Chapter 35
Applications of Sr Isotopes in Archaeology

N.M. Slovak and A. Paytan

Abstract The inclusion of radiogenic strontium isotope (\(^{87}\text{Sr}/^{86}\text{Sr}\)) analysis in archaeological and bioarchaeological research has resulted in the creation of new data by which to evaluate models of migration, culture change, colonization, trade, and exchange. Overwhelmingly, archaeologists have used radiogenic strontium isotope signatures in human enamel and bone apatite to reconstruct ancient mobility patterns and to distinguish between individuals of local and non-local origins at archaeological sites. The method also has been employed to establish the provenience of artifacts, ancient building materials, and foodstuffs as well as to track the origins and migratory patterns of prehistoric animals. The present chapter provides an introduction to the fundamental principles, approaches, applications, and future directions of radiogenic strontium isotope analysis in archaeology.

35.1 Introduction

The application of radiogenic strontium isotope (\(^{87}\text{Sr}/^{86}\text{Sr}\)) analysis\(^1\) in archaeology has revolutionized paleomobility studies. Whereas traditional archaeological investigations relied on artifactual and architectural evidence as proxies for population movement, the retrieval of radiogenic strontium isotope signatures from human and faunal skeletal material allows archaeologists to directly examine past residential mobility and related phenomena. The present chapter provides an introduction to the principles, methods, and applications of radiogenic strontium isotope analysis in archaeology, beginning with a general discussion of the properties of strontium isotopes.

Strontium (Sr) is a trace element that is found in most igneous, metamorphic, and sedimentary rock, as well as in river water, groundwater, seawater, soil, plants, and animals. Strontium has four naturally occurring stable isotopes (\(^{84}\text{Sr}, {86}\text{Sr}, {87}\text{Sr}, \text{and} {88}\text{Sr}\)), three of which are non-radiogenic: \(^{84}\text{Sr} (0.560\%), {86}\text{Sr} (9.870\%), \text{and} {88}\text{Sr} (82.53\%)\). The remaining isotope, \(^{87}\text{Sr} (7.040\%)\), is radiogenic and is formed by the radioactive decay of \(^{87}\text{Rb}\), with a half life of approximately \(4.88 \times 10^{10}\) years (Faure and Mensing 2005). The amount of \(^{87}\text{Sr}\) in a mineral or rock containing \(^{87}\text{Rb}\) depends upon two things: the age of the rock or mineral and its \(^{87}\text{Rb}/^{86}\text{Sr}\) ratio (Faure and Mensing 2005). The highest \(^{87}\text{Sr}/^{86}\text{Sr}\) values are found in very old rocks with high \(^{87}\text{Rb}/^{86}\text{Sr}\) ratios such as granites and shales, while younger rocks and minerals with low \(^{87}\text{Rb}/^{86}\text{Sr}\) ratios have correspondingly low \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios (Faure 1977). Because of their large atomic mass, \(^{87}\text{Sr}/^{86}\text{Sr}\) values change little as they pass from weathered rocks through soils to the food chain (Hurst and Davis 1981; Beard and Johnson 2000; but see Fietzke and Eisenhauer 2006; De Souza et al. 2007; Wakabayashi et al. 2007; Halicz et al. 2008; Ruggeberg et al. 2008). Importantly, variations in \(^{87}\text{Sr}/^{86}\text{Sr}\) values that occur along biogeochemical pathways are corrected for during mass spectrometry, at which time \(^{87}\text{Sr}/^{86}\text{Sr}\) signatures are normalized to the constant value of \(^{86}\text{Sr}/^{88}\text{Sr}\) in natural

\(^1\)Traditionally, many scholars have used the expression “stable strontium isotopes” when referring to \(^{87}\text{Sr}/^{86}\text{Sr}\) signatures. In the present chapter, we prefer to employ the phrase “radiogenic strontium isotopes” in reference to \(^{87}\text{Sr}/^{86}\text{Sr}\) values so as to distinguish this kind of data from recent paleodietary research (Knudson et al. 2010) that utilizes stable \(^{87}\text{Sr}/^{86}\text{Sr}\) signatures.
rocks (Beard and Johnson 2000). Accordingly, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in soil, groundwater, vegetation, and fauna largely reflect underlying $^{87}\text{Sr}/^{86}\text{Sr}$ bedrock values (Capo et al. 1998), with some input from atmospheric sources (see discussion in Bentley 2006 and references therein). A similar principle applies to the marine environment where the isotopic composition of modern seawater, characterized as $^{87}\text{Sr}/^{86}\text{Sr} = 0.7092$ (Veizer 1989), is derived from $^{87}\text{Sr}/^{86}\text{Sr}$ signatures of the input sources of strontium to the ocean and strontium deposition into sediments (Faure 1977).

Building off of the principles of strontium isotope geochemistry, Ericson (1985) first demonstrated that $^{87}\text{Sr}/^{86}\text{Sr}$ in human bones and teeth could be used to study aspects of ancient human behavior. Strontium substitutes for calcium in the foodweb, and is deposited in hydroxyapatite crystal in human tooth enamel and bones (Comar et al. 1957). Given that any mass-dependent fractionation is corrected for, reported $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in human tissue should reflect the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of water, plants, and animals consumed, which in turn should reflect $^{87}\text{Sr}/^{86}\text{Sr}$ bedrock signatures in a given region (Ericson 1985).

