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Strontium Isotope Composition of Skeletal Material Can Determine the Birth Place and Geographic Mobility of Humans and Animals

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ABSTRACT: The Sr isotope composition measured in skeletal elements (e.g., bone, teeth, or antlers) can be used to infer the geographic region that an animal or human inhabited, because different regions tend to have distinct Sr isotope compositions, and natural variations in the relative abundance of Sr isotopes are not changed as Sr is processed through the food chain. Therefore, an organism that ingests Sr from one region can have a Sr isotope composition that is different than that of an organism that ingests Sr from another region. The Sr isotope composition of skeletal elements is a reflection of the concentration-weighted average of dietary Sr that was ingested while that skeletal element was produced. Because different skeletal elements grow and exchange Sr at different stages during the life times of organisms, Sr isotope analysis of different skeletal elements can be used to infer changes in geographic location at different stages in an organism's life. The Sr isotope composition measured in human teeth will reflect the average Sr isotope composition that was ingested as a child, due to the immobile nature of Sr and Ca in teeth after formation, whereas the Sr isotope composition of bone will reflect the average isotopic composition over the last ten years of life, due to continuous biological processing of Sr and Ca in bone. Inferring the average isotopic composition of dietary Sr is best done by analyzing skeletal fragments from control groups, which might be animals that have the same feeding habits as the animal in question, or, in the case of humans, analysis of close family relatives. In cases where it is not possible to construct a Sr isotope database from control groups, it becomes necessary to estimate the isotopic composition of dietary Sr based on geologic principles. We present three case studies from our research that illustrate a range of approaches: (1) results from a criminal case where a deer was illegally harvested and the location of the deer was important to establish, (2) a pilot study of commingled human remains from a burial in Vietnam, associated with the Vietnam Conflict, and (3) a study of 13th and 14th century migration of people from an archeological site in the Southwest United States.

KEYWORDS: forensic science, forensic anthropology, geology, archaeology, strontium isotope composition, skeleton, human, animal, geographic origin, mobility

The only physical evidence in some forensic science investigations is the skeletal remains of an organism. Determination of the birth place or last residence before death of an organism or individual can be instrumental in solving such an investigation. Below we describe a technique that can place constraints on possible geographic residence of an organism through Sr isotope analysis of skeletal material. This technique is based on the fact that the Sr isotope composition of bedrock and soils are variable due to long-lived radioactive decay, and Sr isotopes are not fractionated as Sr is passed through the food chain. Plants from one geographic location have the same Sr isotope composition as the soil and rock upon which they grew. Herbivores in turn will have the same Sr isotope composition as the plants, and carnivorous animals in turn will have the same isotope composition as the herbivores that they eat. Chemically, Sr behaves like Ca, and is therefore concentrated in skeletal elements. The Sr isotope composition of these skeletal elements is a reflection of the concentration-weighted average of the Sr that was ingested. Although it might first be assumed that Ca isotope studies of skeletal elements would be the best approach, radiogenic isotope variations in ^{40}Ca abundances (produced by long-lived ^{40}K decay) are vanishingly small (1), and are on the order of natural, mass-dependent, Ca isotope variations (2).

Below we discuss the origin of Sr isotope variations in the earth, how Sr is processed through the food chain and concentrated in skeletal elements, and discuss three case examples of how the Sr isotope composition of skeletal elements may be used to elucidate geographic information.

Strontium Isotope Variations in the Earth

There are four stable isotopes of Sr: ^{88}Sr (82.53%), ^{87}Sr (7.04%), ^{86}Sr (9.87%), and ^{84}Sr (0.56%). All but ^{87}Sr are nonradiogenic (that is, not the products of radioactive decay), and ^{87}Sr is produced by beta decay of ^{87}Rb (half-life = 48.8×10^9 yrs). The ^{87}Rb $\rightarrow$ ^{87}Sr radioactive decay pair has therefore produced distinctly different ^{87}Sr abundances in different parts of the Earth over its 4.5 billion year history. To measure differences in the ^{87}Sr abundances in various rocks or other samples, ^{87}Sr abundances are typically normalized to a nonradiogenic isotope, ^{86}Sr (the choice of ^{86}Sr produces ratios near unity, which are analytically the most precise; ^{88}Sr or ^{84}Sr would be poorer choices). Using the ^{87}Sr/^{86}Sr ratio, rather than absolute ^{87}Sr abundances, removes variations in ^{87}Sr abundances that reflect natural variations in total Sr; use of the ^{87}Sr/^{86}Sr ratio allows us to isolate the variations in ^{87}Sr abundances that are solely a function of ^{87}Rb $\rightarrow$ ^{87}Sr decay. The Sr isotope composition of a sample at any time is described by the exponential radioactive decay equation

$$[^{87}Sr/^{86}Sr]_{T2} = [^{87}Sr/^{86}Sr]_{T1} + [^{87}Rb/^{86}Sr](e^{[\lambda t]} - 1) \quad (1)$$

$[^{87}Sr/^{86}Sr]_{T1}$ is defined as the ^{87}Sr/^{86}Sr ratio a sample or part of the Earth's crust had at some time in the past (such as formation of a volcanic rock, etc.), λ is the decay constant (1.42×10^{-11} yr^{-1} for

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^{87}Rb), t = T1 − T2 (in years), and $[^{87}Sr/^{86}Sr]_{T2}$ is the ratio at some young time, such as measured today in the laboratory. Due to the long half-life of ^{87}Rb, significant changes in $^{87}Sr/^{86}Sr$ ratios in the Earth generally occur only when the value T1-T2 is on the order of tens of millions of years or more. In addition, T2 does not have to be known to better than several thousand years. The variations in $^{87}Sr/^{86}Sr$ isotope ratios of different areas of the Earth observed today (T2) is a result of variations in (1) the Rb/Sr ratio (as expressed by the $^{87}Rb/^{86}Sr$ ratio), (2) the age of a sample, and to a lesser degree, (3) variations in the $[^{87}Sr/^{86}Sr]_{T1}$ of a sample (typically a rock sample of the crustal "basement").

Some examples may be useful. Consider two volcanic rocks that crystallized with the same $^{87}Sr/^{86}Sr$ ratio at the time of eruption, $[^{87}Sr/^{86}Sr]_{T1}$, and identical Rb/Sr ratios. One rock might be a sample from Hawaii that formed ten years ago, and another might be a sample from Michigan that formed two billion years ago. Clearly the equation above indicates that the $[^{87}Sr/^{86}Sr]$ ratio measured today, $[^{87}Sr/^{86}Sr]_{T2}$, will be much higher for the very old rock from Michigan. Consider a second example of two rocks that formed one billion years ago with the same initial ratio, $[^{87}Sr/^{86}Sr]_{T1}$, but one rock, such as a shale, had a very high Rb/Sr ratio as compared to the second rock, such as a limestone. Again, from the equation above, it should be clear that the shale will have a much higher measured $^{87}Sr/^{86}Sr$ ratio than that of the limestone. It is well known from over three decades of geological research that distinctive rock types have distinctive Rb/Sr ratios. For example, basaltic lavas, limestone, and marble all have very low Rb/Sr ratios, whereas sandstone, shale, and granite commonly have very high Rb/Sr ratios. Clay minerals have some of the highest Rb/Sr ratios, and therefore soils developed on shale units may develop quite distinct Sr isotope ratios relatively quickly.

The variations in Sr isotope ratios found on the surface of the Earth are a function of both the age of the crust and their bulk compositions. Relative to the analytical error of the $^{87}Sr/^{86}Sr$ measurements (±0.00001 to 0.00003), there are huge differences in the Sr isotope compositions of different parts of the Earth. In order to facilitate comparison of the numerically small differences in $^{87}Sr/^{86}Sr$ ratios, Sr isotope compositions may be presented in $\varepsilon^{87}Sr$ notation (3), which is defined as

$$\varepsilon^{87}Sr = ([^{87}Sr/^{86}Sr]_{MEASURED}/[^{87}Sr/^{86}Sr]_{BULK\ EARTH} - 1)\ 10{,}000 \quad (2)$$

where $[^{87}Sr/^{86}Sr]_{MEASURED}$ is the measured $^{87}Sr/^{86}Sr$ and $[^{87}Sr/^{86}Sr]_{BULK\ EARTH}$ is equal to 0.7045; analytical uncertainty as expressed by $\varepsilon^{87}Sr$ values are 0.2 to 0.4 $\varepsilon^{87}Sr$ units.

The geologic history of the U.S.A. (and other countries) has been very long, representing nearly the entire 4.5 billion year history of the Earth. The ages of the crust in the U.S.A. varies from less than one million years old in Hawaii to nearly four billion years old in parts of Minnesota and the Upper Peninsula of Michigan, which produce significant variations in the Sr isotope composition of different regions of the U.S.A. (Fig. 1). A model for Sr isotope variations in the continental United States can be constructed to a first order assuming that $[^{87}Sr/^{86}Sr]$ varies solely as a function of age, although a more complex model would additionally account for variations in bedrock lithology. The purpose of such a model is not to make detailed conclusions regarding the geographic origin of skeletal elements from a specific study, but to act as a general guide to expected isotopic variations in a particular region. Below we discuss different methods on how the average dietary Sr isotope composition of different regions can be determined, using Fig. 1 as a framework, but also detailed consideration of local variations in Sr isotope compositions.

