

# Determination of Geo-dependent Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ Isotopic Ratios for Archaeological Sites from the Inn Valley (Austria): A Model Calculation

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## Introduction

Numerous attempts have been made for an assessment of local bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios, the knowledge of which is essential for the determination of local or primarily non-local bio-archaeological finds at a site. The majority of these investigations focus on the comparison of measured stable strontium isotopic ratios in archaeological bones and teeth with those of the regional geology (e.g. Grupe et al. 1997; Müller et al. 2003; Tütken 2010). It has only occasionally been attempted to introduce other factors such as water and vegetation into the discussion on local bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios (see Drouet et al. 2005; Xin and Hanson 1994). Our study tries to integrate all relevant factors which mix in the consumer's tissues, be it man or animal. Vertebrates ingest strontium with their food and drinking water, whereby the strontium isotopic ratio of plants is influenced to a major degree by the respective ratio of the atmosphere and the soluble mineral soil components.

We undertook detailed re-investigations of several archaeological sites to establish a firm basis for model calculations of local bioavailable strontium isotopic signals, which are in turn based on mixing models integrating strontium isotopic ratios and strontium concentrations of the relevant parameters water, soil, and vegetation (Faure and Mensing 2005; Grupe et al. 2011). The first data sets, the results of which are presented in this chapter, concentrate on the Inn Valley, part of

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the archaeologically highly relevant transalpine Inn-Eisack-Etsch-Brenner passage (see [www.for1670-transalpine.uni-muenchen.de](http://www.for1670-transalpine.uni-muenchen.de)). It was expected that the model calculation should lead to more reliable results with regard to the determination of place of origin of individual finds, which are hitherto frequently fraught with ambiguity.

It should be stressed, however, that model calculations alone do not permit the assessment of the reliability and correctness of this method. It is necessary to test and control the calculated data by analysis of  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios in archaeological vertebrate bone finds from the respective sites. Here, three species were chosen: domestic cattle (*Bos taurus*), domestic pig (*Sus domestica*), and red deer (*Cervus elaphus*). Taken all data together, a reliable image of the local, geo-dependent bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio emerges. This is in turn indispensable for any attempt to quantify immigrant people or imported animals (or animal parts/raw material) at a site to firmly distinguish mobility from migration and trade.

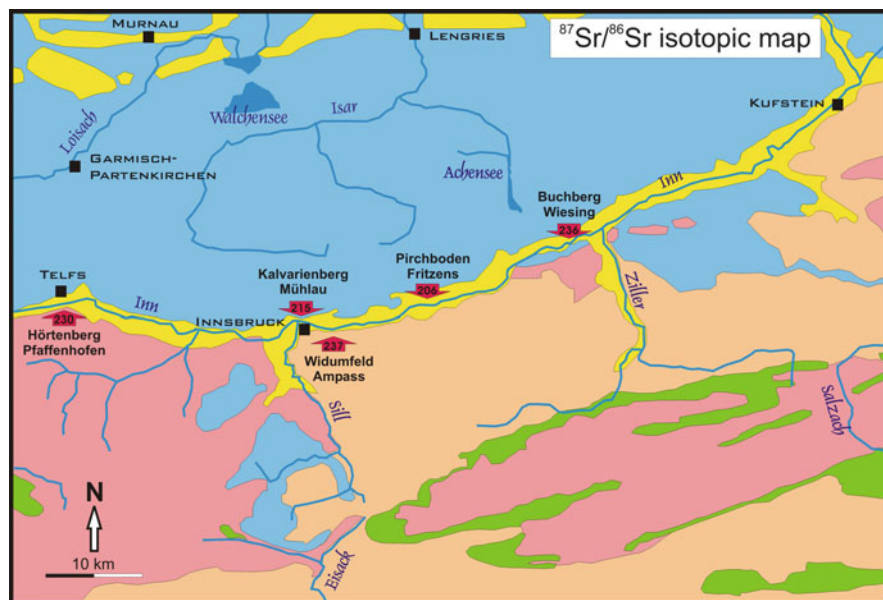
## Material: Samples and Site Description

Between 2013 and 2014, groundwater (naturally composed of atmospheric and soil conditioned components), “weathered soil” (comparable to acid leached soil), and vegetation from selected archaeological sites throughout the Inn Valley were sampled during several field excursions (Fig. 1). Soil samples were taken from the archaeological strata of the chosen sites to establish a baseline soil composition in order to correctly assess the growth of past and wild cultivated plants as closely as possible. Care was taken to exclude possible influences of modern fertilisers. Soil samples were subjected to a leaching step prior to analysis (see below; see also Drouet et al. 2005). Groundwater was taken from neighbouring springs or wells and therefore consists of both precipitation (rain water) and surface water, enriched with soil components. Unfortunately, no archaeological plant material was available for analysis. Modern vegetation samples were therefore taken directly from the location of the soil sample. To avoid species-specific peculiarities, hazelnut branches (*Corylus avellana*) were sampled because of their availability at nearly every site (Göhring 2014).

The following archaeological sites were chosen:

**Fritzens Pirchboden (Code Number 206)** The Pirchboden is a hill site located above the town of Fritzens, the type locality of the Fritzens-Sanzeno culture (sixth century bc; Lang 1998). The archaeological soil consists of argillaceous sand, light brown with crystalline components of boulder pavement of varying size, definitely of moraine origin ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.714$ , profile depth = 100–110 cm). Branches of hazelnut were sampled, and spring water from a nearby well.

