Archaeometry **44**, 1 (2002) 117–135. Printed in Great Britain

THE CHARACTERIZATION OF BIOLOGICALLY AVAILABLE STRONTIUM ISOTOPE RATIOS FOR THE STUDY OF PREHISTORIC MIGRATION*

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Strontium isotope analysis of bone and tooth enamel from prehistoric human skeletons is an important new technique used to address questions regarding migration. Two problems arise in such investigations: (1) levels of strontium isotope ratios in local bedrock, soil, water, plants and animals are variable; and (2) a range of values in human bone and enamel data make it difficult to distinguish some migrants from locals. Analysis of the bones of small animals provides a robust measure of local strontium isotope ratios and a reliable, if conservative, means for determining confidence limits for distinguishing migrants. Data from various geographical areas are presented here in a discussion of variability in strontium isotope values. Examples are provided using modern and prehistoric materials. We conclude with the recommendation that studies involving strontium isotope analysis should incorporate small animal samples for comparative purposes whenever possible.

KEYWORDS: ARCHAEOLOGY, MIGRATION, STRONTIUM ISOTOPES, BONE CHEMISTRY, ARCHAEOMETRY

INTRODUCTION

The use of isotopic tracers in the human body is well established in the field of medicine. In recent years, isotopic tracers have also been employed in environmental sciences and ecological studies to map the geographical movement of certain materials and species (see, e.g., Gosz *et al*. 1983; Rundel *et al*. 1989; Koch *et al*. 1992; Lajtha and Michener 1994; Åberg 1995; Chamberlain *et al*. 1997; Gannes *et al*. 1998). Similar methods were introduced in archaeology two decades ago for the investigation of residential change among prehistoric humans (Ericson 1981, 1985). Stable isotope ratios of several different elements have since been investigated as possible indicators of movement by prehistoric peoples (Katzenberg and Krause 1999). Oxygen (Stuart-Williams *et al*. 1995; White *et al*. 1998, 2000), lead (Carlson 1996; Gulson *et al*. 1997), and strontium (see, e.g., Ericson 1989; Sealy 1989; van der Merwe *et al*. 1990; *Sealy et al*. 1991; Price *et al*. 1994a,b, 1998, 2000, 2001) have been considered to date. Of these, strontium appears to be the most useful.

Two problems have emerged in studies involving the estimation of local isotope levels and the distinction of migrants from locals. In this paper, we address these problems and suggest some solutions. There are several parts to our essay. The first section documents some principles of strontium isotope analysis of human bone for information on residential change. The second section details the two problems and possible solutions. A third section discusses methods for

* Received 4 February 2001; accepted 22 March 2001.
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distinguishing migrant from local individuals in a human population. A fourth section concludes with some recommendations for dealing with these problems in future investigations.

THE PRINCIPLES OF STRONTIUM ISOTOPE ANALYSIS

Radiogenic ^{87}Sr is formed over time by the decay of rubidium (^{87}Rb, half-life ~ 4.7×10^{10} years) and comprises approximately 7.0% of total strontium (Faure and Powell 1972). The other naturally occurring isotopes of strontium are non-radiogenic and include ^{84}Sr (~0.56%), ^{86}Sr (~9.87%) and ^{88}Sr (~82.5%). Strontium isotope ratios in the Earth's crust vary with the age and type of rock. Geologists have employed this principle for some time to measure the strontium isotope composition of bedrock and to determine the ages of various formations through the proportion of ^{87}Rb that has decayed (Fullagar *et al*. 1971; Faure and Powell 1972; Faure 1986).

Variation in strontium isotope compositions in natural materials is conventionally expressed as a 'strontium isotope ratio' ($^{87}Sr/^{86}Sr$), which varies among geological terrains as a function of the relative abundances of rubidium and strontium and the age of the rocks. Ratios of $^{87}Sr/^{86}Sr$ generally vary between 0.700 and 0.750. Geological units that are very old (> 100 mya) and had very high original Rb/Sr ratios will have very high $^{87}Sr/^{86}Sr$ ratios. In contrast, rocks that are geologically young (< 1–10 mya) and that have low Rb/Sr ratios, such as late Cenozoic volcanic fields, generally have $^{87}Sr/^{86}Sr$ ratios less than 0.706 (see, e.g., Rogers and Hawkesworth 1989). Rocks that had very low initial Rb/Sr ratios, such as basalt, can have $^{87}Sr/^{86}Sr$ ratios less than 0.704. These variations may seem small, but they are exceptionally large from an instrumental standpoint and far in excess of analytical error.

Strontium in bedrock moves into soil- and groundwater and into the food chain (Sillen and Kavanagh 1982; Price 1989a). Strontium concentrations and isotope ratios in rock, groundwater, soil, plants and animals thus depend in part on local geology (Dasch 1969; Hurst and Davis 1981; Graustein 1989). Although local levels of elemental strontium in plant and animal tissue vary due to many factors (Burton and Wright 1995; Burton *et al*. 1999), the isotopic composition of strontium is not changed (fractionated) by biological processes, because of the very small relative mass differences of the strontium isotopes (Faure 1986; Blum *et al*. 2000). The strontium isotope compositions of bones and teeth thus match those of the diets of the individuals, which in turn are *assumed* to reflect the strontium isotope composition of the local geology.

Measured in human bones and teeth, these ratios thus can serve as tracers of the geology of the areas in which individuals grew up and in which they died, respectively, because consumed strontium is incorporated into the skeleton during bone formation and remodelling. Bone undergoes continual replacement of its inorganic phase (Jowsey 1971), so that measurements of bone strontium reflect the later years of the life of the individual. Tooth enamel, on the other hand, forms during childhood and undergoes relatively little change (Hillson 1997). Differences in strontium isotope ratios between bone and tooth enamel in a single individual thus reflect changes in the residence history of that person.