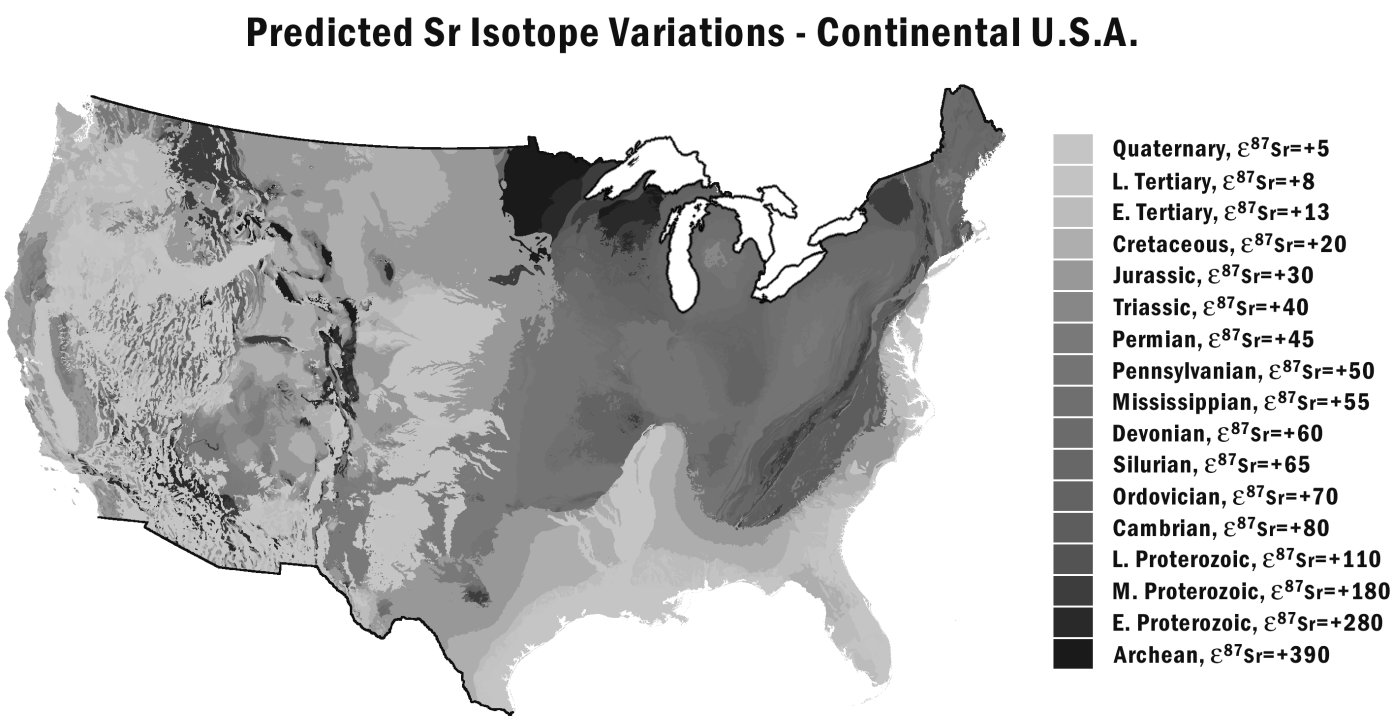

FIG. 1—*Model Sr isotope compositions of the United States showing inferred $\varepsilon^{87}Sr$ values, as calculated by age variations in basement rocks. Based on the digital geologic map by the U.S. Geological Survey (54). Sr isotope evolution equations given in text, and average crustal Rb and Sr contents (55), and an assumed initial $^{87}Sr/^{86}Sr$ of 0.705. The primary purpose of this model is to illustrate the first-order Sr isotope variations that occur in the Earth due to basement-rock ages. Additional complexities will develop due to variations in rock lithology (which changes Rb/Sr ratios), and sedimentary rocks which may contain multiple age and lithologic components.*

Tracing Sources of Sr in the Body

Because Sr is an alkaline-earth metal that is chemically similar to Ca, forensic Sr isotope studies have focused on Ca-bearing phases such as bone, teeth, or antlers. The Sr isotope composition of an organism's bones or teeth will reflect the integrated Sr isotope composition of its diet during the period of time that certain parts of its body were forming or open to chemical and isotopic exchange. Using Sr isotopes to trace the geographic origin of an organism relies critically on the fact that the $^{87}Sr/^{86}Sr$ ratio of the soil and water will be reflected in the plants, which will be passed on to a herbivore, which will be passed on to a carnivore. In the simplest case where a food chain is based on rock, soil, and water of constant $^{87}Sr/^{86}Sr$ ratio, the Sr isotope ratio of each point along the composition chain will be the same. Strontium is a high-mass element where any mass-dependent isotope fractionation would be vanishingly small during geological or biological processing. Even if mass-dependent fractionation of Sr did occur in nature, the most common method of mass spectrometric analysis of Sr would remove any such fractionation affects (see 4 or 5). The lack of Sr isotope fractionation between skeletal elements and dietary input has been proven by numerous workers for a variety of different organisms and skeletal elements, including antlers from reindeer (6), marine and freshwater fish teeth and bones (6,7), elephant, mammoth, and mastodon bone and tusk material (8–11), human tooth and bone (12), and plants (13–15).

Biological processing of Sr was extensively studied in the 1950's because of the potential for radioactive ^{90}Sr ingestion from the atmosphere, which was produced by aboveground nuclear weapons testing (16). More recently Sr/Ca ratios in bone have been used to infer paleodiets (17,18), and it is now recognized that there is a decrease in the Sr/Ca ratio of bones up the food chain because only 20 to 40% of ingested Sr is absorbed, as compared to the 40–80% of dietary Ca that is absorbed. Additionally, it was found that the placenta wall served as an effective barrier against Sr in a developing fetus but that juvenile mammals do not discriminate against Sr (16,19–22).

Use of Sr/Ca ratios for inferring diet or trophic levels is subject to uncertainties in Sr-Ca partitioning among various skeletal parts, as well as variations in Sr/Ca ratios of soil or bedrock sources. For example, high-precision measurements have shown that in humans, tooth enamel generally has lower Sr contents as compared to bone (12,23,24). In addition, dietary Sr/Ca ratios may change during an organism's lifetime, and it may be important to compare identical skeletal components among individuals, such as only using the third molar or the middle section of a femur. Overall, skeletal Ca (and by analogy Sr) in human bones is essentially completely exchanged on a six year basis, hence analysis of bones will be representative of the average dietary Sr over the last six years of life (25). A six-year residence time for skeletal Ca is a body average and significantly longer or shorter times can be inferred for different skeletal elements as well as for different sexes, levels of maturity, health, and race (26–29). In contrast, it seems likely that tooth enamel does not exchange Sr after it is formed (30), making tooth enamel an excellent proxy for the average dietary Sr isotope composition that was ingested when the tooth formed.

Many of the uncertainties involved in interpreting elemental ratios and abundances are minimized if isotopic compositions are used (12,23,24). For example, if the Sr isotope composition of an individual's diet is constant, their bones and teeth will have the same Sr isotope composition, and yet the Sr contents of skeletal components may vary due to variations in Sr partitioning. The Sr isotope composition of bone and teeth of an individual who migrated from one locality to another is likely to be different; the composition of the tooth should reflect the person's birth place and the Sr isotope composition of the bone should reflect where the person resided later in life. It is important to recognize that this type of analysis may not provide unique geographic constraints in cases where the Sr isotope composition of different regions are similar. Tighter constraints are likely to be obtainable in such cases if isotopic analysis of possible family members is done, based on the assumption of similar Sr isotope compositions of their diet.

One of the complicating factors in using the Sr content and isotope composition of skeletal elements to infer past events is the pristine nature of a sample. Burial and fossilization of skeletal elements can mobilize Sr, leading to partial or total isotopic equilibration with components such as ground water. It is generally observed that there is a progression in the degradation of skeletal material that includes addition of carbonate minerals into the pores of skeletal elements by fluids, followed by recrystallization of the fine-grained primary hydroxyapatite into coarser domains of hydroxyapatite (7). The recrystallization process is particularly troublesome, because the coarse recrystallized material incorporates some of the carbonate material that was introduced into the skeletal element. Typically, carbonate material in a skeletal element can be removed by leaching in weak acetic acid, which dissolves carbonate but does not attack the hydroxyapatite (7,23). In the case of recrystallized hydroxyapatite that has incorporated carbonate, this leaching method is not effective at isolating the primary Sr isotope composition, and the measured Sr content and isotope composition will be a mixture of the primary skeletal hydroxyapatite and the Sr that was externally introduced during secondary carbonate deposition.

Acetic acid leaching of bones has been successful in allowing the primary Sr content and isotope composition to be determined in buried prehistoric human bones from the arid Southwestern United States (12,23), but in another study, such a leaching technique was not able to recover the primary Sr content and isotopic composition from human bones that had been buried for only a short time (~20 years) in the hot humid climate of Southeast Asia (see below). In situations where the primary Sr isotope compositions of bone cannot be determined because of degradation of the skeleton, isotopic analysis of tooth enamel is the only approach that will isolate the primary Sr isotope compositions. Tooth enamel is relatively nonporous, which reduces the ability of ground water to introduce exotic material, and this has been confirmed by other studies (31).

Assessing the Isotope Composition of Dietary Sr for Samples of Unknown Origin

The Sr isotope composition of skeletal material reflects the concentration-weighted average of the Sr that was ingested, integrated over the time period that the particular skeletal component was open to Sr exchange with the entire organism. Ultimately, the degree of detail to which Sr isotopes can be used to elucidate geographic information is largely a function of the diversity of isotopic composition of dietary Sr. In the case of a foraging animal, detailed geographic information may be obtainable for animals that forage over a restricted range, whereas only regional information may be obtainable from organisms that forage over a wide range. In the case of humans, the utility of using Sr isotopes to infer geographic information is probably more difficult today than it was just two or three decades ago, because of the increased national and international diversity of Ca (and Sr) sources in food.