**Innsbruck-Mühlau, Kalvarienberg (Code Number 215)** The soil was taken from the sidecut of a road. It consists of brown rendzina with calcareous fragments and some crystalline pebbles ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.70876$ , profile depth = 25–30 cm). It was



**Fig. 1**  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic map of the Inn Valley between the cities of Telfs and Kufstein. The investigated archaeological sites are marked with numbers. The Inn Valley is one of the largest East–west extending valleys of the Alps filled with glacial and fluvial sediments (yellow), occasionally cut through by tertiary sandy outcrops. Moraine and fluvial sediments of the Inn Valley prevalently enriched with crystalline rocks have mixed  $^{87}\text{Sr}/^{86}\text{Sr} < 0.709\text{--}0.7135$ . The Inn Valley forms the boundary between the Northern Calcareous Alps (blue, various types of calcareous rocks,  $^{87}\text{Sr}/^{86}\text{Sr} = 0.707\text{--}0.709$ ) and the Central Alps which are made primarily of crust-dominated crystalline rocks (pink—of acid magmatic origin, and beige—of sedimentary and acid volcanic origin,  $^{87}\text{Sr}/^{86}\text{Sr} > 0.71$ ). Lacustrine sediments, partly of tertiary age and often positioned on high Inn terraces underlain by crystalline rocks, have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios  $> 0.7135$ . Mantle-dominated rocks are shown in green ( $^{87}\text{Sr}/^{86}\text{Sr} < 0.707$ )

covered by landslide material interspersed with carbonate fragments only. Vegetation (hazelnut branches) and water from a well were sampled next to church at the Kalvarienberg.

**Pfaffenhofen/Ilm, Hörtenberg (Code Number 230)** The Hörtenberg is a conspicuous hill near the town of Pfaffenhofen and is composed of quartz-phyllite. The Iron Age archaeological site (Fassbinder 2010) is located at the northern hill slope near the “Maierhof” farm. The soil sample was taken from a road sidecut, 100 cm below surface. It consists of grey soft clay ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.72533$ , profile depth 100–130 cm), whereby the whole fine-grained loamy profile intercalated with chalk bands suggests a lacustrine sequence. The vegetation sample again consists of hazelnut branches; spring water originates from a well inside the “Maierhof” farm.

**Wiesberg Buchberg (Code Number 236)** The Buchberg is a conspicuous hill built by Triassic carbonate rocks overlain by glacial sediments loaded with rounded

crystalline components, and was once a Bronze Age settlement site (Pöll 2014). The archaeological soil sample consists of sandy silt ( $^{87}\text{Sr}/^{86}\text{Sr}=0.71081$ , profile depth = 50–60 cm). The vegetation sample is hazelnut; spring water originates from a fountain in the nearby village of Jenbach.

*Ampass Widumfeld (Code Number 237)* The Widumfeld is located east of the town of Ampass and is of archaeological relevance from the Iron Age until Roman times (Tomedi et al. 2001; Castellan and Tomedi 2006). The soil sample ( $^{87}\text{Sr}/^{86}\text{Sr}=0.71736$ , profile depth 50–60 cm) contains small pebbles of quartz-phyllite and probably relates to an argillaceous sandy valley or lake filling from post-Roman times. We assume that the sediment source has remained similar during post-glacial times and that no fundamental change in strontium isotopic ratios of the reservoir has taken place. Again, vegetation samples consist of hazelnut branches; spring water originates from the nearby forest.

## Analytical Methods

*Bone* The surfaces of the bone samples were manually removed by grinding, and the remaining sample was washed ultrasonically in distilled water until the water remained clear. The air-dried sample was then defatted with diethylether for 5 h in a Soxhlet, air-dried and etched ultrasonically with HCOOH (98 %) for 5 min. Finally, the sample was washed in distilled water until the wash solution reached a pH of 5–6. The air-dried sample was then ashed for 12 h at 800 °C in a muffle furnace and homogenised to a fine powder. This bone meal was dissolved in 1 mL concentrated HNO<sub>3</sub> (65 %) on a hot plate at 100 °C and dried. For the column separation, the sample was dissolved in 1 mL 6 N HNO<sub>3</sub>. This solution was used for the separation of the strontium fraction for the isotopic analysis by mass spectrometry, after a chromatographic column separation by use of Sr SPEC resin. All acids were of ultrapure quality. Distilled water was always of doubly distilled quality.

*Water* 1.5 L water was filtered through an MN 615 filter (if necessary). Ten millilitre of the sample was then evaporated to dryness and dissolved for 14 h in 65 % HNO<sub>3</sub> in a Teflon beaker on a hot plate at 100 °C, followed by a chromatographic column separation (see above).

*Soil* The soil samples were dried and sieved with a 500 µm sieve. Three hundred milligram of this sample was dissolved in a Teflon beaker in 2 mL 35 % HCl (suprapure) for 12 h at 120 °C, cooled to room temperature and centrifuged at 11,000 rpm for 10 min. The clear solution was separated and evaporated to dryness, followed by a chromatographic column separation (see above).

*Wood* From 150 mg of hazelnut wood each, only the annual rings were analysed; all other parts were removed mechanically. The wood was dried at 80 °C in an oven to constant weight, and then ashed for 12 h at 800 °C. Four hundred milligram of the ash was processed by chromatographic column separation (see above).