Results from the past decade of research into the composition of skeletal tissues have amply demonstrated both the possibility and the practicality of using strontium isotopes to identify immigrants at archaeological sites. Sealy examined the origins of recent and prehistoric residents of the Cape region of South Africa (Sealy *et al*. 1991, 1995; Cox and Sealy 1997). Price *et al*. (1994a) and Ezzo *et al*. (1997) investigated strontium isotopes at Grasshopper Pueblo in north-central Arizona and found very high rates of migration in this population. Price *et al*. (1994b, 1998; Grupe *et al*. 1997) studied migration in Bell Beaker populations in southern Germany and reported rates of migration of at least 25% among these groups. Sillen and Sealy (1995)

and Sillen *et al*. (1998) employed strontium isotopes to examine diet and ecozone use in early fossil hominids in South Africa. Price *et al*. (2000) demonstrated high rates of immigration at the Classic period Mexican city of Teotihuacan and identified migrant burials in foreign enclaves within the city. Price *et al*. (2001) documented high rates of migration in the Linearbandkeramik culture, the first farmers of Central Europe.

PROBLEMS IN STRONTIUM ISOTOPE ANALYSIS

These studies have provided new and important results. However, two problems arise in such investigations: (1) levels of strontium isotope ratios in bedrock, soil and water within a local area vary, so that a single, representative ratio is difficult to ascertain; and (2) there is a range of values in human bone and enamel isotope ratios, making it difficult to distinguish some migrants from locals. The remainder of this paper is essentially a discussion of these two issues and a proposal for solution.

Variation in local strontium isotopes

Over the past 25 years, geologists have measured the strontium isotope composition of bedrock in many areas of the world (Fullagar *et al*. 1971; Faure 1986). Ecologists and archaeologists have used these values as a baseline for the study of animal and human migration, assuming that bedrock values are reiterated through the food chain. However, it is now clear that local strontium isotope values in the food chain cannot be taken directly from known values for bedrock geology.

The direct use of strontium isotopes from bedrock geology is confounded by several factors. A useful distinction can be made between geological substrate strontium and biologically available strontium. In the laboratory, experiments have shown that plants take up the $^{87}Sr/^{86}Sr$ ratios of soil in which they are planted (Hurst and Davis 1981), but in the real world this may not always be true. Isotopic ratios in the local environment are composed of a mixture of strontium derived from both atmospheric sources and mineral weathering (Miller *et al*. 1993). For example, atmospheric contributions are a major source of lead in modern soils, and the same may be true for Sr in some areas of the world. Chadwick *et al*. (1999, 494) found that in highly weathered geological environments, such as the oldest islands of the Hawaiian chain, at least as much strontium may be delivered to plants by rainwater as from the weathering of local rocks. Another study in the Sangre de Cristo Mountains of New Mexico revealed that 50–75% of the strontium in local vegetation derived from atmospheric deposition (Graustein and Armstrong 1983; Miller *et al*. 1993). However, the impacts of such atmospheric contributions are probably minimal in most areas, and particularly in much of prehistory.

In addition, various minerals within a single rock can have enormous variability in their $^{87}Sr/^{86}Sr$ ratios. For example, a rock such as granite can have two feldspars with radically differing $^{87}Sr/^{86}Sr$. Plagioclase feldspar contains most of the calcium, and hence the strontium, and has very low rubidium; hence it has a low $^{87}Sr/^{86}Sr$ close to 0.70. Conversely, potassic feldspars such as microcline or orthoclase, which are the most abundant minerals in granite, have high levels of rubidium and low levels of strontium. Their $^{87}Sr/^{86}Sr$ can substantially exceed unity (Fullagar *et al*. 1971). A single rock type can consequently vary in its $^{87}Sr/^{86}Sr$ due to different proportions of these minerals. For example, Fullagar *et al*. (1971) reported the values for Salisbury Pluton whole-rock $^{87}Sr/^{86}Sr$ as ranging from 0.7111 to 1.1970. Moreover, in the general case, these minerals will not weather at the same rate.

Figure 1 *Strontium isotope values for plants (open triangle) and available soil (solid triangle) on different rock types in the Sterkfontein Valley in South Africa (Sillen et al. 1998).*

Local soils can exhibit a range of $^{87}Sr/^{86}Sr$ values, rather than any precise whole-rock average, depending on the differential weathering of minerals and the mixing of various sources of sediment in the soil. Alluvial soils in particular tend to exhibit an $^{87}Sr/^{86}Sr$ value that averages source materials. Regardless of the mean $^{87}Sr/^{86}Sr$ for local bedrock, groundwater values can be towards either end of the available range, while the residual materials retain $^{87}Sr/^{86}Sr$ towards the other end of the range. In addition, groundwater may incorporate deeper, older waters that alter its baseline $^{87}Sr/^{86}Sr$ value (Jørgensen *et al*. 1999*)*.

Examination of $^{87}Sr/^{86}Sr$ relationships among rock, soil, plants and water by Sillen and Sealy (1995) and Sillen *et al*. (1998) has demonstrated such local variability in strontium isotope levels. Samples of soil (14), rock (five), plants (48) and water (one) were collected within a 15 km radius around the early hominid site of Swartkrans in the Sterkfontein Valley in South Africa. The geology of this area is diverse and includes dolomite, shale, andesite/basalt, quartzite/greywacke, mafics and granite. The strontium isotope values from this study for plants and whole soil on different rock types in this area are shown in Figure 1.

The Sillen *et al*. study demonstrates that: (1) whole soils in the area are highly variable; (2) plants are less variable across the region, but distinct from whole soil and bedrock values; (3) available strontium in the soil is closely correlated with plant values, regardless of substrate values; and (4) the surface water in the area has a lower strontium isotope ratio than surrounding plants and soils. A distinction was observed between vegetation along the stream in the area and the non-riparian veld zone. Plants along the stream reflected $^{87}Sr/^{86}Sr$ values of the stream waters derived from the local dolomite, while plants in the drier veld away from the stream more closely reflected available soil strontium. A small sample of three fossil mice (*Mystromis*

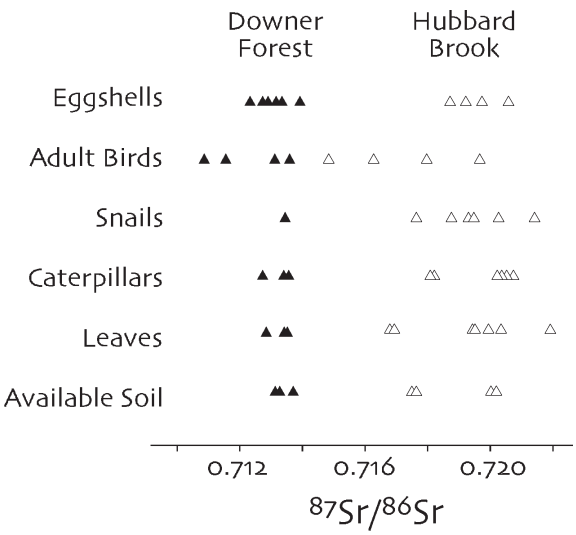

Figure 2 *$^{87}Sr/^{86}Sr$ values from available soil, plants, snails, caterpillars, birds and eggshells from two forest localities in the northeastern USA (from Blum et al. 2000).*

albicaudatus) averaged 0.71982 ± 0.0022, which is indistinguishable from the local stream water and riparian plants from the environment in which they lived.