The best way to determine the average Sr isotope composition of dietary Sr is to analyze the same skeletal element of organisms from known geographic locations. In the case of humans, isotopic

analysis of potential close relatives of a known geographic location is likely to be a very accurate approach. The database from these control groups can then be used to determine the geographic origin of the organism in question. However, it is likely that the initial stages of an investigative case might first look to a compilation of measured Sr isotope compositions of soil and bedrock, taking data that are scattered throughout the scientific literature. Such an approach is more detailed than using the first-order predictive model of Fig. 1, but is still likely to only answer general questions, such as, "Was this person born in California or Florida?".

Use of Bedrock Samples to Establish the Isotopic Composition of Dietary Sr

Using the compilation approach, reasonably precise estimates of the isotopic composition of dietary Sr can be obtained from the concentration-weighted average of the Sr isotope composition of the bedrock from a surrounding area. The Sr isotope composition of different rocks from all over the world have been determined from over three decades of work by isotope geochemists, and rocks from most areas of the world have been directly measured or their Sr isotope composition can be inferred from rocks of similar age and composition. An excellent check may be made by comparison with Sr isotope analyses of ground and surface water samples, which provide an assessment of the isotopic composition of local "mobile" Sr. In terms of assessing the Sr isotope composition of an organism's diet, there are three main components: soil (source of nutrients for plants), precipitation, and dry fall (e.g., atmospheric dust deposited on plant leaves) (6,32,33). The relative importance of these components is a function of their Sr concentration; the soil component is generally the major factor, followed to a lesser degree by the precipitation and dry fall components. In some parts of an ecosystem, however, the atmospheric inputs (precipitation and dry fall) can impact the Sr isotope budget (34,35).

The isotopic composition of soil Sr is controlled by rock type and mineral phases that have been chemically and physically broken down. However, the isotopic composition of Sr available to a plant cannot be approximated by a bulk soil or rock analysis, because plants are only able to use the Sr that is easily exchangeable. The isotopic composition of exchangeable Sr may be estimated by analyzing a weak HCl- or ammonia acetate-leach of a bulk soil sample. It must be recognized, however, that soil formation and evolution is dynamic, and the Sr isotope composition of a soil can be changed through time by climatic and anthropogenic effects (6).

In Fig. 2, we illustrate a case example from the Grasshopper Pueblo, Arizona (also discussed below), where the local concentration-weighted Sr isotope composition that is determined from modern rodents or bones and teeth from 13th and 14th century burials may be compared to the isotopic compositions determined on soil and bedrock samples from the area. As discussed in Appendix 1, $^{87}Sr/^{86}Sr$ variations in rocks are expected to inversely vary with Sr contents in an exponential manner, and a model curve can be constructed that closely matches the soil and bedrock samples. Similarly, a mixing line between a high-Sr, low-$^{87}Sr/^{86}Sr$ and low-Sr, high-$^{87}Sr/^{86}Sr$ endmembers of the model curve closely matches the observed variations. Although measurements on samples and predictive models demonstrate the wide range of isotopic compositions that may exist in one geographic location, the low Sr contents of the high-$^{87}Sr/^{86}Sr$ components minimize their contribution to dietary Sr. This is well illustrated by integrating the model curves (see Appendix 1), which produce comparatively low, concentration-weighted $^{87}Sr/^{86}Sr$ ratios that closely match the isotopic compositions measured for local modern field mice, or those of bones from 13th and 14th century individuals who are buried on the site and have isotopic compositions that are interpreted to reflect those of the local dietary Sr (Fig. 2) (12).

Use of concentration-weighted isotopic compositions is an important modification to the first-order predictive model (Fig. 1; Appendix 1), because the low Sr contents of the high-$^{87}Sr/^{86}Sr$ components in a region will not be significant contributions to dietary Sr. A practical approach to a specific case would be to fit a mixing or theoretical-based curve to measured soil and bedrock samples (or acid leaches of such samples) and calculate the concentration-weighted isotopic composition using the equations presented in Appendix 1. For a given geological age for a region, use of concentration-weighted isotopic compositions will produce significantly different interpretations as compared to using simple averages of measured geologic samples (Fig. 3).

Analytical Methods

The details of preparing bone and teeth material for Sr isotope analysis in our lab have been reported in a number of articles (12,23). In general, bone and teeth samples are prepared by first re-

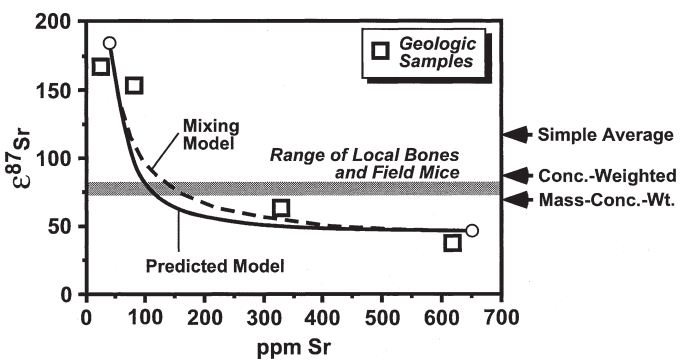

FIG. 2—*Comparison of model $\varepsilon^{87}Sr$-ppm Sr variations, measured compositions of local geologic samples, and field mice (teeth and bones) and human bones from the Grasshopper Pueblo, Arizona. The $\varepsilon^{87}Sr$ values of local dietary Sr for the 13th and 14th century burials (bones) and modern field mice are similar, ranging from $\varepsilon^{87}Sr = +72$ to $+85$, and reflect the concentration-averaged Sr isotope composition of the region. In contrast, model variations (see Appendix 1) and measured soil and rock samples vary greatly in their $\varepsilon^{87}Sr$ values; integration of the curves produces similar concentration-weighted $\varepsilon^{87}Sr$ values, whereas simple averages do not.*

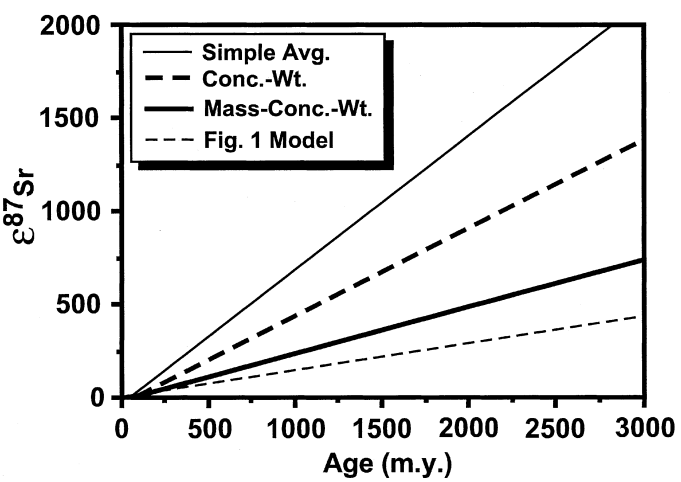

FIG. 3—*Comparison of $\varepsilon^{87}Sr$ variations as a function of bedrock age (in millions of years) for different models for Sr isotope variations. See Appendix 1 for details of the models.*

moving extraneous soil particles by use of a dental pick and ultrasonication in double-distilled water. Powders of bone, teeth, or antlers are then prepared using a drill, followed by leaching in 1M acetic acid. The leaching step is critical in cases involving skeletal material that is not in pristine condition, so that external Sr that may have been introduced as carbonate can be removed (see above). Skeletal material is then combusted for 8-12 h at ~800°C to burn away all organic material, followed by dissolution in concentrated nitric acid. Strontium contents may be determined by isotope dilution on the sample used for isotopic analysis. Reproducibility of Sr contents are typically not as good as those observed for rock samples, and likely reflect differences in the "dry" weight of the sample that are due to variations in ashing. In addition, for samples that are not in pristine condition, leaching using acetic acid has variable effectiveness, particularly for heterogeneous samples. Following dissolution, Sr is separated from all other cations by ion-exchange chromatography, which may involve an ion-specific resin such as Sr-Spec resin (E-I Chrom Industries), or traditional cation exchange resin (e.g., Bio-Rad AG 50W X 8).

The purified Sr is typically analyzed by thermal ionization mass spectrometry (TIMS). All the reported Sr isotope data (Tables 1–3) were obtained at the University of Wisconsin-Madison Radiogenic Isotope Laboratory using a Micromass Sector 54 mass spectrome-

TABLE 1—*Measured $^{87}Sr/^{86}Sr$ isotope ratios of white-tailed deer antler samples.*

Sample	ε_{Sr}	$^{87}Sr/^{86}Sr$	Year Killed	Location of Kill
Michigan Control Group				
S-1	151.8	0.715198	1993/94?	Sect 28
S-3	126.9	0.713440	1995	Sect 29
S-3*	126.9	0.713439		
S-3†	127.0	0.713447		
S-4-A	135.5	0.714044	1991	Sect 28
S-4-B	136.4	0.714111	1991	Sect 28
S-5	128.6	0.713559	1991	Sect 27
S-5*	128.6	0.713558		
S-5†	128.6	0.713562		
S-6	171.3	0.716571	1994	Sect 28
S-6*	171.2	0.716563		
S-6†	171.4	0.716579		
S-7	140.4	0.714393	1993	Sect 29
Wisconsin Control Group				
S-8-A	70.2	0.709443	1997	Sect 29
S-8-A*	70.0	0.709429		
S-8-B	70.5	0.709469	1997	Sect 29
S-16	67.9	0.709286	1988	Sect 4
S-16†	68.1	0.709300		
S-15	72.0	0.709569	1996	Sect 4
S-17	69.6	0.709405	1993	Sect 4
S-19	83.0	0.710348	1998	Sect 23
S-19†	83.0	0.710345		
S-20	79.8	0.710121	1998	Sect 23
Deer in Question				
K-1	70.3	0.709453	1996	
K-1*	70.2	0.709449		
K-1†	70.2	0.709448		
K-1*	70.2	0.709443		

* Reanalysis of same sample. † Analysis of different sample split processed through the entire analytical procedure. S-4-A and S-4-B from same antler, -A base, -B top. S-8-A and S-8-B from same antler, -A base, -B top. Year killed refers to the year the deer was harvested; all the deer were harvested during gun or archery season in their respective states. Location of kill refers to the section number where the deer were harvested, all the Michigan deer are from township 47N range 39W and all the Wisconsin deer are from township 11N range 8E.