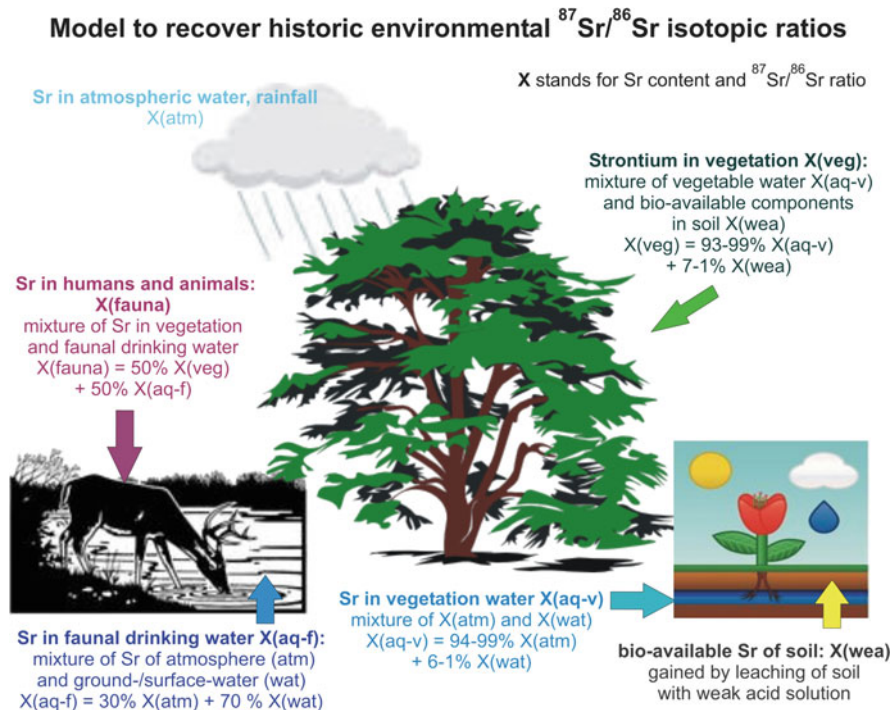
*Mass Spectrometry*  $^{87}\text{Sr}/^{86}\text{Sr}$  was analysed with a Thermal Ionisation Mass Spectrometer Finnigan MAT 261.5 on single tungsten filaments. Significant extant Rb was evaporated from the loaded filament by controlled preheating before the isotopic Sr composition was measured. For quality control and to check for the proper operation of the mass spectrometer, a certified reference material was measured under the same conditions as the samples ( $\text{SrCO}_3$ , NIST SRM 987,  $^{87}\text{Sr}/^{86}\text{Sr}$ :  $0.710210 \pm 0.000056$  STD,  $n = 110$ ). Isotope mass fractionation during analysis was corrected by referencing to an invariant  $^{88}\text{Sr}/^{86}\text{Sr}$  value of 8.37521. Total analytical uncertainty (precision + accuracy) for  $^{87}\text{Sr}/^{86}\text{Sr}$  on natural samples is assumed to be  $<50$  ppm. Standard Reference Material SRM 1400 “Bone ash” (NIBS, Washington DC) was used with regard to the wet ashing and Rb–Sr separation. Measurement precision was  $\pm 0.00001$ .

## **Strontium Mixing Model for the Assessment of Local Geo-dependent Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ Isotopic Ratios in Archaeological Fauna**

Humans and animals ingest strontium with food and drinking water hence strontium from these sources with their source-specific isotopic ratio mixes in the consumer’s body. The  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio in a consumer’s tissue is therefore always a mixed isotopic ratio. In plants,  $^{87}\text{Sr}/^{86}\text{Sr}$  is largely due to the atmospheric water and the solubilised mineral component in soil. It became obvious in the course of our research that local faunae and florae are not supplied with a uniform water composition, but that the mix of groundwater [groundwater  $X(\text{wat})$ ] and ocean or rainwater  $X(\text{atm})$ ] could exhibit completely different values (see Fig. 2).

Chosen end members for the water composition are those of the North Atlantic ocean water ( $^{87}\text{Sr}/^{86}\text{Sr}(\text{atm}) = 0.709024$ ,  $\text{Sr}(\text{atm}) = 0.0013$  ppm; Veizer 1989) and of the locally sampled groundwater  $X(\text{wat})$ , respectively. Groundwater can be described as rainwater, the composition of which has been altered by solubilised mineral components in the soil. It is necessary to stress that no surface water has been sampled for this study, but rather water from shallow fountains (5–6 m maximum) or from springs near their bottom. Due to the residence time of the water in the soil, a measurable uptake of mineralogical components which are clearly related to the parent rock is evidenced (Table 1).

For an assessment of the other components contributing to the mixed isotopic ratio measured in the consumer’s skeleton, soil and vegetation samples were taken and measured. Soil was sampled from the archaeological sites at a level equivalent to the surface at the time of the prehistoric settlement whenever possible. This way, the strontium isotopic ratio of such soil was measured that had once driven the prehistoric plant growth and was introduced into the prehistoric food chain. The soil samples were processed and leached to access a  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio comparable to the former bioavailable one. Since no archaeological plant material was available,



**Fig. 2** Scheme for the recovery of local geo-dependent  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios. In this model, strontium from soil, vegetation, and water is analysed to evaluate the bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio which characterises the place of residence in historical times (see text)

modern hazelnut was chosen not only because it was abundant at most of the former archaeological localities, but especially because the main roots reach into a depth of 30–40 cm and thus below the agricultural horizon.

For this pilot study, five archaeological sites from the Inn Valley were chosen, whereby the local geology is characterised by lacustrine-fluvial sediments (sites nos 230 and 237), or by glacial sediments of the last glaciation period (sites nos 206 and 236). In contrast, the Innsbruck/Mühlau Kalvarienberg site (no 215) exhibits a soil composition which is best described as weathered soil on carbonate rock (rendzina). Archaeozoological remains of the three vertebrate species mentioned above were available from all five sites.