Another study, by Blum *et al*. (2000), examined changes in Sr/Ca, Ba/Ca and $^{87}Sr/^{86}Sr$ between trophic levels in two different forest ecosystems in the northeastern USA. The available soil exchange pool, plants, caterpillars, snails and birds were measured in this study. $^{87}Sr/^{86}Sr$ values did not change through this food chain (Fig. 2) with the exception of the migratory fowl, which reflected a mix of values from summer and winter homes. Also of interest in this study are the differences between the two study areas. The Downer Forest area is very homogeneous and $^{87}Sr/^{86}Sr$ values are consistent through the food chain. The Hubbard Brook area is heterogeneous; two distinct $^{87}Sr/^{86}Sr$ values are observed in the available soil strontium and these differences are reflected in the food chain (Fig. 2). The clear message here is that local variation is just that, variable from place to place.

Our own investigations at the site of Grasshopper in east-central Arizona document similar patterns. A study of Grasshopper Pueblo, a fourteenth century AD pueblo in east-central Arizona, provides an example. Grasshopper Pueblo was settled during a period of drought in the American Southwest and abandoned approximately 125 years later. Grasshopper was a large 500-room pueblo, arranged in three major blocks, each constructed at about the same time at the beginning of the occupation: there were three rectangular plazas associated with each of the three blocks, ten small outlying blocks of rooms and a number of smaller habitation units (Reid and Whittlesey 1999). Almost 700 burials were discovered beneath room floors, in the plazas and away from residential areas.

The local geology of the local site is diverse and $^{87}Sr/^{86}Sr$ values vary widely. Samples of rock and soil taken in the immediate vicinity of the site show a range of values from 0.70893 to 0.71627 (Table 1). Grasshopper sits near the boundary of sandstone and limestone formations, and both were measured for this study. In contrast, variability in human and animal bone was less by several orders of magnitude. Samples of modern mice from agricultural fields adjacent to the archaeological site averaged 0.71000 ± 0.00031, while 16 samples of human bone from

Table 1 *Rock and soil $^{87}Sr/^{86}Sr$ data from the Grasshopper area, east-central Arizona (Ezzo et al. 1997)*

Sample	*Analyte*	*$^{87}Sr/^{86}Sr$*
Rock	Naco Limestone	0.70893
Rock	Supai Limestone	0.71627
Rock	Sandstone (Vosberg Mesa)	0.71018
Sediment	Local soil (Vosberg Mesa)	0.71523

Grasshopper had an average $^{87}Sr/^{86}Sr$ value of 0.71018 ± 0.0005 (see Table 4), close to the value for the Vosberg Mesa sandstone. The mean value for 69 enamel samples was 0.71111 ± 0.0014.

Biologically available strontium

The moral of these studies seems obvious: biologically available strontium isotope ratios can differ substantially between bedrock and other environmental values. As Sillen *et al*. (1998, 2466) noted, 'The large difference in strontium isotope composition between plant and available Sr on the one hand, and whole soil Sr, on the other, suggests that potential applications of $^{87}Sr/^{86}Sr$ relationships should use biologically available strontium as a starting point, rather than substrate geology *per se*.' The question then becomes how best to measure this biologically available strontium value. The solution of this problem is possible through the study of animal populations.

Although there can be significant $^{87}Sr/^{86}Sr$ heterogeneity in rocks, soils and plants in a given local area, animal skeletal tissues often display a remarkable homogeneity in these strontium isotope values. There are a number of studies of different species that document this pattern. Table 2 lists some of these by location, species, analyte, number of samples and the mean $^{87}Sr/^{86}Sr$, standard deviation and coefficient of variation, along with published references. The first seven studies were undertaken by the Laboratory for Archaeological Chemistry in Madison. The Hubbard Brook project by Blum *et al*. (2000) has been discussed above. Salmon strontium isotope ratios were measured as part of a study of lifetime migration (Koch *et al*. 1992). Studies of elephant and rhino bone, tusk and horn were undertaken in an attempt to determine the original homes of illegally poached animals in South Africa (van der Merwe *et al*. 1990; Vogel *et al*. 1990; Lee-Thorp *et al*. 1992, reported in Hall 1994; Hall-Martin *et al*. 1993).

Although sample sizes are sometimes less than ideal, the very low standard deviation and coefficient of variation exhibited by these examples is remarkable, and documents the generally low variability in $^{87}Sr/^{86}Sr$ values in animal populations, in spite of potentially varied geological sources. A slight increase in variability can be seen in the table with increasing size of animal and home range. Animals that obtain their foods from a larger and more geologically varied region can be expected to exhibit greater variability in $^{87}Sr/^{86}Sr$ values. This homogeneity of $^{87}Sr/^{86}Sr$ in local animal skeletal materials provides a solution to the problem of assessing biologically available strontium isotope levels.