TABLE 2—*Bone elements from commingled human remains from the Vietnam Conflict.*

Sample	Sr ppm	$\varepsilon^{87}Sr$	$^{87}Sr/^{86}Sr$
S-1	275.1	136.1	0.71409
S-2 (ash 1)	113.6	160.3	0.71579
S-2 (ash 2)	167.1	203.3	0.71882
S-2 (ash 2)	168.9	202.6	0.71877
S-2 (ash 3)	177.5	192.1	0.71803
S-2 (ash 4)	179.5	207.5	0.71912
S-3 (ash 1)	287.8	152.6	0.71525
S-3 (ash 2)	301.3	156.0	0.71549
S-4	196.0	152.7	0.71526
S-5 (ash 1)	230.0	132.3	0.71382
S-5 (ash 2)	273.7	132.9	0.71386
S-6	197.5	149.6	0.71504
S-7 (ash 1)	263.9	154.9	0.71541
S-7 (ash 2)	314.7	159.4	0.71573
S-8	219.7	153.2	0.71529
S-9	497.3	145.4	0.71474
S-10	299.9	131.0	0.71373
S-11	185.4	136.6	0.71412
S-12	175.4	134.4	0.71397
S-13 (ash 1)	429.8	144.5	0.71468
S-13 (ash 2)	304.1	140.0	0.71436
S-14	194.6	141.2	0.71445
S-15	337.3	154.0	0.71535

TABLE 3—*Tooth elements from commingled human remains from the Vietnam Conflict.*

Sample	Type	Sr ppm	$\varepsilon^{87}Sr$	$^{87}Sr/^{86}Sr$
Individual 1				
S-16	Molar 3	73.5	103.6	0.71180
S-17	Canine	94.6	99.4	0.71150
S-18	Incisor	121.7	104.6	0.71187
S-19 (ash 1)	Molar 3	67.2	97.4	0.71136
S-19 (ash 2)		85.8	105.5	0.71193
S-19 (ash 3)		71.4	98.2	0.71142
S-19 (ash 3)		69.4	98.4	0.71143
S-20	Canine	114.3	103.9	0.71182
Individual 2				
S-21	Molar 2	148.4	92.5	0.71102
S-22	Canine	169.0	95.2	0.71121
S-23 (ash 1)	Incisor	187.0	101.5	0.71165
S-23 (ash 2)		161.6	98.9	0.71147
S-24 (ash 1)	Canine	175.4	99.4	0.71150
S-24 (ash 2)		185.5	97.1	0.71134
S-24 (ash 2)		170.6	97.3	0.71135
S-25	Molar 3	87.2	87.2	0.71064
S-25		102.4	86.6	0.71060
Individual 3				
S-26 (ash 1)	Molar 3	115.8	53.5	0.70827
S-26 (ash 2)		115.0	53.7	0.70828
S-26 (ash 2)		117.7	52.5	0.70820
S-27 (ash 1)	Canine	177.1	60.5	0.70876
S-27 (ash 2)		188.5	59.2	0.70867
S-27 (ash 2)		180.7	59.5	0.70869
S-27 (ash 2)		182.9	59.7	0.70871
S-27 (ash 2)		186.9	59.3	0.70868
S-28	Incisor	200.0	77.2	0.70994
S-29 (ash 1)	Molar	134.5	60.2	0.70874
S-29 (ash 2)		126.8	60.6	0.70877
S-30 (ash 1)	Incisor	162.0	102.5	0.71172
S-30 (ash 2)		200.3	112.4	0.71242
S-30 (ash 3)		265.7	102.1	0.71169

ter. All Sr isotope ratios were collected using a multi-collector dynamic analysis, which removes all Faraday collector biases, and normalized to an $^{86}Sr/^{88}Sr$ ratio of 0.1194. The external precision of this analysis method for the $^{87}Sr/^{86}Sr$ measurement is 0.004 to 0.002%, and is typical for modern multi-collector TIMS instruments. Long-term reproducibility of Sr isotope measurements is checked by analysis of the NIST Sr carbonate isotope reference material SRM-987. The $^{87}Sr/^{86}Sr$ ratio measured for SRM-987 at the University of Wisconsin-Madison is 0.710259 ± 0.000013 (n = 150; 2-standard deviations) over the last three years.

Case Examples

We present three case studies of using Sr isotopes to trace geographic origins of animals and humans. The first case involves deer, where a control group of known geographic origin is used to interpret results from unknown samples. The second involves human remains where geographic origin is unknown, but is inferred from a database of Sr isotope compositions from potential localities. A third case is a summary of previously published work where migration of ancient humans is documented using detailed teeth and bone analysis.

Geographic Fingerprinting of White-Tailed Deer Antlers

In a criminal investigation, it was necessary to determine if an antlered white-tailed deer was harvested in the Upper Peninsula of Michigan near Bruce Crossing, MI, or if it was harvested in Central Wisconsin near Portage, WI. The only physical evidence for this investigation was the antlers from the deer in question. Strontium isotope analyses of the antlers is easily able to resolve this question because these two areas are geologically very distinct. The area of Bruce Crossing, Michigan is underlain by the Jacobsville Sandstone Formation, which is a Late Proterozoic rock unit that consists of quartz sandstone and shale that were derived by erosion of the surrounding Archean and Proterozoic rocks (36,37). The area in Wisconsin is underlain by Upper Cambrian sedimentary rocks (sandstones, carbonates, and shales) of the Trempealeau and Tunnel City Groups and the Galesville Sandstone (38). The large differences in the ages of these rocks imply that there should be large differences in the $^{87}Sr/^{86}Sr$ of the two areas (Fig. 1). Six antler samples from each of the two suspect areas were taken from archives that were collected by official wardens from hunters who had legally harvested deer in previous years.

The Sr isotope composition of the six antlers from Michigan range from $\varepsilon^{87}Sr$ values of +126.9 to +171.3, and the Sr isotope composition of the antlers from Wisconsin range from $\varepsilon^{87}Sr$ values of +68.0 to +83.0 (Fig. 4; Table 1). There is no overlap in the Sr isotope composition of the antlers from these two areas, and the difference in Sr isotope composition between the Wisconsin and Michigan deer antlers is over 200 times the analytical uncertainty of a single measurement. Figure 4 readily shows that the antler from the deer in question is an exact isotopic match to the antlers that were obtained from Wisconsin deer (Fig. 4). From these Sr isotope data we can confidently conclude that the deer in question did not come from near Bruce Crossing, MI, and that the Sr isotope composition of antler from the deer in question is consistent with the deer having been harvested from near Portage, Wisconsin.

This case demonstrates the most robust application of Sr isotopes to forensic geographic fingerprinting, because a specific geographic question was asked: Did the deer live in Wisconsin or Michigan? If it had not been possible to tightly constrain the possible geographic locations, there may be significant overlap in isotopic compositions of possible geographic localities, greatly increasing the uncertainty in geographic fingerprinting.

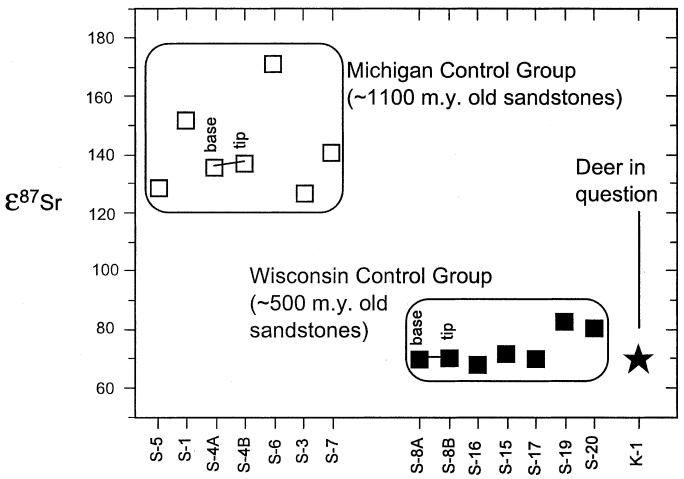

FIG. 4—*Plot of average $\varepsilon^{87}Sr$ measured in white-tailed deer antlers from Wisconsin and Michigan, plotted against sample number (Table 1). The measured $\varepsilon^{87}Sr$ values of white-tailed deer from the Upper Peninsula of Michigan are much higher as compared to the $\varepsilon^{87}Sr$ values of white-tailed deer from Central Wisconsin, reflecting the fact that the rocks from the Upper Peninsula of Michigan are much older (~1,100 million years) as compared to those of Central Wisconsin (~500 million years). These control groups define the average dietary Sr isotope composition from the two areas. The deer in question has a Sr isotope composition that is not consistent with an origin from the Upper Peninsula of Michigan; the $\varepsilon^{87}Sr$ values are, however, an exact match to the Sr isotope composition of the Wisconsin control group.*

Commingled Skeletal Material from the Vietnam Conflict

A second case example involves an attempt to identify commingled human remains of casualties from the Vietnam Conflict. The goals of this project were to link the commingled remains to each casualty, and to attempt to identify where each individual resided in the United States. In this case, skeletal material was thought to belong to three individuals, although the poor preservation prevented skeletal reconstruction. The commingled bones and teeth had been buried in a shallow grave in Vietnam for over two decades. Based on dental reconstructions, teeth samples had been grouped by U.S. Army dentists to each of three individuals. Additionally, it was believed, based on military records, that one individual was born and lived in North Central California for much of his life, the next individual was born in the Upper Peninsula of Michigan and resided in Detroit (Michigan) for much of his life, and the third individual was born in Vermont and resided in Massachusetts for much of his life. Because of their limited time in Vietnam, there would be only small contributions of Sr from Vietnam to their bones, and no contribution to their teeth (see above).