For the calculation of the local bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio which should be reflected in the skeleton of residential animals (and also in humans; see Table 2 in the appendix), the following published data were used:

For the atmospheric water component  $X(\text{atm})$ , we took the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio and the strontium content of modern ocean water ( $0.709024 \pm 0.000032$ ; Veizer 1989). Other values published by e.g. Graustein and Armstrong (1983) or Dupré et al. (1994) are influenced by aerosol and dust particle burdens (Xin and Hanson

**Table 1** Measured  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios in archaeological animal bone finds, and calculated geo-dependent  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, affected by local parameters of water, soil, and vegetation

Sample no.	Locality	Groundwater			Hazelnut			Soil leached		
		$^{87}\text{Sr}/^{86}\text{Sr}$ (wat) measured	STDev	f(atm.aq-v) <sup>a</sup> in %	$^{87}\text{Sr}/^{86}\text{Sr}$ (veg) measured	STDev	f(aq-v, veg) <sup>b</sup> in %	$^{87}\text{Sr}/^{86}\text{Sr}$ (wea) measured	STDev	
206	Pirchboden Fitzens	0.710884	0.0001	96	0.713703	0.000046	92.8	0.714 <sup>c</sup>	0.000073	
215	Kalvarienberg Mühiau/Innsbruck	0.70799	0.000142	99.5	0.708756	0.000043	99.9	0.708384	0.000139	
230	Hörtenberg Pfaffenhofen/Inn	0.715413	0.000124	94	0.713103	0.000046	99.9	0.725331	0.000027	
236	Buchberg Wiesing/Inn	0.710884 <sup>f</sup>	0.0001 <sup>f</sup>	96	0.71046	0.000059	99.9	0.716711	0.000068	
237	Widumfeld Ampass	0.716387	0.000065	95	0.716514	0.000076	96.4	0.717355	0.0001	
Localised										
Sample no.	$^{87}\text{Sr}/^{86}\text{Sr}$ (fauna) calculated	$^{87}\text{Sr}/^{86}\text{Sr}$ (fauna) STDev (mean)	+Cut-off calculated error	-Cut-off calculated error	+Cut-off adopted+ 0.001	-Cut-off adopted- 0.001	$^{87}\text{Sr}/^{86}\text{Sr}$ pig (bone)	$^{87}\text{Sr}/^{86}\text{Sr}$ cow (bone)	$^{87}\text{Sr}/^{86}\text{Sr}$ deer (bone)	
			0.715083	0.710398	<b>0.71354</b>	<b>0.71154</b>				
206	<b>0.71254</b>	0.000781	0.715083	0.710398	<b>0.71354</b>	<b>0.71154</b>	0.710892 <sup>d</sup> <b>0.713180<sup>e</sup></b> 0.710987	0.714179, <b>0.711711</b>		
215	<b>0.708032</b>	0.000078	0.708368	0.7079	<b>0.709032</b>	<b>0.707032</b>	0.709437 0.710142 0.713157	0.709928 0.710964 0.709271		
230	<b>0.714913</b>	0.000604	0.716264	0.712638	<b>0.715913</b>	<b>0.713913</b>		<b>0.714405</b> <b>0.715243</b> 0.716443		

(continued)

Table 1 (continued)

Sample no.	Localised				Local animal bone findings				
	$^{87}\text{Sr}/^{86}\text{Sr}$ (fauna) calculated	$^{87}\text{Sr}/^{86}\text{Sr}$ (fauna) STDev (mean)	+Cut-off calculated error	-Cut-off calculated error	+Cut-off adopted+ 0.001	-Cut-off adopted- 0.001	$^{87}\text{Sr}/^{86}\text{Sr}$ pig (bone)	$^{87}\text{Sr}/^{86}\text{Sr}$ cow (bone)	$^{87}\text{Sr}/^{86}\text{Sr}$ deer (bone)
236	<b>0.710807</b>	0.000174	0.711254	0.710214	<b>0.711807</b>	<b>0.709807</b>	<b>0.711722</b> 0.712871 <b>0.711522</b>	<b>0.711136</b> 0.712383 <b>0.709828</b>	<b>0.716071</b> <b>0.716724</b>
237	<b>0.716342</b>	0.000043	0.716551	0.716294	<b>0.717342</b>	<b>0.715342</b>	<b>0.716362</b> <b>0.716544</b> <b>0.716576</b>	<b>0.716648</b> <b>0.716429</b> <b>0.716634</b>	<b>0.716071</b> <b>0.716724</b>

<sup>a</sup>Abundance of atmospheric water in vegetation water (water absorbed from vegetation, ca. 95 % is assumed)

<sup>b</sup>Abundance of vegetation water in vegetation (correlated with f(atm, aq-v), ≤100 % is precondition)

<sup>c</sup>Isotopic ratio is estimated, according to soil type

<sup>d</sup>Normal: isotopic ratio lies outside the cut-off range (sample is non-local)

<sup>e</sup>Bold: isotopic ratio lies inside the cut-off range (sample is local)