The homogeneity of bone can be further demonstrated using a large data set of strontium and calcium values in animal bone. Our own unpublished study of strontium levels in an ecosystem in northern Wisconsin reveals that bone shows a robust Sr/Ca average that is less variable than

Table 2 *The mean, standard deviation (s.d.) and coefficient of variation (c.v.) of $^{87}Sr/^{86}Sr$ levels in natural populations. The selected locations represent a limited geographical area and a sample size (n) greater than five*

*Location**	*Species*	*Material*	*N*	*Mean $^{87}Sr/^{86}Sr$*	*s.d. $^{87}Sr/^{86}Sr$*	*c.v. (%)*	*Reference*
Teotihuacan, MX	Rabbit	Bone	8	0.70463	0.00005	0.0071	Price *et al*. (2000)
Cahokia, IL	Squirrel	Bone and enamel	5	0.70925	0.00012	0.0169	Unpublished
Aztalan, WI	Rabbit	Bone	5	0.70922	0.00004	0.0056	Unpublished
Vermont, WI	Deer	Bone	12	0.71029	0.00022	0.0310	Unpublished
Oneida, WI	Deer	Bone	6	0.71295	0.00041	0.0575	Unpublished
Grasshopper, AZ	Mouse	Bone and enamel	10	0.71000	0.00031	0.0437	Ezzo *et al*. (1997)
Dillingen, D	Snail	Shell	5	0.70838	0.00027	0.3812	Unpublished
Hubbard Brook, NH	Snail	Shell	6	0.71923	0.00134	0.1863	Blum *et al*. (2000)
Hatchery, ME	Salmon	Bone	5	0.71982	0.0009	0.1250	Koch *et al*. (1992)
Hatchery, OR	Salmon	Bone	5	0.70919	0.00010	0.0141	Koch *et al*. (1992)
Addo Park, SA	Elephant	Bone	6	0.71153	0.00008	0.0112	Vogel *et al*. (1990)
Namibian Desert, SA	Elephant	Bone	6	0.72380	0.0013	0.1796	Vogel *et al*. (1990)
Etosha Park, SA	Rhino	Horn	8	0.71837	0.00306	0.4260	Hall-Martin *et al*. (1993)
Addo Park, SA	Rhino	Horn	7	0.71340	0.00212	0.2972	Hall-Martin *et al*. (1993)
Pilanesberg, SA	Rhino	Horn	7	0.70675	0.00389	0.5504	Hall-Martin *et al*. (1993)
Umfolozi, SA	Rhino	Horn	12	0.71693	0.00096	0.1339	Hall-Martin *et al*. (1993)
Mkuze, SA	Rhino	Horn	16	0.71161	0.00112	0.1574	Hall-Martin *et al*. (1993)
Hluhluwe, SA	Rhino	Horn	8	0.71511	0.00208	0.2909	Hall-Martin *et al*. (1993)

* Location abbreviations include the following: MX = Mexico, IL = Illinois, WI = Wisconsin, AZ = Arizona, D = Germany, NH = New Hampshire, SA = South Africa, ME = Maine, OR = Oregon.

Table 3 *Strontium/calcium ratios in soils, plants, and animals from northern Wisconsin*

	Mean	*s.d.*	*n*	*Coefficient of variation*	*Range*
Soils	0.018615	0.026983	47	1.449535	0.099398
Plants	0.003768	0.001541	864	0.409010	0.009573
Deer	0.000889	0.000200	59	0.224992	0.000897
Marten	0.000379	0.000075	51	0.197435	0.000355
Bobcat	0.000270	0.000048	25	0.178234	0.000211

Table 4 *$^{87}Sr/^{86}Sr$ for animal and human bone from archaeological sites. The key to locations is the same as in Table 2. Values in parentheses are sample sizes (n)*

Location	*Species*	*Animal mean ± s.d. $^{87}Sr/^{86}Sr$*	*Human mean ± s.d. $^{87}Sr/^{86}Sr$*
Grasshopper, AZ	Mouse	0.71000 ± 0.00031 (10)	0.71018 ± 0.0005 (16)
Dillingen, D	Snail	0.70838 ± 0.00027 (5)	0.70867 ± 0.0001 (7)
Teotihuacan, MX	Rabbit	0.70463 ± 0.00005 (8)	0.70501 ± 0.0008 (57)
Aztalan, WI	Rabbit	0.70922 ± 0.00004 (5)	0.701021 ± 0.0004 (21)

Sr/Ca in the associated soils by several orders of magnitude (Table 3). If bones represent merely a random sample of the local strontium, we would expect not only strontium isotopes but also strontium elemental levels to be as variable as the plants and underlying soils. On the other hand, if bones representatively sample strontium from enough sources to reflect some regional average, then we would expect to see a significant reduction in the variability of strontium levels and isotope ratios in the bones compared to the plants and soils. Analysis of more than a thousand samples of soils, waters, plants and bones shows that Sr/Ca ratios for soils in the region have a range of 0.09940, those for plants growing on these soils have a range of 0.00957, herbivore bones show a range of 0.00090, and carnivore bones have a miniscule range of only 0.00021(cat)/0.00036(marten)—a compression of several orders of magnitude (Table 3).

Although biopurification itself reduces the magnitude of Sr/Ca, and hence its range, with each increase in trophic level, the magnitude-adjusted coefficient of variation also shows a reduction in Sr/Ca variability from 145% in soils to 18% (cat) to 20% (marten) in carnivores.

Bone formation acts as a powerful averaging mechanism for local $^{87}Sr/^{86}Sr$ variability. In recognition of the fact that bone $^{87}Sr/^{86}Sr$ reflects the assimilation of locally available $^{87}Sr/^{86}Sr$ over time, as the tissue is formed and remodelled over time and over a range of available foodstuffs, it is not surprising that bone should provide a summary measure of the local $^{87}Sr/^{86}Sr$ ratio. Thus it is clear that animal skeletal tissue can provide a good estimate of biologically available strontium isotope ratios for a local area, averaging variability of that area into a summary value. The next important question concerns the relationship between animal skeletal tissue and human remains in terms of $^{87}Sr/^{86}Sr$ values.

Studies in the Laboratory for Archaeological Chemistry have generally involved the use of small animals as indicators of local strontium isotope levels (Table 4). Plants are not used

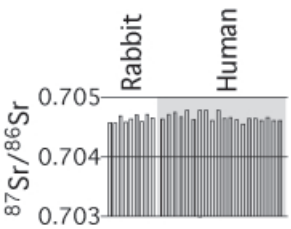

Figure 3 *Strontium isotope ratios in the bones of rabbits and humans from the ancient city of Teotihuacan (Price* et al. *2000).*

because, as we have seen, they are much more variable than animal bone in a given area. We have collected various species, including mice, guinea pigs, rabbits, squirrels and snails, from areas around prehistoric sites and cemeteries investigated with strontium isotopes. Small mammals and snails are relatively easy to obtain and have limited home ranges. These herbivores eat a mix of plant materials from their local area. The strontium isotope ratio in their diet is averaged over their lifetimes in their bones. These modern samples thus provide an estimate of the local strontium isotope levels available to humans as well.

