by contamination in the periosteal and endosteal regions that, however, do not exceed a few hundred micrometers even in fossil bone (Williams 1988). In both modern and archaeological teeth, lead concentrations decrease from the outside to the enamel/dentin-junction (Purchase, Fergusson 1986). Finally, the distribution of both $^{87}Sr/^{86}Sr$ and lead stable isotopes among the samples match the regional geological differences, and both exhibit a north-to-south variation (Ströbele *et al.* 2012, Sjøgren *et al.* 2016) (see below).

Primarily non-local individuals immigrated to the site during childhood or early juvenile age. With the exception of the young female (grave 300), where a plausible explanation for her migration would be exogamy, the young age of the other individuals at residence change suggests migration in (family) groups (in Roman times also as slaves). The female from burial 300 died between 25 and 30 years, and had changed her place of residence as a juvenile (12–19 years). This individual appears non-local also for other reasons, in particular its artificially deformed skull. Distance covered by the immigrants was however most probably not very large. Since similar $^{87}Sr/^{86}Sr$ isotopic ratios occur at many places, the most parsimonious place of origin should be chosen. A strontium isotopic map of the region in question has been published by Sjøgren *et al.* (2016) and according to it, individuals with $^{87}Sr/^{86}Sr$ isotopic ratios > 0.71 have most probably spent their childhood in the Black Forest region (*Figure 1*) that extends up to 100 km to the south of Stettfeld. Non-local individuals with isotopic ratios < 0.709 and especially the female no 300 could have originated from the east of the site at similar distances. Possible places of origin can thus be found at distances that could be covered by foot in a few days. Therefore, all individuals should have originated from more or less the same microregion.

When lead stable isotopic ratios are compared with regional geological data provided by Durali-Müller *et al.* (2007) and Ströbele *et al.* (2012, 2015), all specimens but two exhibit isotopic ratios that are compatible with galena from several sites in the Black Forest or those at Wiesloch, the deposit at closest distance to the burial site (*Figure 1*). The mining regions in question are located to the east of the Rhine river at distances matching those of the possible places of origin of the non-locals. The conclusion from both strontium and lead stable isotopes is that the non-locals originated from some places in the Black Forest area that had been sought after for its ore deposits (Hildebrandt 2004). Therefore, the people at Roman Stettfeld could have had contact with lead derived from several of the available ore deposits in relative vicinity to the settlement, and it comes to no surprise that the majority of the measured lead data are consistent with isotopic ratios from several such sites and should even consist of mixed isotopic ratios. In addition, every mining and metal working site will lead to a spatial dispersion of lead by dust and aerosols. It may well be possible that people with different occupations might be distinguishable from each other by lead stable isotopes in the skeleton, but this was not the aim of this study and cannot be achieved by only 12 individual skeletons tested.

Age-trends and a comparison of the distribution of lead isotopic signatures with $^{87}Sr/^{86}Sr$ are suitable for the confirmation of hypotheses related to migration. Only two individuals (burials 26 and 348B) exhibit enamel lead isotopic ratios that fall outside the respective ratios of lead ore deposits in the Black Forest. This implies that both individuals might not have originated from the same microregion as the others, but could have travelled a longer distance. Recent archaeometallurgical research at Wiesloch came to the conclusion that Roman mining and smelting at this site was only performed with local material (Ströbele *et al.* 2015). This is in agreement with the lead isotopic signatures measured in the other skeletons from Stettfeld. The individual 348B had been buried together with 348A in the same grave what does not necessarily imply that they were related to each other. They might as well simply have died around the same time. While both individuals were primarily not local to the site of their recovery according to the Sr isotopes, they had at least contact with different lead sources during their childhood. Therefore, while the analysis of $^{87}Sr/^{86}Sr$ alone suggests origin from more or less the same microregion according to the principle of parsimony, additional information by lead stable isotopes and comparison with local such ratios opens up the probability of far distance mobility and migration of selected individuals.

CONCLUSION

Detecting immigrants to a site by $^{87}Sr/^{86}Sr$ isotopic ratios has become routine in archaeobiology since decades. Because the strontium metabolism in humans has been studied in detail, the measurement of strontium concentrations in the skeletal apatite is capable of giving additional information on the probable age at residence change. This has implications for the reconstruction of past social peculiarities, e.g. whether immigrants had migrated during childhood, whether they had married in, or whether migration was a reaction of pull factors such as the occurrence of raw material and the prospect of labour.

In more recent times, apatite lead stable isotopic ratios have also proven their suitability for georeferencing skeletal finds. This method faces difficulties in case of metal working societies, where a lead industry and the exploitation of several ore deposits will result in mixed isotopic ratios, a common result in archaeometallurgical research. Therefore, it depends on the specific archaeological question to be solved whether lead stable isotopes will be suitable for georeferencing people. In case of the Stettfeld skeletons, it was due to outliers in lead isotopic ratios that the parsimonious interpretation of $^{87}Sr/^{86}Sr$ as being indicative of a restricted catchment area might have to be expanded, and travelling larger distances of particular individuals can no longer be fully excluded.

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APPENDIX:

Literature data on modern strontium and lead concentrations in bones and teeth (compilation by Neußer 2017)

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