<sup>f</sup>Isotopic ratio is that of sample 206

For the cut-off values, a fixed value (±0.001) was used, see text



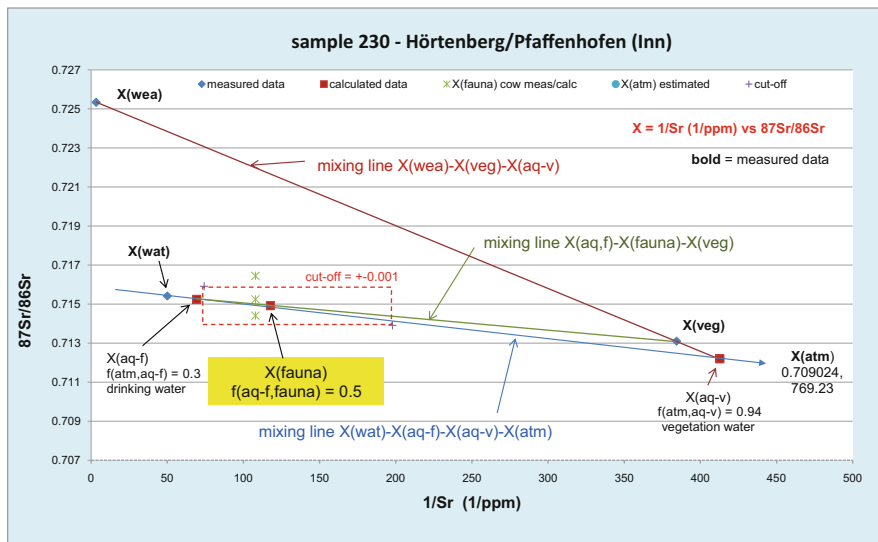
1994). Strontium concentrations in precipitation can be highly variable but usually do not exceed 1 ppb (Xin and Hanson 1994). For Central Belgium, Drouet et al. (2005) measured a mean concentration of  $1.1 \pm 0.3$  ppb ( $n=9$ ). For our model calculation, we took an atmospheric strontium concentration  $\text{Sr}(\text{atm})$  of 0.0013 ppm. This value correlates with that of sample BP1 determined from a post-glacial environment in Central Belgium (Drouet et al. 2005).

$^{87}\text{Sr}/^{86}\text{Sr}$  of groundwater  $X(\text{wat})$  was measured; the strontium content was estimated. Both the stable strontium isotopic ratio and the strontium content of groundwater adjust to the respective local geological settings dependent on the residence time in the soil. Xin and Hanson (1994) published measurements of both soil and groundwater in the Peconic river watershed (New York) and found  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios between 0.71 and 0.7113 and strontium contents between 25 and 34 ppb. It is obvious that both the strontium concentration and the strontium isotopic ratio adjust to the geological conditions according to the residence time below ground (Voerkelius et al. 2010). For Denmark, Frei and Frei (2011) published decreasing mean strontium concentrations between 1 and 0.125 ppm for surface water with a  $^{87}\text{Sr}/^{86}\text{Sr}$  variability between 0.708 and 0.711. Accordingly, we chose a strontium concentration of 0.06 ppm for carbonatic water with a  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio  $<0.709$ , 0.03 ppm Sr for soils with a  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio between 0.709 and 0.7135, and 0.02 ppm Sr for soils with a  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio  $>0.7135$ .

With these isotopic and concentration data, the stable strontium isotopic ratio of the water consumed by the fauna [drinking water  $X(\text{aq-f})$ ] can be calculated. We suggest that the proportion of atmospheric water  $f(\text{atm}, \text{aq-f})$  in the drinking water averages 30 %. This percentage correlates with the one calculated from data of Xin and Hanson (1994) as a mixture of soil water (10–50 cm depth) and atmospheric (rain) water component.

With regard to the strontium concentrations in soil [ $\text{Sr}(\text{wea})$ ] and the frequency distribution of the water components in the atmosphere [ $f(\text{atm}, \text{aq-f})$ ], and the fauna [ $(\text{aq-f}, \text{fauna})$ ], variations of 50 % were assumed. Standard deviations (STDev) were taken into account with regard to the measurement error. To present an example, the complete model calculation is shown in Table 2 (see appendix) for sample no. 230 and visualised by the resulting mixing diagram (Fig. 3).

It is depicted from Fig. 3 that the data points for drinking water  $X(\text{aq-f})$  and the sampled groundwater  $X(\text{wat})$  plot closely together ( $X$  stands for both the isotopic ratio and the strontium concentration of the respective components). With regard to this position in the mixing diagram, drinking water cannot combine with the measured soil components  $X(\text{wea})$  to the strontium mixing in the vegetation  $X(\text{veg})$ . Consequently, the water taken up by the vegetation must have a composition different from that of the drinking water of the fauna. The vegetation water  $X(\text{aq-v})$  should rather plot at the intersection of the mixing lines groundwater  $X(\text{wat})$ —atmospheric water  $X(\text{atm})$  and soil  $X(\text{wea})$ —vegetation  $X(\text{veg})$  (Fig. 3). The calculated data for the vegetation water in Fig. 3 are  $^{87}\text{Sr}/^{86}\text{Sr}(\text{aq-v}) = 0.712189$