Values for animal tissue and human bone in four archaeological studies are shown in Table 4. These samples come from both archaeological and modern contexts. The mice from Grasshopper and the snails from Dilligen are modern. One-half of the rabbits from Teotihuacan are modern and the remaining half are prehistoric; no differences were observed between the modern and ancient samples. The squirrels from Aztalan are prehistoric. These four sites are in diverse geological environments and of varying ages, ranging from 7000 to 600 years ago. In two of these examples, Grasshopper and Dillingen, the animal values are statistically identical to the human values. In the other two cases, Teotihuacan and Aztalan, the values for animals and humans are very close, and the differences probably reflect human foods coming from a wider range of geologies than the animal diet.

In the case of Teotihuacan, for example, the human values were measured on 57 individuals, including a large proportion that migrated into this ancient city (Price *et al*. 2000). Comparison of the rabbit $^{87}Sr/^{86}Sr$ values with non-migratory humans from two areas of burial at Teotihuacan (Cueva del Pirul and Cueva de las Varillas) documents the very close relationship between the rabbits and the native human inhabitants of the city. Figure 3 shows strontium isotope ratios in the bones of rabbits and humans from two localities in the city. The rabbit and indigenous human bone are essentially identical.

The results of these studies demonstrate that small animal remains provide (1) a homogeneous signal of local strontium levels and (2) a strong correspondence with the indigenous prehistoric humans. It is clear from the above that animal bone provides a good proxy for measuring the strontium isotope ratios that are biologically available to the local human population. Measurement of small animal skeletal tissue should be done in conjunction with studies of prehistoric humans.

What species? Several aspects of this procedure, however, remain to be resolved. One question concerns the species of animal that it is best to use to establish the biologically available strontium values. There is no simple answer to this question, since different species will be present in different regions and will have varying home ranges. Our research has focused on small animals, since they are easier to collect in some number. However, smaller animals have smaller

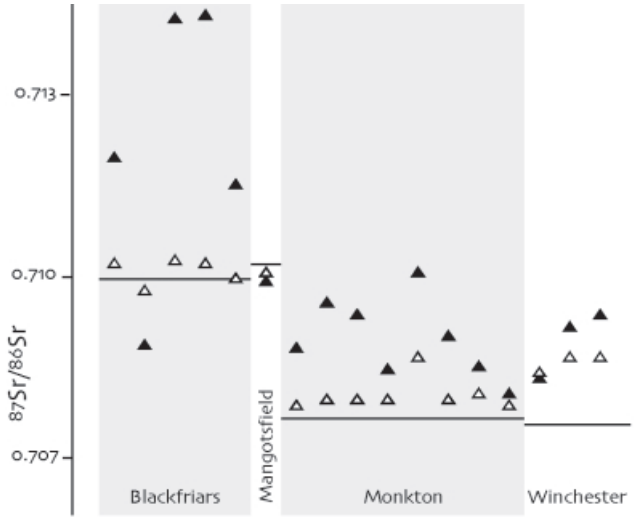

Figure 4 *Available soil (line), enamel (solid triangle) and dentine (open triangle) $^{87}Sr/^{86}Sr$ values from four archaeological sites in England (Budd* et al. *2000). Note that the dentine values are much closer to those of the soil.*

home ranges, and may not incorporate all sources of strontium in the area of a given site. Larger animals utilize larger areas, of course, but modern specimens are often difficult to obtain in quantity. We have collected deer, mice, guinea pigs, squirrels, rabbits, snails and other species in our studies. Certainly, the snails were easy to catch and clearly incorporate locally available strontium. Snails collected from a substantial radius around the site of interest should reflect the range of strontium isotope ratios present in the area.

Modern or fossil animals? A second question concerns whether modern or fossil animals are more appropriate for establishing the biologically available level of $^{87}Sr/^{86}Sr$. Several factors must be considered in answering this question. Modern animals may differ from local sources of strontium isotopes for several reasons: consumption of imported foods, or pollution from fertiliser or airborne sources of strontium. Domestic animals today, and modern humans, eat foodstuffs that originate in a variety of different geologies, making measurement of strontium isotope ratios irrelevant (Åberg *et al*. 1998). Pollution is another source of contrasting strontium isotopes in a local area. Comparison of strontium isotope ratios in greenhouse plants and outdoor species suggested that fly ash from coal-fired energy plants contributed substantially to the soil strontium pool in the UK (Hurst and Davis 1981; Straughn *et al*. 1981). In some areas, agricultural fertilisers can make a significant Sr contribution to local groundwater. For example, on the coastal plain in Maryland, Early Tertiary marine carbonates have a relatively low $^{87}Sr/^{86}Sr$ value of about 0.708. However, fertilisers with relatively high values (~0.715) have raised the $^{87}Sr/^{86}Sr$ values of groundwater to levels between 0.713 and 0.715 (Böhlke and Horan 2000). The effect is primarily dependent on the age of the groundwater; deeper and older (35 or more years since recharge) groundwater has $^{87}Sr/^{86}Sr$ values between 0.708 and 0.710, more consistent with the marine sedimentary basement (Böhlke and Horan 2000, Fig. 4).

Fossil animal bone at archaeological sites is more generally subject to the vagaries of contamination or diagenesis. There is an enormous literature on this subject, and it is clear that bone often is subject to contamination by a variety of elements, including strontium (Lambert

et al. 1985; Nelson *et al*. 1986; Price 1989b; Sillen 1989; Sealy *et al*. 1991; Wang and Cerling 1994; Nielsen-Marsh and Hedges 2000a). A number of techniques, some more successful than others, have been developed to remove these contaminants (see, e.g., Price *et al*. 1992; Sillen and Sealy 1995; Koch *et al*. 1997; Nielsen-Marsh and Hedges 2000b). More important for the application of strontium isotope analysis, however, is the nature of this contamination in terms of isotopic ratios. Contamination by strontium isotopes will not change the local value of the bone, because the 'contamination' is normally the same biologically available solution in the food chain (Grupe *et al*. 1997).

This issue of using modern or fossil animal tissue may best be resolved by consideration of a final question concerning the specific tissue to be analysed for biologically available strontium. In this context, the question is whether to use tooth enamel or bone.