Strontium isotope analyses and Sr contents were measured on 15 bone fragments (Table 2) and 15 tooth fragments (5 teeth fragments from each of the three individuals, as identified by dental reconstructions; Table 3). The Sr isotope composition of the bone samples is higher ($\varepsilon^{87}Sr$ values from +130 to +220), as compared to the tooth samples ($\varepsilon^{87}Sr$ values from +50 to +110; Fig. 5A), and the Sr contents measured for the bone samples are highly variable (100 to 500 ppm Sr). In contrast, the Sr contents measured for the teeth define a relatively restricted range, 75 to 280 ppm Sr (Fig. 5B). On a plot of Sr content versus $\varepsilon^{87}Sr$ there are no clear group-

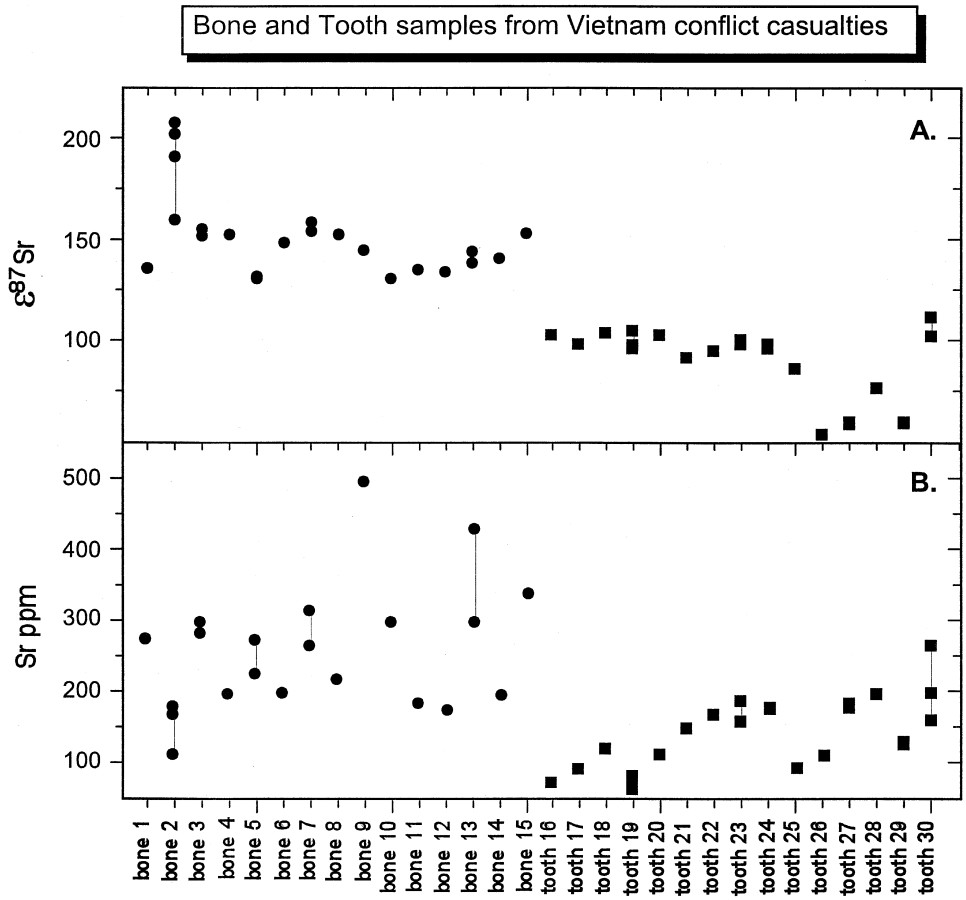

FIG. 5—A. *Plot of measured ε^{87}Sr values of commingled bones and teeth of casualties from the Vietnam Conflict plotted against sample number (Tables 2 and 3). Duplicate measurements of different splits of the same sample, as processed through the entire analytical procedure, are connected by tie lines. The Sr isotope composition of all the bone fragments are significantly higher than those measured for teeth. Duplicate measurements of bones and the single tooth that contained enamel and dentine (tooth #30) are much larger than the analytical uncertainty of a measurement. The large isotopic heterogeneity in these duplicate analyses are interpreted to reflect an open system behavior of Sr in porous skeletal material. Duplicate measurement of nonporous tooth enamel are more reproducible, suggesting that there has been little open-system behavior for Sr in the tooth enamel samples.* B. *Plot of measured Sr concentration of commingled bones and teeth of casualties from the Vietnam Conflict, plotted against sample number (Tables 2 and 3). Duplicate measurements of different splits of the same sample powder, processed through the entire analytical procedure, are connected by tie lines. Similar to the Sr isotope determinations, duplicate measurements of the Sr content of porous skeletal elements such as bones and tooth sample 30 are more variable as compared to the nonporous tooth enamel samples.*

ings of bone elements (Fig. 6); one sample (#2) has a high ε^{87}Sr value ($>+160$) and the other samples cluster between ε^{87}Sr values of $+130$ to $+160$.

Replicate analyses involved complete processing of different splits of the powdered bone sample through dissolution, chemical separation, and mass analysis, but yielded poor reproducibility for bone samples, which likely reflects their poor preservation state. For example, four replicate analyses of bone sample 2 define a range of ε^{87}Sr values from $+160$ to $+210$, and Sr contents range from 100 to 200 ppm. Replicate analyses of four other bone samples (samples 3, 5, 7, and 13) is better, where ε^{87}Sr values agree to a precision to ± 2 ε^{87}Sr units, but reproducibility of the Sr contents is poor, ranging from ± 7 to ± 30 ppm Sr. Replicate analyses of the same sample ashing are precise to ± 0.4 ε^{87}Sr units and Sr contents are precise to $\pm 1\%$, indicating that the poor reproducibility is not due to analytical error. The variability in the Sr isotope composition of the replicate bone analyses is correlated with Sr contents; there is a positive correlation between the measured Sr isotope composition and content between replicate analyses (Fig. 6), suggesting that some samples contain two Sr components, reflecting a mixture of primary Sr and that which was added through diagenesis after burial.

In contrast, the Sr isotope composition and Sr contents of tooth samples, except for sample 30, define three distinct groups (Fig. 7), which correspond to the dental reconstructions that were made independently (and unknown to us at the time of analysis) by U.S. Army dentists. Sample 30 is the only tooth sample that contained enamel and dentine, whereas all other samples were composed entirely of enamel. Three replicate analyses of sample 30 define a range of ± 5 ε^{87}Sr units and ± 52 ppm Sr. We believe that the Sr isotope integrity of sample 30 has been compromised by burial; like the bones, the porous dentine of this sample may have exchanged Sr with ground waters after burial, and we therefore discard sample 30 from any geographic interpretations. Replicate analyses of six tooth samples that are composed solely of enamel are more precise than those of bone samples, where ε^{87}Sr values reproduce to ± 0.2 to ± 4 ε^{87}Sr units, and Sr contents reproduce to ± 8.3 to ± 12 ppm Sr.

Strontium content-isotope composition variations can be divided into groups that correlate with the dental element analyzed (Fig. 7).

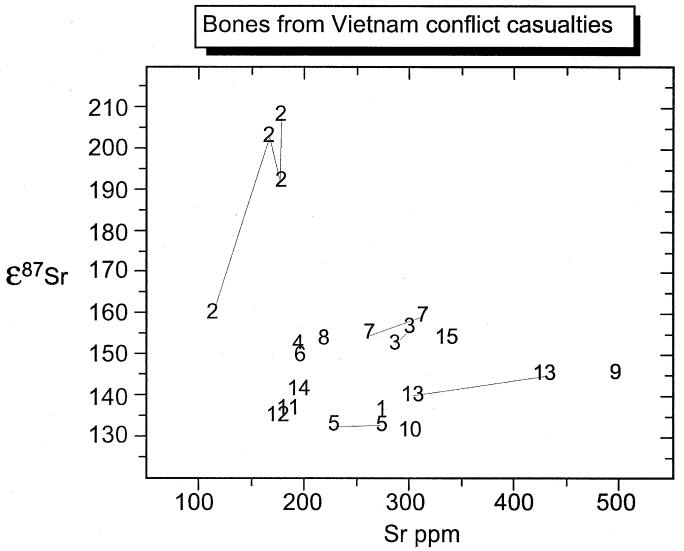

FIG. 6—*ε^{87}Sr-Sr content variations of bone fragments from casualties from the Vietnam Conflict (duplicate measurements shown by sample numbers connected by tie lines). Duplicate measurements are generally positively correlated, suggesting that the high ε^{87}Sr values of the bones are due to addition of radiogenic Sr by percolating ground waters. We interpret the similarity in ε^{87}Sr values of the majority of bone fragments to reflect open-system isotopic exchange during burial in the hot humid climate of Vietnam.*

Within each of the three groups, as defined by dental reconstructions, the incisor enamel has the highest Sr contents, the canine element contains intermediate Sr contents, and molar elements have the lowest Sr contents. We suggest that this correlation between dental element and Sr concentration reflects natural changes in diet as different dental components were grown. Incisors develop first, followed by canine elements, followed by molar elements (39). It is well known that Sr/Ca ratios decrease during "biopurification" (16), and the decrease in Sr contents with increasing age is exactly what we would expect as dietary sources change from mother's milk or dairy products to meat and grains. There is also a slight change in ε^{87}Sr value of the different tooth elements, where ε^{87}Sr values decrease from incisor to molar elements, and we believe that these changes in isotope composition of teeth also reflects changes in Sr sources of the dietary components. We note that although we expect diet changes to produce teeth that decrease in their Sr contents with age, Sr isotope compositions could increase or decrease, depending upon the geographic regions for dietary Sr.