**Fig. 3** The mixing diagram illustrates the mixing of three components with different strontium contents and different stable strontium isotopic ratios: groundwater  $X(wat)$ , leached soil  $X(wea)$ , and vegetation  $X(veg)$ . *Straight lines* represent the non-linear mixing of the respective end members.  $X(aq-v)$  = vegetation water is a mixture of groundwater  $X(wat)$  and rain/atmospheric water  $X(atm)$  with a proportion  $f(atm, aq-v)$  of 94 %.  $X(aq-f)$  = drinking water is a mixture of the same components with a proportion  $f(atm, aq-f)$  of only 30 %. Strontium in vegetation [ $X(veg)$ ] is made up of 99.9 % strontium from vegetation water  $X(aq-v)$  and only 0.1 % of strontium from soil water  $X(wea)$ . The geo-dependent bioavailable strontium composition in faunal bone [ $X(fauna)$ ] is a 1:1 mixture of strontium from drinking water  $X(aq-f)$  and from vegetation  $X(veg)$ . All investigated samples reflect a far less abundance of strontium from soil than from the surrounding water component for the creation of the local bioavailable  $^{87}Sr/^{86}Sr$  isotopic ratio than expected. Acronyms: see appendix

and  $Sr(aq-v) = 0.00242$  ppm, whereby the atmospheric contribution to the vegetation water  $f(atm, aq-v) = 94$  %. Note the nonlinear variation of  $f(atm, aq-v)$  along the straight mixing line in the diagram (see Faure and Mensing 2005). With a proportion  $f(aq-v, veg)$  of more than 99 %, the influence of the strontium component in vegetation water to the whole vegetation is remarkably high.

We would like to stress the dependencies between the proportion of rainwater in vegetation water [ $f(atm, aq-v)$ ] and the proportion of vegetation water in the whole vegetation [ $f(aq-v, veg) = \text{factor } K$  (see Table 2)].  $K$  is calculated by use of the strontium concentrations from vegetation, vegetation water, and leached soil. In the case of  $K = 1$ , data points  $X(veg)$  and  $X(aq-v)$  would plot in the same place, and a 100 % dependency of  $^{87}Sr/^{86}Sr$  of the whole vegetation from  $^{87}Sr/^{86}Sr$  in vegetation water would be the case. Since this is rather improbable, we chose an  $f(atm, aq-v)$  which renders  $K < 1$  automatically. This way,  $f(atm, aq-v)$  declines to 0.94 (sample 230). But still, only 6 % of the  $^{87}Sr/^{86}Sr$  ratio in the whole vegetation would be due to the soil component; 94 % would still stem from the water component.

While such a value seems to be very high, it is not unusual in nature (see e.g. Green et al. 2004; Grupe et al. 2011).

The strontium concentration of the leached soil  $\text{Sr}(\text{wea})$ , that of the vegetation water  $\text{Sr}(\text{aq-v})$  and its abundance in the vegetation  $f(\text{aq-v, veg}) = 0.9994$ , permits the calculation of the strontium concentration in the vegetation  $\text{Sr}(\text{veg})$  and results in 0.0026 ppm. This value, however, is far lower than some other published strontium concentrations in wood. Stem wood of different tree species had a mean concentration of about 5 ppm (Dijksta et al. 2003). Drouet et al. (2005) measured a mean concentration of 2.6 until 2.8 ppm for the genus *Fagus*, and Lambertz and Welling (2010) report a range of 1–10 ppm Sr in dry wood of various species. Just as in other tissues, Sr binds at Ca sites in plants which are mainly components of the cell wall (such as pectin acids, cellulose, and lignine; Torre et al. 1992), whereby the cellulose has a particularly low affinity to Ca and therefore also for Sr. Calculation of our model is performed under the assumption that in the course of mixing the components vegetation  $X(\text{veg})$  and water  $X(\text{aq-f})$  to form the Sr concentration in fauna  $X(\text{fauna})$ , the total Sr content of the vegetation is of no relevance but rather the bioavailable proportion of strontium only. We assume that it is only the strontium component which solubilised in the vegetation water is available to the consumer. This would in turn correspond to 0.05–0.1 % of the total strontium content of the plant.

Surprisingly, the calculated strontium concentration in animal bone [ $\text{Sr}(\text{fauna})$ ] is at first glance also very low (0.0085 ppm Sr according to the mixing diagram) and no longer comparable with the average total Sr content in bone (130–140 ppm; see Grupe et al. 1997). While the total Sr content in a bulk bone sample is the result of a long accumulation process lasting many years due to the long biological half-life of the apatite, the mixing diagram rather reflects the actual, time-dependent uptake in the course of the remodelling processes. Therefore, the resulting Sr content of 0.0085 ppm only is a function of the known slow remodelling of bone. Likewise, the low Sr content in vegetation which results from the mixing diagram is due to the time-dependent mineral uptake and precipitation into the plant tissue.

For an assessment of the local bioavailable and geo-dependent stable strontium isotopic ratio in vertebrate bone, possible strontium components provided by the diet are vegetation  $X(\text{veg})$  and drinking water  $X(\text{aq-f})$ . Assuming that both components mix with the same proportion [ $f(\text{aq-f, fauna}) = 0.5$ ], then the local  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio for the find with the code number 230 is 0.714913 (Table 2, appendix).

Table 2 also presents an error estimation for the data. For the measured  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, the standard deviation of the measurement value was used, but no error estimation was carried out with regard to the Sr contents. Even a 100 % deviation only led to marginal changes. For the calculation of the proportion of the mixing components, a variation of 50 % was assumed with the exception of  $f(\text{atm, aq-v})$ , which varies between 0.1 and 2 %. A higher variation would be incompatible with the fact that  $K = f(\text{aq-v, veg})$  must not exceed 1. For the sample no. 230, error estimation resulted in  $^{87}\text{Sr}/^{86}\text{Sr} = 0.714913 \pm 0.000604$  (0.084 %, STDev, mean of errors).

## Results

Table 1 presents the results obtained at all five sites chosen for this pilot study.