Enamel or bone? As noted, bone is subject to substantial diagenesis and post-mortem change. On the other hand, tooth enamel—a denser, harder and more inert substance than bone—is much less susceptible to diagenesis (see, e.g., Dreissens and Verbeeck 1990; Kolodny *et al*. 1996; Hillson 1997; Sharp *et al*. 2000). The higher organic content and reduced crystal size in dentine increase its susceptibility to contamination (Kohn *et al*. 1999). There is some suggestion of *in vivo* uptake of strontium on the surface of enamel (Grupe *et al*. 1999; Hörn and Müller-Söhnius 1999), but this has not been identified analytically (Brudevold and Söremark 1967; Montgomery *et al*. 1999). A number of studies have documented the more resistant nature of enamel to post-mortem chemical and physical changes. Boecherens *et al*. (1994), in a study of fossil dinosaur teeth, have demonstrated that enamel was more resistant than dentine to trace element contamination, even in material more than 100 million years old.

Budd *et al*. (2000) measured strontium abundance and $^{87}Sr/^{86}Sr$ ratios in 14 prehistoric and medieval human teeth from different individuals from four sites in the UK. They compared values in available soil, dentine and enamel to examine the integrity of the original biogenic material. The study assumed that dentine, as a more porous material, would behave more like bone in terms of contamination. Dentine is different from bone, because it does not undergo remodelling during the lifetime of an individual (Hillson 1997). The Budd *et al*. study documented significant differences between enamel and dentine in most instances, and minor differences between the dentine and soil (Fig. 4). The dentine values were substantially closer to those for the soil than the enamel values. The authors argued that strontium abundance in the majority of enamel samples was similar to modern, unburied values. The study concludes that enamel is less susceptible to diagenesis than dentine and, by extension, bone.

Kohn *et al*. (1999) have undertaken perhaps the most detailed study of diagenesis in fossil teeth to date, using the latest instrumentation to examine elemental abundances in 4 mya fossil and modern animal tooth enamel from East Africa at a micrometre to sub-micrometre scale. Their study documented physical contamination of enamel by a number of elements (Fe, Mn, Si, Al, Ba and possibly Cu) and chemical alteration of U, REE, F and 'possibly' Sr. Isotopic ratios were not measured. Concentrations of secondary minerals ranged from 0.3% in enamel to *c*. 5% in dentine, documenting the more resistant nature of enamel and very low levels of contamination. Evidence for strontium contamination in enamel was equivocal; no significant differences could be observed between modern and fossil specimens. Unfortunately, this study compared samples from separate areas in East Africa where the natural abundance and availability of strontium may have differed. Variation in biologically available strontium, barium and other elements incorporated into the skeleton would mean that comparison of different areas was futile without knowledge of baseline local levels. Nevertheless, the fact that strontium

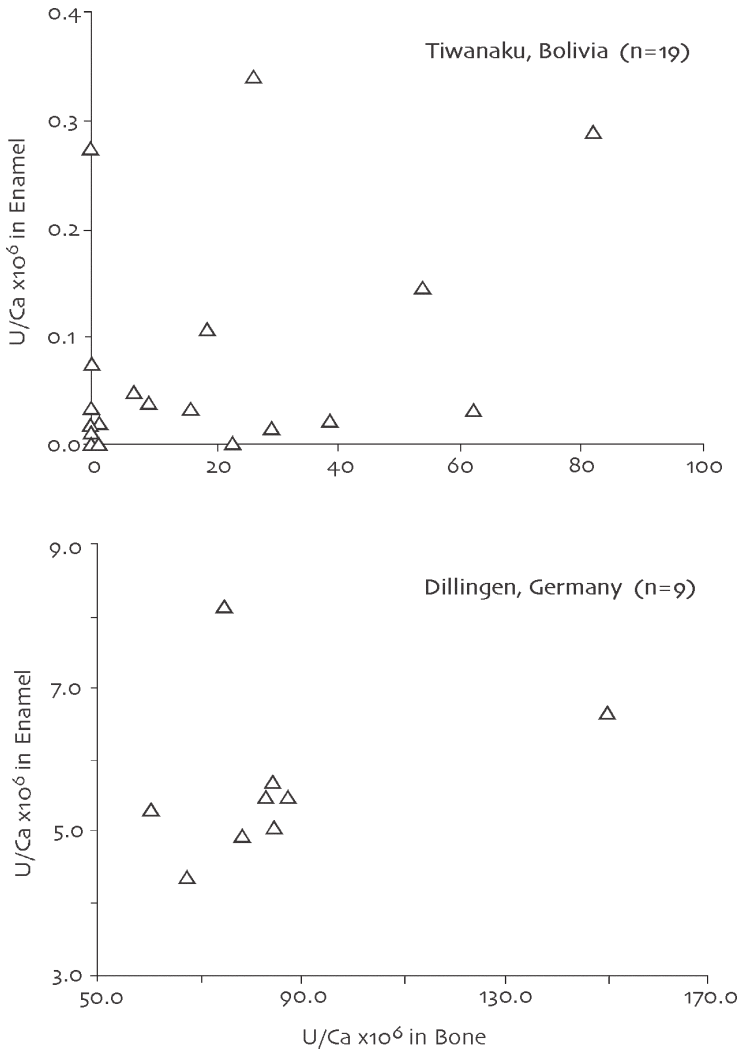

Figure 5 *Uranium levels (U/Ca × 10^8) in archaeological bone versus tooth enamel samples from two prehistoric sites in Bolivia and Germany. Each paired bone/enamel sample is from a single individual. Note that levels in enamel are generally very low and several orders of magnitude lower than in bone.*

cannot clearly be shown to have contaminated fossil enamel supports an argument for the use of this material.

Kohn *et al*. (1999) also suggest that measurement of uranium and the rare earth elements in archaeological enamel can provide some indication of the degree of contamination present. Such data can also be used to document the susceptibility of bone to diagenesis, compared to enamel. We have measured uranium levels in samples of human bone and tooth enamel in two prehistoric contexts (Fig. 5): Linearbandkeramik burials from 7000 BP in Germany and Tiwanaku period burials from Bolivia, dating from approximately 1000–1500 BP. Uranium levels on the graph are reported as U/Ca × 10^6. In both cases, uranium levels (an indicator of contamination)

are lower in enamel by between two and three orders of magnitude, and are either absent or very low in absolute abundance.