The teeth of individuals 1 and 2 overlap in terms of their ε^{87}Sr values and hence cannot be geographically distinguished. For individuals 1 and 2, we would expect a childhood diet that reflects an average ε^{87}Sr value of about +90 to +105. The teeth from individual 3, however, are statistically distinct from individual 1 and 2, particularly if one discards the incisor data (the one sample that contains dentine). The Sr isotope composition of teeth from individual 3 indicates a childhood diet that had an average ε^{87}Sr value

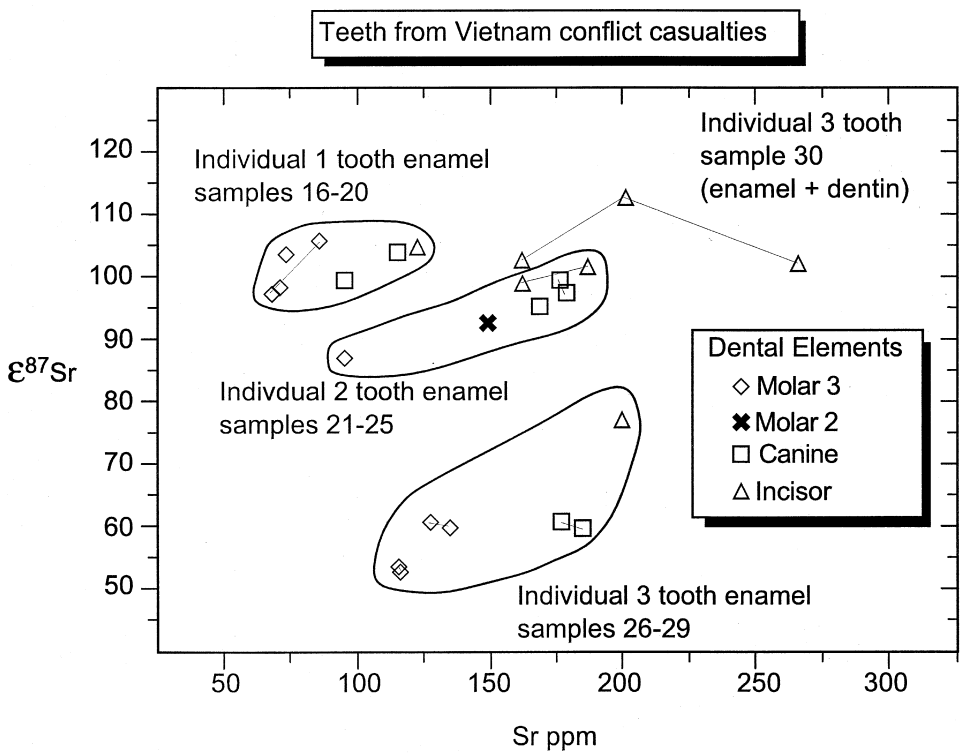

FIG. 7—*ε^{87}Sr-Sr content variations of tooth samples from casualties of the Vietnam Conflict. Samples are grouped according to the dental reconstruction of the three individuals involved in the incident; different dental elements (e.g., canine, incisor, and molar) are identified by different symbols. The Sr content of different dental elements from a single individual vary systematically, where incisors have higher Sr contents, as compared to canine teeth, as compared to molars. This variation in the Sr content of dental elements is interpreted to ultimately reflect the biopurification of Sr, as dietary Sr had sources from higher trophic levels as successive dental elements formed. The Sr isotope composition of teeth from individual three are unique and we interpret this individual to have been the person who lived in California. The Sr isotope composition of teeth from individuals 1 and 2 are not unique, so a positive determination of the residence of these individuals is impossible given the current data set.*

of about +50 to +60. Therefore, it should be possible to assign a geographic location to individual 3. However, unlike the white-tailed deer antler case, we do not have control groups from the three suspected geographic locations (North Central California, Michigan, Vermont/Massachusetts). We are therefore forced to infer the Sr isotope composition of the dietary Sr from the bedrock geology of the three areas that these individuals inhabited.

The Sr isotope composition of the bedrock geology of North Central California is distinct as compared to that of Vermont/Massachusetts and Michigan. Geologically, this area of California is underlain by two bedrock types: Mesozoic granitic rocks (ε^{87}Sr values −14 to +61; 40), and Neogene basalts of the Great Basin (ε^{87}Sr values from −7 to +21; 41). Nearby Mesozoic sandstones and shales of the Great Valley Sequence have ε^{87}Sr values of −4 to +108 (42), with a weighted average of +34, and may represent the Sr isotope composition of the majority of California agricultural products. Fresh water that is discharging from the Sacramento and San Joaquin Rivers, which drain the Great Valley, has an ε^{87}Sr value of +28 (43). In summary, we expect individuals who obtained their dietary Sr from this region of California to have ε^{87}Sr values between +30 and +60, which encompasses the range of individual 3 (Fig. 7).

The geology in Vermont and Massachusetts is diverse. The individual from this area was born in St. Johnsbury, Vermont and resided in Whitinsville, Massachusetts. Bedrock geology in the region includes Paleozoic limestone (marble) and mica schist (44). Although no Sr isotope data on either rock type is available in the immediate St. Johnsbury area, a local study of the Sr isotope compositions of ground water suggests that appropriate ε^{87}Sr values range from +84 to +94 (45). However, schists of the same rock group in Southeastern Vermont have much higher ε^{87}Sr values of +298 to +342 (46). Limestone and marble of this age are commonly deposited from seawater, which, worldwide, had ε^{87}Sr values of +50 to +57 at the time (47). We therefore interpret the ground water Sr isotope compositions to represent the concentration-weighted average of these diverse rock suites. The bedrock near Whitinsville, Massachusetts is dominantly granitic gneiss of Latest Proterozoic age, and one unit has a concentration-weighted average ε^{87}Sr of +356 (48). In summary, we expect this individual to have had dietary Sr that had ε^{87}Sr values of perhaps +80 to +100, although a variable contribution of Sr from the schists and granite may push that value >+100. Our estimate of +80 to +100 is strongly biased toward the Sr isotope compositions of ground water.

The third individual was born in Ontonagon, Michigan, on the south shore of Lake Superior. Geologically, this area is located on the southern edge of the Late Proterozoic Mid-Continent rift. The rocks of the rift exposed in this area are dominated by basaltic volcanic flows and minor rhyolitic volcanics of the Portage Lake volcanic series, which in turn are overlain by conglomerate, shale, and sandstone. Immediately to the south of the rift in this area are Archean gneisses of the Canadian Shield. Basaltic rocks of the rift have ε^{87}Sr values ranging from +30 to +108 (49), with a concentration-weighted average of +62. Rhyolites have a very wide range of ε^{87}Sr values from +199 up to +11,840 (49–50), with a Sr concentration-weighted average of +877. Archean gneisses have a similarly wide range and high average ε^{87}Sr values.

The Nonesuch Shale, which is expected to have a more radiogenic isotope composition (higher ε^{87}Sr values) than the rest of the rift sediments due to its high clay content, has a range of ε^{87}Sr values from +192 to +1,302 (50); the concentration-weighted average of the Nonesuch Shale is +463. There is not a large database for the Sr isotope composition of the rift-related sandstones, but based on the Sr isotope composition measured for the white-tailed deer antler samples from the Michigan control group, an ε^{87}Sr of +140 can be inferred. Unfortunately, no ground water Sr isotope studies are published for this region, which would greatly help in assessing the concentration-weighted average ε^{87}Sr value for the region. It is therefore very difficult to estimate the Sr isotope composition of dietary Sr for this individual.

The third individual is thought to have spent much of his life in Detroit, which sits at the eastern edge of the Michigan Basin. Strontium isotope ratios measured on rocks of the Michigan Basin (Southeastern Michigan and Northwestern Ohio) indicate a narrow range of ε^{87}Sr values, from +55 to +64 (51–53). It therefore seems most likely that an individual who obtained their dietary Sr from these areas of Michigan would have an ε^{87}Sr value in the range of +60 to perhaps +200. Strontium isotope data for ground waters would significantly restrict this possible range.

In summary, our inferences regarding dietary Sr isotope compositions based on bedrock geology lead to the following conclusions: the individual from California should have ε^{87}Sr = +30 to +60, the individual from Vermont/Massachusetts should have ε^{87}Sr = +80 to +90, and the individual from Michigan could have ε^{87}Sr = +90 to +200. Considering these variations we are confident in inferring that the teeth of individual 3 belong to the person who lived in California. The teeth of individuals 1 and 2 are from the individuals who lived in Michigan and Vermont/Massachusetts, but based on the current data it is impossible to confidently determine which set of teeth correspond to which of the two individuals.