While it is highly plausible that the  $^{87}\text{Sr}/^{86}\text{Sr}$  of the vegetation is largely due to the water that has been taken up by the plant, the low influence (1–6 %) of the bioavailable soil strontium component is surprising at first glance. This phenomenon is however particularly conspicuous when the  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios of groundwater and soil are significantly different from each other (see samples with the code numbers 230, 236). To relate the expected  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio in residential fauna to the bioavailable strontium isotopic ratio of the soil alone may therefore be totally misleading. Groundwater and vegetation indispensably need to be considered in addition.

The variability of local  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in fauna defines the cut-off value which differentiates between local and non-local individuals. One possibility of defining this cut-off value is the traditional statistical way by use of the threefold error ( $=3 \times \text{STDev}$ ). Other authors decided for a fixed value such as  $\pm 0.001$  (highly significant in terms of the measurement precision; see Grupe et al. 1997) which is also used in our study because on average, this value agrees well with the  $3 \times \text{STDev}$  of the calculated  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in fauna (Table 1;  $3 \times \text{STDev}_{\text{mean}} = 0.00101$ ). By defining the cut-off values as  $^{87}\text{Sr}/^{86}\text{Sr}(\text{fauna}) \pm 0.001$  this way, the variation of *calculated*  $^{87}\text{Sr}/^{86}\text{Sr}$  of local vertebrates with the code number 230 would range from 0.713913 to 0.715913. The *measured*  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in the three cattle bones from the site were  $0.714405 \pm 0.000135$ ,  $0.715243 \pm 0.000264$ , and  $0.716443 \pm 0.000141$  ( $\pm 2 \times \text{STDev}$ ). The first two individuals should therefore have been of local origin; the third one can no longer be considered local.

## Discussion

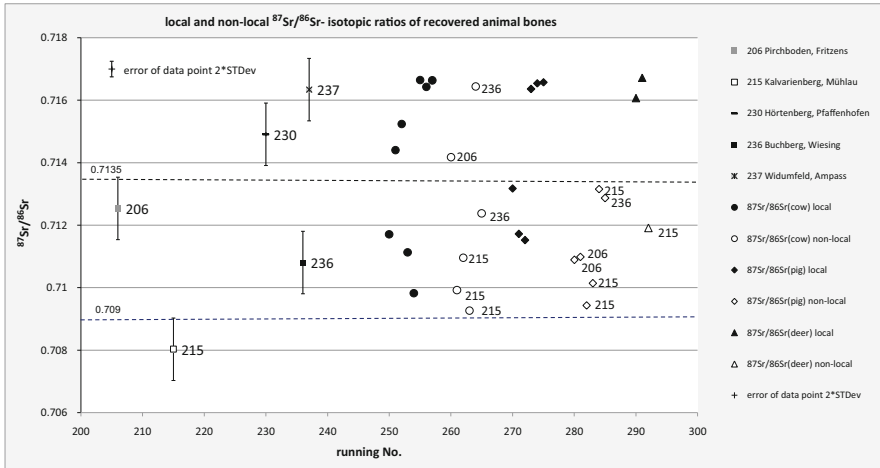
In sum, the analysis of archaeological animal bone finds and the modelled local, bioavailable  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios lead to the definition of three domains which correspond with the regional soil types (Fig. 4):

Domain 1:  $^{87}\text{Sr}/^{86}\text{Sr} < 0.709$ ; brown calcareous soil, rendzina.

Domain 2:  $0.709 \leq ^{87}\text{Sr}/^{86}\text{Sr} \leq 0.7135$ ; moraine soil, often in hillside location, coarse detrital and mostly crystalline material.

Domain 3:  $^{87}\text{Sr}/^{86}\text{Sr} \geq 0.7135$ , fluvio-lacustrine sediments (silty alluvial deposits, fine sand and silt, lacustrine clay partly of tertiary origin).

The majority of the investigated animals exhibit  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios between 0.709 and 0.7135 and must have spent their lives on moraine soil. This was expected since the Inn Valley glacier had once been one of the largest glaciers in the whole alpine area, filling the Inn Valley to a depth of several hundred metres.



**Fig. 4** Test of local or non-local origin of recovered animal bones from all five sites. Calculated local geo-dependent  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratios are indicated by site code numbers with the corresponding cut-off error bar ( $\pm 0.001$ ). Marked domains of different isotopic ratios ( $^{87}\text{Sr}/^{86}\text{Sr} < 0.709$ ;  $0.709-0.7135$ ;  $> 0.7135$ ) correlate with local geological sediment/soil types (see text). Non-local animals according to the model calculation are numbered and marked in *white* and local animals in *black*

At other locations such as Pirchboden (code no. 206) and Buchberg (code no. 236), animals had been slaughtered which had obviously been raised elsewhere. A complete agreement of all data (calculated local  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio and measured ratios of cattle, pig, and red deer) was found at only one site (Widumfeld, Ampass, code no. 237), and no agreement at all was found between calculated and measured data for Innsbruck-Mühlau, code no. 215).

In sum, the model calculations permit for a much finer-scaled solution of isoscapes in the Inn Valley as the available geological map (Fig. 1). Furthermore, an association of single non-local bone finds to defined regions is possible beyond the mere detection of non-local finds by way of the exclusion principle.