DISTINGUISHING LOCAL AND MIGRANT INDIVIDUALS

A second problem that has arisen in strontium isotope studies of prehistoric human migration involves distinguishing local and migrant individuals. In an ideal situation, the enamel values of the migrants should be completely different from the bone and enamel values of indigenous persons. In reality, however, this distinction is not always clear. A range of isotope ratio values for both bone and tooth enamel is often observed in such studies. Extreme values are not a problem, but there is no objective criterion for distinguishing among individuals with values close to the local range.

An example of this range of variation in a human population can be seen in the Grasshopper study. Figure 6 shows the distribution of $^{87}Sr/^{86}Sr$ values at Grasshopper. There are a total of 85 $^{87}Sr/^{86}Sr$ measurements on 16 bone (black bar) and 69 tooth enamel (white bar) samples. Bone and tooth samples from the same individual are paired in the graph. The mean value for all the samples is 0.71094, s.d. = 0.0014 and the range is 0.70869–0.71748. It is clear from the graph that the distinction between migrants and locals is not always obvious. Extreme values are certainly migrants, but what about the individuals with values closer to the centre of the distribution? How do we objectively determine which individuals are local and which are outsiders?

To resolve this problem, it is useful to consider the sources of variation in $^{87}Sr/^{86}Sr$ in prehistoric human skeletal remains in addition to residential change from one geological region to another. Importantly, the inherent natural variation in $^{87}Sr/^{86}Sr$ levels in a human population is unknown, although—on the basis of comparison with animal populations—it is presumed to be low. Several factors introduce additional variability into this system, especially diet, length of residence and multiple movement.

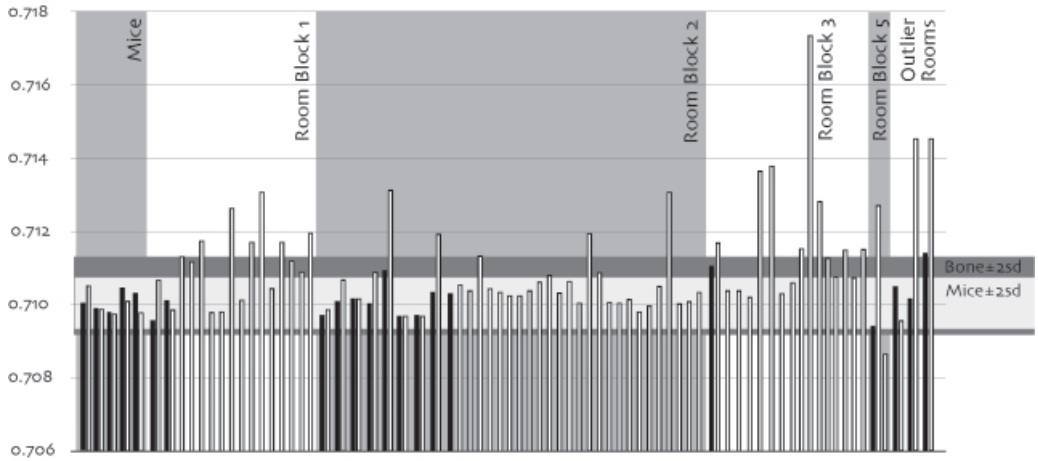

Figure 6 *Strontium isotope values for human bone (solid bar) and tooth enamel (open bar) at Grasshopper Pueblo, Arizona (Price* et al. *1994b; Ezzo* et al. *1997). The confidence limit of ± 2 s.d. for the mice is shown, as is a band based on human bone values.*

Diet is the proximate source of strontium that is incorporated into bone. Human diets are omnivorous, and include a number of species of plants and animals with greatly varying home ranges. Depending on the local geology, such foods may come from one or more $^{87}Sr/^{86}Sr$ sources. Wide-ranging edible species such as large herbivores may incorporate strontium from several sources into their diet and tissue. Species of edible plants may also range over several $^{87}Sr/^{86}Sr$ sources. Depending on the catchment of foodstuffs for the human population, several $^{87}Sr/^{86}Sr$ sources may be included in the dietary intake of strontium. Variation in individual diets will result in some variation in $^{87}Sr/^{86}Sr$ levels in bone.

Length of residence is also a contributing factor in variation in bone $^{87}Sr/^{86}Sr$ values. As noted previously, bone is remodelled throughout life. The rate of turnover in bone depends on the specific kind of bone and part of the skeleton involved. While dental enamel undergoes virtually no change after it is formed, bone tissues themselves remodel at different rates depending upon the ratio of active osteoclasts (responsible for hydroxyapatite precipitation) and osteoblasts (responsible for dissolution).

In general, trabecular bone remodels rapidly, while dense cortical tissue remodels more slowly (Simmons and Grynaps 1989; Mulhern and Van Gerven 1997; Teitelbaum 2000). Cortical bones, such as the diaphyses of the femur and tibia, remodel over a period of decades, while bones with abundant trabecular tissue, such as ribs and the iliac crest, can remodel over a few years (Jowsey 1961; Jowsey *et al*. 1965; Parfitt 1983; Eriksen 1986; Hill 1998). The turnover rates for several different skeletal tissues are modelled in Figure 7. Note that the turnover rate in the trabecular bone of the iliac crest is much faster than in the cortical bone of the mid-shaft femur. Thus, the $^{87}Sr/^{86}Sr$ value in the bones of a migrant individual shifts towards the local value over time, depending on the type of bone analysed. The longer a migrant individual has been in residence, the closer bone values will be to local values. Different periods of residence among the migrant individuals will introduce some variation in bone values in a population.