It is possible that the ppm Sr − ε^{87}Sr variations (Fig. 7) provide some constraints on the geographic origin of individuals 1 and 2. The significant decrease in ε^{87}Sr from the early stage of tooth development (incisors ε^{87}Sr = +100) to later stages of tooth development (molars ε^{87}Sr = +86) is more consistent with the individual who moved from Ontonagon, Michigan to Detroit, Michigan, as compared to the individual who moved from Vermont to Massachusetts. Based on bedrock analyses and the discussion above, the individual who moved from Ontonagon to Detroit should have experienced a decrease in ε^{87}Sr values over time, whereas the individual who moved from Vermont to Massachusetts should have experienced an increase in ε^{87}Sr of dietary Sr. Significantly tighter constraints may be obtained through isotopic analyses of ground water/municipal water, as well as isotopic analysis of the baby teeth of potential family members.

Bone-Tooth Pairs from Burials of the Grasshopper Pueblo, Arizona

Isotopic analysis of pristine bone-tooth pairs from known intact skeletons provide an exceptional approach to forensic studies, and this is well illustrated in a study of the 14th century Grasshopper Pueblo, Arizona. These data were previously published by our group as part of a study of settlement and immigration during the 13th and 14th centuries in the Southwest United States (12,23), and here we discuss the implications those data have for forensic studies that require constraints on the geographic origin and movement of individuals. Unlike the Vietnam samples, where the primary Sr isotope composition of bones has been completely lost during several decades of diagenesis in a humid environment, bones from the Grasshopper site have retained their primary isotopic compositions during 600 to 700 years of burial in the arid climate of Arizona (12,23).

Construction and population of the Grasshopper Pueblo spanned approximately 150 years, and during its peak, contained 600 to 700 residents (12 and references within). It had long been suspected, therefore, that the population was a diverse mixture of longtime residents and immigrants who came to the site at various times in their lives. As noted above, it is anticipated that the Sr isotope composition of bones will largely reflect the isotopic composition of dietary Sr during later life, and to the degree that the residents had spent a number of years at the location, the $\varepsilon^{87}Sr_{Bone}$ values would also reflect those of the primary local food sources (largely maize grown on the site). The "local" Sr isotope composition of the site is taken to be the range determined from field mice (Fig. 8), given their limited geographic range, and there is general agreement between the $\varepsilon^{87}Sr_{Bone}$ and $\varepsilon^{87}Sr_{Teeth}$ of the mice. Individuals who have $\varepsilon^{87}Sr_{Teeth}$ values which exceed those of the local values are interpreted to be immigrants, and a likely locality for their higher values would have been the Shoofly Village to the west, which lies on high-ε^{87}Sr Precambrian basement rocks (12).

A number of bone-teeth pairs have similar ε^{87}Sr values (Fig. 8), and these are interpreted to reflect individuals who lived their entire lives at Grasshopper. In contrast, a number of individuals have $\varepsilon^{87}Sr_{Teeth} >> \varepsilon^{87}Sr_{Bone}$, and such relations are interpreted to reflect cases where an individual was born elsewhere (at a high-ε^{87}Sr region), but immigrated to Grasshopper later in life. As discussed above, such an interpretation is based on the premise that the Sr isotope composition of teeth reflects the dietary Sr consumed during childhood, and that no further isotopic exchange occurred after tooth growth ceased. Using standard isotope flux and exchange equations (see Appendix 2), we can calculate the time between immigration and death at Grasshopper, based on the degree to which the $\varepsilon^{87}Sr_{Bone}$ values have changed from their original values ($=\varepsilon^{87}Sr_{Teeth}$), toward those of the local isotopic composition. Assuming a Sr residence time in bones, a series of "immigrant isochrons" can be calculated for $\varepsilon^{87}Sr_{Teeth} - \varepsilon^{87}Sr_{Bone}$ variations (Fig. 8) as expressed in fraction of Sr residence times. If a Sr residence time in bone of six years is assumed it would suggest that immigrants migrated to Grasshopper 3 to 15 years before they died. Shorter times between immigration and death would be calculated if the residence time for Sr in bone is shorter, and this uncertainty may produce errors in the interpretation. Additional uncertainties are introduced because of the range of possible local Sr isotope compositions, which is indicated by the range in ε^{87}Sr values for field mice at the Grasshopper site, as well as the fact that some of the $\varepsilon^{87}Sr_{Bone}$ values are lower than those of the field mice (Fig. 8).

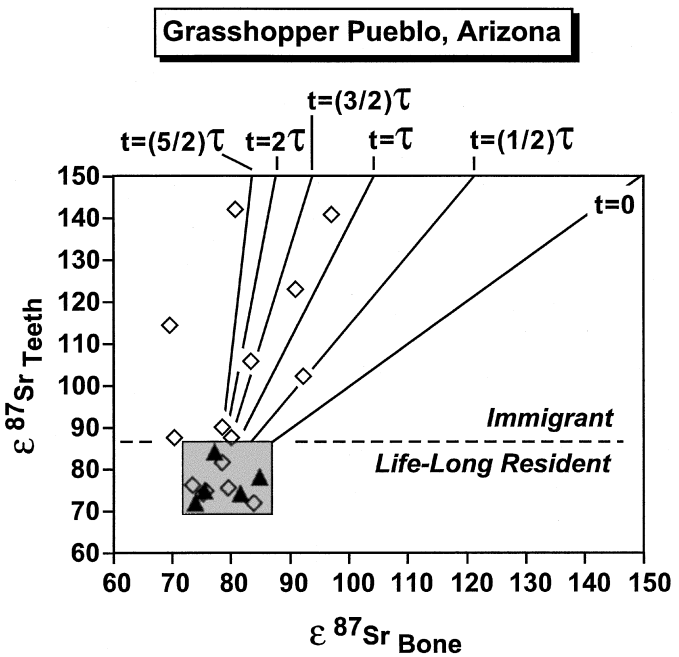

FIG. 8—*$\varepsilon^{87}Sr_{Teeth}$-$\varepsilon^{87}Sr_{Bone}$ variations for pairs from the same skeletons (open diamonds) from Grasshopper Pueblo burial sites. Gray box indicates isotopic composition for local Sr, as determined by analyses of local field mice (filled triangles). Individuals that have $\varepsilon^{87}Sr_{Teeth}$ values which overlap those of the local isotope composition ($\varepsilon^{87}Sr = +70$ to $+86$) are interpreted to have lived their entire lives at Grasshopper, whereas individuals who have $\varepsilon^{87}Sr_{Teeth} > +86$ are interpreted to be immigrants. Individuals who died immediately after immigrating to Grasshopper would have $\varepsilon^{87}Sr_{Teeth} = \varepsilon^{87}Sr_{Bone}$, which would lie on the "zero" age isochron. However, an immigrant who continued to reside at Grasshopper would continue to undergo Sr isotope exchange in their bones, and their $\varepsilon^{87}Sr_{Bone}$ values would gradually approach the local Sr isotope compositions; $\varepsilon^{87}Sr_{Teeth}$ values would remain unchanged due to the immobile nature of tooth Sr. The labeled lines are "immigrant isochron" ages, representing the time (in fraction of Sr residence time in bones) between immigration and death. Isochrons calculated using standard isotope flux and exchange equations (see Appendix 2), and an assumed local $\varepsilon^{87}Sr$ value of +78. If a six year Sr residence time in bone is assumed then immigrants migrated to Grasshopper 3 years ($t = {}^{1}/_{2}\tau$) to 15 years ($t = {}^{5}/_{2}\tau$) before they died.*

The applicability of Sr isotope analysis of bone-teeth pairs to forensic studies of modern humans will be limited by the availability of control groups (ideally obtained by isotopic analysis of baby teeth of close potential family members), and the temporal isotopic homogeneity of a person's diet, during tooth formation as well as during the last years of life when bone Sr is open to isotopic exchange. We suspect that individuals who consumed Ca (Sr) sources from geographically-diverse regions, which would impart Sr isotope heterogeneity, will be the hardest to "fingerprint" isotopically, and such cases may be most common in the last decade or so when food sources have been globally diverse. Such effects, however, will not be the same for all regions or countries, and may be compensated by isotopic analysis of any available bone or teeth from potential family members.

Conclusions

The geographic residence of humans and animals can be inferred from the Sr isotope compositions of Ca-bearing skeletal elements. The confidence to which a unique geographic fingerprint may be obtained rests on the isotopic distinctiveness of possible geographic locations and the degree to which the isotopic composition of dietary Sr is known. In order of decreasing confidence, examples include: (1) A test of two or more isotopically distinct geographic localities, where control groups from the possible localities can be measured. This type of case is analogous to the white-tailed deer study presented here. For studies involving humans, the best control may be obtained by isotopic analysis of baby teeth from close family members, or bones from deceased close family members. (2) A test of two or more geographic regions, where control groups are not available, but for which detailed isotopic data on rocks, soil, or ground waters are available from potential geographic locations. Because lithologic and age variations can be large in basement rocks, concentration-weighted Sr isotope compositions may be best estimated primarily from ground water and soil moisture, and secondarily from integration of ppm Sr $-$ ε^{87}Sr data obtained on local geologic samples. It is possible that isotopic analysis of mu-

nicipal water supplies may also be excellent representatives of dietary Sr. However, if significant non-local dietary Sr sources were consumed, the lack of a control group will introduce significant uncertainty in this approach. (3) A test of two or more geographic regions, where neither control group nor detailed isotopic data for rocks, soil, or local water are available from potential geographic locations. In such cases, general geographic constraints may be provided using a predicted model based on basement rock ages, which may also be refined based on lithology and integrative models.