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Two hundred and thirty mean Hortenberg, Plattflohofen/ Iln	0.3	0.725331	0.000027	-3.209E-05	0.725438	0.00260	0.9994	0.5	-6.785E-06	0.715711	0.0085	<b>0.714913</b>	Cut-off=3x error	Cut-off= value±0.001
Estimated limit or calc./ measured error	0.45	0.725358		-2.858E-05	0.725422	0.00233		0.75	-6.234E-06	0.715826	0.0135	0.000450	0.716264	<b>0.715913</b>
Estimated limit or calc./ measured error	0.15	0.725304		-3.594E-05	0.725544	0.00288		0.25	-7.338E-06	0.715606	0.0051	0.000758	0.712638	<b>0.713913</b>

f(atm, aq-f)—Abundance of atmospheric water (=rainwater) component in the drinking water

f(atm, aq-v)—Abundance of atmospheric water (=rainwater) component in the vegetable water

f(aq-v, veg)—Abundance of vegetable water component in the vegetation

f(aq-f, fauna)—Abundance of drinking water component in the fauna

$^{87}\text{Sr}/^{86}\text{Sr}(\text{atm})$ —Sr isotopic ratio in the atmosphere (ocean water, rainwater, taken from literature)

$^{87}\text{Sr}/^{86}\text{Sr}(\text{wat})$ —Sr isotopic ratio in the groundwater (sampled water from wells or groundwater horizon)

$^{87}\text{Sr}/^{86}\text{Sr}(\text{aq-f})$ —Sr isotopic composition of drinking water (mixture of rain and groundwater) incorporated into fauna

$^{87}\text{Sr}/^{86}\text{Sr}(\text{aq-v})$ —Sr isotopic composition of vegetable water (mixture of rain and groundwater) assimilated in vegetation

$^{87}\text{Sr}/^{86}\text{Sr}(\text{veg})$ —Sr isotopic ratio in the vegetation (wood, measured)

$^{87}\text{Sr}/^{86}\text{Sr}(\text{fauna})$ —Sr isotopic composition of the fauna (localised Sr isotopic ratio, human and animal, calculated)

$^{87}\text{Sr}/^{86}\text{Sr}(\text{wea})$ —Sr isotopic composition of weathered soil, bioavailable component (gained by leaching of sampled soil with 1 N HCl, details see “Analytical methods”)

Sr(atm) ppm—Sr content of the atmospheric or rainwater (taken from literature)

Sr(wat) ppm—Sr content of the groundwater (taken from literature)

Sr(aq-f) ppm—Sr content in drinking water, mixture of ground and rainwater

Sr(aq-v) ppm—Sr content in vegetable water, mixture of ground and rainwater

Sr(fauna) ppm—Sr content incorporated into the fauna, time-dependent assimilated (human and animal, calculated)

Sr(veg) ppm—Sr content of the vegetation (wood, time-dependent assimilated, calculated)

Sr(wea) ppm—Sr content of the weathered soil (taken from literature)

## Abbreviations

f(atm, aq-f)	Abundance of the atmospheric water (=rainwater) component in the drinking water
f(atm, aq-v)	Abundance of the atmospheric water (=rainwater) component in the vegetation water
f(aq-v, veg)	Abundance of the vegetation water component in the vegetation
f(aq-f, fauna)	Abundance of the drinking water component in the fauna
$^{87}\text{Sr}/^{86}\text{Sr}$ (atm)	Sr isotopic ratio in the atmosphere (ocean water, rainwater, taken from the literature)
$^{87}\text{Sr}/^{86}\text{Sr}$ (wat)	Sr isotopic ratio in the groundwater (sampled water from wells or groundwater horizon)
$^{87}\text{Sr}/^{86}\text{Sr}$ (aq-f)	Sr isotopic composition of drinking water (mixture of rain and groundwater) incorporated into the fauna
$^{87}\text{Sr}/^{86}\text{Sr}$ (aq-v)	Sr isotopic composition of vegetable water (mixture of rain and groundwater)
$^{87}\text{Sr}/^{86}\text{Sr}$ (veg)	Sr isotopic ratio in the vegetation (wood, measured)
$^{87}\text{Sr}/^{86}\text{Sr}$ (fauna)	Sr isotopic composition of the fauna (localised Sr isotopic ratio, human and animal, calculated)
$^{87}\text{Sr}/^{86}\text{Sr}$ (wea)	Sr isotopic composition of weathered soil, bioavailable component (gained by leaching of sampled soil with 1 N HCl; for details see methods section)
Sr(atm) (ppm)	Sr content of the atmospheric or rainwater (taken from the literature)
Sr(wat) (ppm)	Sr content of the groundwater (taken from the literature)
Sr(aq-v) (ppm)	Sr content in vegetation water, mixture of ground and rainwater
Sr(aq-f) (ppm)	Sr content in drinking water, mixture of ground and rain water
Sr(veg) (ppm)	Sr content of the vegetation (wood, time-dependent assimilated, calculated)
Sr(fauna) (ppm)	Sr content incorporated into the fauna, time-dependent assimilated (human and animal, calculated)
Sr(wea) (ppm)	Sr content of the weathered soil (taken from literature)
X(...)	correlation of $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr content for the special case

Equation of the mixing hyperbola in coordinates of the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio and the Sr concentration (Sr) (Faure and Mensing 2005)

$(^{87}\text{Sr}/^{86}\text{Sr})_M = a/(\text{Sr})_M + b$ , with

$$a = \frac{\text{Sr}_A \text{Sr}_B \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_B - \text{Sr}_B \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_A}{\text{Sr}_A - \text{Sr}_B};$$

$$b = \frac{\text{Sr}_A \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_A - \text{Sr}_B \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_B}{\text{Sr}_A - \text{Sr}_B};$$



The numerical values of  $a$  and  $b$  entirely depend on the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios and Sr concentrations of the components  $A$  and  $B$  in the mixture  $M$ .

$$\text{Sr}_M = \text{Sr}_A \times f_A + \text{Sr}_B \times (1 - f_A);$$

$f_A$  is the abundance of  $A$  in the mixture  $M$

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