A third source of variability in bone $^{87}Sr/^{86}Sr$ values may arise from multiple residence by some individuals. The basic assumption in these studies has been that a migrant individual moves from one place to another—from one strontium isotope ratio geology to another—once during his or her lifetime. It is entirely possible, even likely, in the context of migration that some individuals may have had multiple residences during their lifetimes. The bones of these

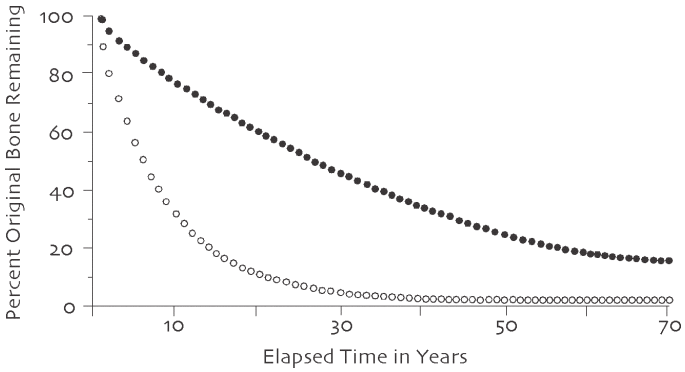

Figure 7 *Estimated bone turnover as a percentage of original bone in a normal 30-year-old adult in different skeletal tissues (data from Jowsey 1961; Jowsey* et al. *1965; Parfitt 1983; Eriksen 1986; Hill 1998). The solid dot is the mid-shaft femur; the open dot is the anterior iliac crest.*

individuals would combine the source $^{87}Sr/^{86}Sr$ levels of the different areas of residence and thus exhibit greater variation.

The moral of this story is that there is variation in $^{87}Sr/^{86}Sr$ in prehistoric human populations, beyond that introduced by migrant individuals, and we do not know exactly how much variation exists. For this reason, an objective criterion is needed to distinguish between local and migrant individuals in archaeological data.

We return to the Grasshopper study to demonstrate how migrants were identified (Price *et al*. 1994b; Ezzo *et al*. 1997). Because of the range in values present in the samples at Grasshopper (Fig. 6), two different criteria were employed in the original study to distinguish between migrants and locals: (1) a cut-off value based on the human bone values and (2) a cut-off value derived from the $^{87}Sr/^{86}Sr$ ratios in field mice. Two horizontal bands are shown in Figure 6, indicating the mean ± two standard deviations for (1) human bone and (2) mice. The ten mice samples have a mean of 0.71000 ± 0.00031, and the range for ± 2 s.d. is 0.70938–0.71062. The 16 human bone samples have a mean of 0.71018 ± 0.0005, and the range for ± 2 s.d. is 0.70918–0.71118. Using the mouse criterion, 30 of the 69 individuals in the study are determined to be migrants; the human bone criterion designates 21 of 69 as outsiders. Archaeological evidence supports the higher number (Ezzo and Price 2002). In either case, between 30% (human bone criterion) and 43% (mouse bone criterion) of the human individuals analysed at Grasshopper had migrated to the site (Fig. 6).

A second example may also be informative. Dillingen is a site from the early Neolithic of Germany, dating to approximately 5200 BC. Figure 8 shows the distribution of seven bone and 16 enamel samples from the site. In this example, bone values are very homogeneous, with a mean of 0.70865 ± 0.00012. Five modern snail shells were collected from the area around the site and exhibit a mean value of 0.70838 ± 0.00027, showing slightly higher variation. Again, two bands are indicated in the figure at ± 2 s.d. for snails (0.70784–0.70892) and human bone (0.70841–0.70889). In this example, the confidence limits based on bone are narrower than those based on the snails. Using the bone criterion, nine of the 16 individuals are indicated as migrants; the snail criterion designates eight of the 16 as outsiders. In either case, it appears that at least half of the burials at Dillingen were migrants to the site.

The two examples from Grasshopper and Dillingen in this study point out that human bone and animal values often produce similar results. Use of the less heterogeneous values will result in a less conservative estimate of migrants in the sample. Nevertheless, we recommend the use of animal values from enamel or shell to estimate local biologically available $^{87}Sr/^{86}Sr$ levels,

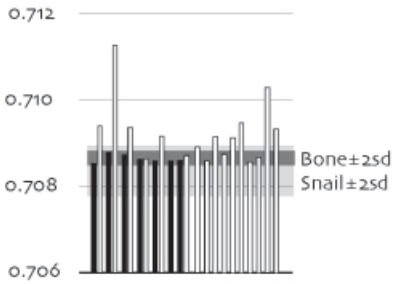

Figure 8 *Strontium isotope ratios from the Dillingen, Germany, study. Open bars are tooth enamel; solid bars are bone. Paired bone and tooth bars are from the same individual. Two bands show confidence limits at the mean ± 2 s.d. for human bone and snail shell from the area of the site.*

as human bone values will be more varied for reasons discussed above, and in most instances less reliable.

CONCLUSIONS AND RECOMMENDATIONS

The above discussion has focused on two problems in strontium isotope studies of prehistoric migration, specifically the measurement of biologically available strontium isotope ratios and the distinction between migrant and local individuals. The use of prehistoric and/or modern samples of small animals is advocated in order to establish the biologically available level and to distinguish migrant individuals. Where possible, we recommend measurement of tooth enamel in fossil animals from the archaeological site under consideration. Tooth enamel is less susceptible to diagenesis than bone. If such samples are not available, modern species can be substituted, with concern for contamination from imported foods or pollutants. Comparison of enamel in the fossil and modern specimens of the same species would provide some control for diagenesis, assuming no modern contamination. A mix of small and large animals, or samples of the same species from across the presumed prehistoric human diet catchment area, will probably provide the best estimate for the diet catchment of the human population, as well as a means for distinguishing individuals who have migrated into the local human population.

We recommend that a confidence limit for separating migrants and indigenous individuals be established using the mean of biologically available strontium isotope ratios (determined from animal samples) ± 2 s.d. While this criterion is arbitrarily selected, as discussed above, used conventionally it becomes an objective means of identifying residential change in prehistoric contexts.

Strontium isotope analysis of prehistoric human remains is an important new technique for studying migration. While there are minor problems, such as those discussed in this study, in general terms the technique is robust, exciting and useful. We look forward to forthcoming studies and more fascinating information about our mobile ancestors.

ACKNOWLEDGEMENTS

This study has involved the assistance and advice of a number of groups and individuals. We would particularly like to thank Joe Ezzo, Paul Fullagar, Clark Johnson, Linda Manzanilla, Nick van der Merwe, Bill Middleton, Peter Schröter and the White Mountain Apache. Two reviewers of the manuscript, Christine White and Christine Hastorf, provided useful advice. The National Science Foundation and the Graduate School of the University of Wisconsin-Madison have funded most of our research on strontium isotopes and prehistoric migration, and we are most grateful for their assistance.

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