Refinement of the residence time of Sr in human bones should allow precise ages to be calculated for the timing of movement of individuals who have moved between isotopically distinct regions. Such movement ages will be most precise where closely-related control groups are available. Diagenetic alteration of porous bone material, which is most likely in uncontained burials in warm humid climate, may prevent use of bone-teeth pairs as a means for evaluating geographic mobility of individuals in such cases.

Acknowledgments

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APPENDIX 1

Calculation of Concentration- and Mass-weighted Isotopic Compositions of the Crust Based on Parent/daughter Fractionation due to Rayleigh Fractionation during Crystallization of Magmas

Variations in Sr isotope compositions are due to long-lived radioactive decay of the ^{87}Rb-^{87}Sr decay system. To describe the isotopic variations due to radioactive decay, we define R_{meas} as measured isotope ratio $^{87}Sr/^{86}Sr$ and R_i as the initial $^{87}Sr/^{86}Sr$ a rock had at some time in the past. Also define λ = decay constant and t = time (in yrs)., and R_i = initial isotope ratio.

$$R_{meas} = R_i + \frac{^{87}Rb}{^{86}Sr}\,(e^{\lambda t} - 1) \qquad \text{(A1-1)}$$

Because $e^x \approx 1 + x$ for $x << 1$, (A1-1) becomes

$$R_{meas} = R_i + \frac{^{87}Rb}{^{86}Sr}\,\lambda t \qquad \text{(A1-2)}$$

Assume a constant conversion constant k between $^{87}Rb/^{86}Sr$ and the $[Rb]/[Sr]$ wt. ratio:

$$R_{meas} = R_i + \frac{[Rb]}{[Sr]}\,k\lambda t \qquad \text{(A1-3)}$$

Because Rb and Sr concentration variations in the Earth are fundamentally due to magmatic crystallization, which can be described by the Rayleigh fractionation model, Rb and Sr variations will follow:

$$[Sr] = [Sr]_0\,F^{(D_{Sr}-1)} \text{ and } [Rb] = [Rb]_0\,F^{(D_{Rb}-1)} \qquad \text{(A1-4)}$$

where F is the fraction of magma remaining during the crystallization of a magma (from 0 to 100% crystallized, F goes from 1 to 0), and D_{Rb} and D_{Sr} are the bulk crystal-liquid distribution coefficients.

$$\frac{[Rb]}{[Sr]} = [Rb]_0\left(\frac{1}{[Sr]_0}\right)^{\gamma}[Sr]^{(\gamma-1)} \qquad \text{(A1-5)}$$

where $\gamma = \left[\frac{D_{Rb}-1}{D_{Sr}-1}\right]$, which may be considered a constant. Equation (A1-3) then becomes:

$$R_{meas} = R_i + k\,[Rb]_0\left(\frac{1}{[Sr]_0}\right)^{\gamma}[Sr]^{(\gamma-1)}\lambda t \qquad \text{(A1-6)}$$

solving for $[Sr]$ produces

$$[Sr] = \left\{\frac{[R_{meas} - R_i]}{k\,[Rb]_0\lambda t}\,[Sr]_0^{\gamma}\right\}^{\left[\frac{1}{(\gamma-1)}\right]} \qquad \text{(A1-7)}$$

We define

$$A = \frac{([Sr]_0)^{\gamma}}{k\,[Rb]_0\,\lambda t},\; B = A\,R_i,\; C = \frac{1}{(\gamma-1)} \qquad \text{(A1-8)}$$

Equation (A1-7) then becomes

$$[Sr] = [A\,R_{meas} - B]^C \qquad \text{(A1-9)}$$

The concentration-weighted average isotope ratio R is

$$R_{C-Avg} = \frac{\int R\,f(R)\,dR}{\int f\,(R)\,dR} \qquad \text{(A1-10)}$$

Recast in terms of equation (A1-9), this becomes

$$R_{C-Avg} = \frac{\int R_{meas}\,[A\,R_{meas} - B]^C\,dR_{meas}}{\int [A\,R_{meas} - B]^C\,dR_{meas}} \qquad \text{(A1-11)}$$

Integration within the limits R_{meas}^{Min} and R_{meas}^{Max} produces

$$R_{C-Avg} = \frac{m\,|\,_{R_{meas}^{Min}}^{R_{Max}}}{n\,|\,_{R_{meas}^{Min}}^{R_{meas}^{Max}}}$$

where $m = \dfrac{[A\,R_{meas} - B]^{(C+1)}\,[A\,R_{meas}\,(C+1) + B]}{A^2\,(C+1)(C+2)}$ (A1-12)

and

$$n = \frac{[A\,R_{meas} - B]^{(C+1)}}{A\,(C+1)}$$

Equation (A1-12) will produce the concentration-weighted isotope ratio R that reflects the range of R_{meas} that is produced over a continuous crystallization interval F, followed by isotope evolution over time t. Equation (A1-12) assumes that each crystallization interval is equally represented in the crust. However, this does not account for the fact that the mass of magmas associated with early crystallization will represent larger volumes than the magmas that remain after extensive crystallization. A mass- and concentration-weighted average isotope composition R is calculated below to account for the decreasing mass contribution to the crust of magmas that have undergone greater crystallization.

We define a mass-weighted $[Sr]$ as

$$[Sr]_{MW} = F\,[Sr] \tag{A1-13}$$

where F is as defined in equation (A1-4).
Substituting equation (A1-9) produces

$$[Sr]_{MW} = F(A\,R_{meas} - B)^C \tag{A1-14}$$

Recall that

$$[Sr] = [Sr]_0\,F^{(D_{Sr}-1)} = (A\,R_{meas} - B)^C \tag{A1-15}$$

solving for F produces

$$F = \left[\frac{(A\,R_{meas} - B)^C}{[Sr]_0}\right]^{\left[\frac{1}{(D_{Sr}-1)}\right]} \tag{A1-16}$$

Equation (A1-14) then becomes

$$[Sr]_{MW} = [Sr]_0\left[\frac{(A\,R_{meas} - B)^C}{[Sr]_0}\right]^{\left[\frac{D_{Sr}}{(D_{Sr}-1)}\right]} \tag{A1-17}$$

Following equation (A1-10), the mass- and concentration-weighted isotope composition R is defined as

$$R_{M-C-Avg} = \frac{\int R_{meas}\,[Sr]_{MW}\,dR_{meas}}{\int [Sr]_{MW}\,dR_{meas}} \tag{A1-18}$$

Substitution of equation (A1-17), followed by integration within the limits R_{meas}^{Min} and R_{meas}^{Max}, produces

$$R_{M-C-Avg} = \frac{\left(\frac{o}{p}\right)\Big|_{R_{meas}^{Min}}^{R_{meas}^{Max}}}{\left(\frac{q}{r}\right)\Big|_{R_{meas}^{Min}}^{R_{meas}^{Max}}} \tag{A1-19}$$

where

$$o = [Sr]_0\,(D_{Sr} - 1)(A\,R_{meas} - B)\left\{\left(\frac{[A\,R_{meas} - B]^C}{[Sr]_0}\right)^{\left[\frac{D_{Sr}}{(D_{Sr}-1)}\right]}\right\} \times \{B\,[D_{Sr} - 1] + A\,[D_{Sr} - 1 + C\,D_{Sr}]\,R_{meas}\}$$

$$p = A^2\,(D_{Sr} - 1 + C\,D_{Sr})([2 + C]\,D_{Sr} - 2)$$

$$q = (D_{Sr} - 1)\,[Sr]_0\,(A\,R_{meas} - B)\left\{\left(\frac{[A\,R_{meas} - B]^C}{[Sr]_0}\right)^{\left[\frac{D_{Sr}}{(D_{Sr}-1)}\right]}\right\}$$

$$r = A\,(D_{Sr} - 1 + C\,D_{Sr})$$

APPENDIX 2

Calculation of Bone-teeth Residence Times for Sr Isotopes

Sr isotope exchange during bone remodeling can be calculated using standard flux equations and definitions (56,57).

We define the Sr residence time in bone as

$$\tau_{Sr} = \frac{M_{Sr}}{J_N} \tag{A2-1}$$

where J_N is the total Sr flux through bone and M_{Sr} is the total moles of Sr in bone.

We further define R_{bone} as the Sr isotope composition ($^{87}Sr/^{86}Sr$ ratios) for bone, and R_N as the Sr isotope ratio of flux J_N into bone after a change in geographic location. The differential equation that defines the change in R_{bone} with time, using the above definitions and formulation of Hodell et al. (56) is

$$\frac{d\,R_{bone}}{dt} = \frac{J_N R_N}{M_{Sr}} - \frac{R_{bone}}{\tau} \tag{A2-2}$$

rearrangement and integration produces:

$$\int \frac{d\,R_{bone}}{(R_N - R_{bone})} = \frac{1}{\tau}\int dt \tag{A2-3}$$

solving the integrals and setting boundary conditions for the integration constants (56) produces:

$$R_{bone}(t) = R_N - [R_N - R_{Tooth}]\,e^{[(t-t_0)/\tau]} \tag{A2-4}$$

where R_{tooth} is assumed to be equal to the initial isotopic composition of bone, R_N is the new isotope composition of the diet, and t_0 is the time of migration. For our purposes, we can set $t = 0$ (today). Solving for t_0, the time since migration based on any bone-tooth pair, equation (4) becomes:

$$t_0 = -\tau \ln\left[\frac{R_N - R_{bone}}{R_N - R_{Tooth}}\right] \tag{A2-5}$$

R_N, the dietary Sr isotope composition after migration, which must be assessed using the approaches outlined in the text.

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