Tooth enamel forms during infancy and childhood, after which its chemical composition does not change (Hillson 1996). Conversely, human bone remodels continuously (Parfitt 1983). Assuming that an individual consumed only locally grown foods during his or her lifetime, $^{87}\text{Sr}/^{86}\text{Sr}$ values in tooth enamel should reflect childhood diet and, by extension, childhood locale, while $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in bone will reflect adult diet and, ideally, adult locale (Ericson 1985; Sealy et al. 1991). Different $^{87}\text{Sr}/^{86}\text{Sr}$ values in enamel and bone from a single individual may indicate dietary changes over time, which in turn, may indicate residence change (Ericson 1985) (Fig. 35.1).

As originally noted by Ericson (1985), the ability to trace prehistoric mobility using $^{87}\text{Sr}/^{86}\text{Sr}$ in human tissue is constrained by several factors. First, there must be sufficient geologic variability between the different residence areas under study such that variations in $^{87}\text{Sr}/^{86}\text{Sr}$ values can be detected. Conversely, there must be sufficient geologic homogeneity within a region if one is relying primarily on geologic data to reconstruct the $^{87}\text{Sr}/^{86}\text{Sr}$ range for a locale, although it should be noted that archaeologists regularly incorporate other classes of data (e.g. faunal and human isotopic values) when determining local $^{87}\text{Sr}/^{86}\text{Sr}$ signatures for a region (see Sect. 35.2.1). Second, radiogenic strontium isotope analysis may not be as effective at tracking movement between coastal areas if the inhabitants of those areas relied solely on marine foods. In such instances, individuals’ $^{87}\text{Sr}/^{86}\text{Sr}$ values will reflect the marine $^{87}\text{Sr}/^{86}\text{Sr}$ signature ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7092$, Veizer 1989), rather than terrestrial $^{87}\text{Sr}/^{86}\text{Sr}$ values. Third, $^{87}\text{Sr}/^{86}\text{Sr}$ values among human populations can

![Fig. 35.1 Potential outcomes and interpretations involved in strontium isotope analysis of human tooth enamel and bone. Although option 1 is the most frequent interpretation in either outcome, the remaining options should be ruled out (or at least considered) in strontium isotope studies](image-url)
serve as accurate markers of prehistoric mobility only if a dependency on imported foods can be ruled out. The consumption of foreign foods, particularly those high in Ca and Sr (e.g., dairy products, leafy greens, legumes, and fish), and Sr-rich food additives such as sea salt (Wright 2005) can significantly alter $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in human bones and teeth, even when consumed in small amounts (Burton and Wright 1995); thus a consideration of total dietary intake is necessary when interpreting $^{87}\text{Sr}/^{86}\text{Sr}$ results.

### 35.2 Materials and Methods

#### 35.2.1 Determining Local $^{87}\text{Sr}/^{86}\text{Sr}$ Signatures for a Region

The first step in conducting radiogenic strontium isotope analysis in archaeology is establishing local $^{87}\text{Sr}/^{86}\text{Sr}$ values for the regions or sites under study. A broad range of $^{87}\text{Sr}/^{86}\text{Sr}$ signatures for an area can be estimated from geological maps and later refined by measuring $^{87}\text{Sr}/^{86}\text{Sr}$ values in local, exposed bedrock and whole soil. Very old rocks with high Rb/Sr ratios generally exhibit $^{87}\text{Sr}/^{86}\text{Sr}$ signatures above 0.710 while younger rocks will have $^{87}\text{Sr}/^{86}\text{Sr}$ values less than 0.704. Although these types of data will provide a reliable estimate of geologic $^{87}\text{Sr}/^{86}\text{Sr}$ values for a region, they may not be indicative of human $^{87}\text{Sr}/^{86}\text{Sr}$ values. As initially noted by Sillen et al. (1998) and subsequently demonstrated and discussed by others (Price et al. 2002; Poszwa et al. 2004; Bentley 2006; Hedman et al. 2009), $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in geologic substrate can deviate significantly from bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values in the food chain. In order to best capture biologically available $^{87}\text{Sr}/^{86}\text{Sr}$ values for a region, it is recommended that researchers measure $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from small, local animals whose diets have been shown to reflect average $^{87}\text{Sr}/^{86}\text{Sr}$ regional values (Price et al. 2002).

Scholars have relied on a variety of fauna to calculate biologically available $^{87}\text{Sr}/^{86}\text{Sr}$ signatures, largely because modern animal diets often include imported foods and fertilizers that may skew $^{87}\text{Sr}/^{86}\text{Sr}$ faunal values (Price et al. 2002; Bentley 2006). On the other hand, archaeological bone, and to a lesser extent tooth enamel, is susceptible to diagenetic contamination. Given that $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in modern and archaeological fauna are subject to potential alteration, it is advisable to incorporate both types of data (preferably archaeological tooth enamel rather than bone) when establishing biologically available strontium isotope levels. Additionally comparative $^{87}\text{Sr}/^{86}\text{Sr}$ measurements from multiple species of local fauna likely will yield the most accurate representations of biologically available $^{87}\text{Sr}/^{86}\text{Sr}$ values (Price et al. 2002).

The bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ signature for a region generally is represented as a range of values, which is calculated using the mean of biologically available strontium isotope levels (as determined by local fauna) ± 2 s.d. (Price et al. 2002). These confidence limits, while admittedly arbitrary, have allowed researchers to differentiate between individuals of local and non-local origins at various archaeological sites. In some cases, however, biologically available strontium isotope ranges are inadequate proxies for human dietary Sr.

In her study of human skeletons from the archaeological site of Tikal, Guatemala, for example, Wright (2005) noted that approximately half of her human sample exhibited $^{87}\text{Sr}/^{86}\text{Sr}$ signatures higher than the $^{87}\text{Sr}/^{86}\text{Sr}$ range established for local fauna, plants, and water. Wright attributed these elevated human $^{87}\text{Sr}/^{86}\text{Sr}$ values to the consumption of imported sea salt, whose $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7092 was significantly higher than the local Tikal signature ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7078–0.7081$) (Wright 2005). A similar trend was noted at the archaeological site of Ancón, Peru (Slovak et al. 2009) where 34 of 35 human skeletons exhibited $^{87}\text{Sr}/^{86}\text{Sr}$ values notably higher than the biologically available $^{87}\text{Sr}/^{86}\text{Sr}$ range for the region (Fig. 35.2). Based on carbon and nitrogen isotope data from a subset of the Ancón skeletal sample, Slovak et al. (2009) and Slovak and Paytan (2009) demonstrated that marine resources comprised nearly half of ancient Ancóneros’ diets. The authors argue that the regular consumption of seafood likely raised inhabitants’ $^{87}\text{Sr}/^{86}\text{Sr}$ values from the local range ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7063–0.7068$) toward the $^{87}\text{Sr}/^{86}\text{Sr}$ of seawater.

In both of these case studies, local $^{87}\text{Sr}/^{86}\text{Sr}$ values for a human population would be better determined by
referencing the human $^{87}\text{Sr}/^{86}\text{Sr}$ data rather than using a $^{87}\text{Sr}/^{86}\text{Sr}$ range based on fauna and/or plants. This method, originally proposed by Wright (2005), removes individuals with outlying $^{87}\text{Sr}/^{86}\text{Sr}$ values from the data set, such that the revised body of data conforms to a normal distribution and more accurately reflects a local $^{87}\text{Sr}/^{86}\text{Sr}$ signature.

Ultimately, and perhaps frustratingly, there is no one method that best establishes a local $^{87}\text{Sr}/^{86}\text{Sr}$ signature for all sites and all individuals. In instances where archaeologists suspect that most of the human study population is locally born and most individuals consumed local, terrestrial foods, biologically available strontium isotope levels based on fauna or plants likely can identify prehistoric migration. On the other hand, if residential mobility is suspected or if imported or marine foods were consumed by some or all of the population, then statistical analyses of the human data is probably more effective at differentiating between local and non-local individuals.

Finally, the incorporation of additional isotopic tracers, particularly carbon ($\delta^{13}\text{C}$), nitrogen ($\delta^{15}\text{N}$), and oxygen ($\delta^{18}\text{O}$), also may help to clarify ambiguous $^{87}\text{Sr}/^{86}\text{Sr}$ data. Briefly, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values in human tissue reflect $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ signatures in different classes of plants and animals consumed and can be used to assess the relative amounts of marine and terrestrial foods in paleodiet (DeNiro and Epstein 1978, 1981; Schoeninger and DeNiro 1984). While neither carbon nor nitrogen isotope analysis can differentiate between individuals of local and non-local origin, they are important to migration studies because they can identify potential dietary biases that would affect strontium isotope signatures such as the ingestion of marine foods (for a more detailed discussion of carbon and nitrogen isotope applications in archaeology, see also Schwarcz and Schoeninger 2011).

Oxygen isotope signatures ($\delta^{18}\text{O}$), on the other hand, can detect prehistoric migration and have been used independent of, and in tandem with, strontium isotope signatures to identify paleomobility (White et al. 1998; Dupras and Schwarz 2001; Müller et al. 2003; Bentley and Knipper 2005; Knudson and Price 2007). $\delta^{18}\text{O}$ values in human tissue ultimately reflect the $\delta^{18}\text{O}$ in drinking water (Longinelli 1984; Koch et al. 1989), which in turn depends on a number of factors including temperature, distance from the sea, and elevation (Gat 1980; Yurtsever and Gat 1981; White et al. 1998). Oxygen isotope data, therefore,
provides an alternative line of evidence by which to track migration and monitor $^{87}\text{Sr}/^{86}\text{Sr}$ results.

### 35.2.2 Sample Selection and Sampling Techniques for Human Remains

#### 35.2.2.1 The Potential for Diagenesis

Diagenesis, or the molecular alteration of skeletal material after interment, is one of the major obstacles facing applications of radiogenic strontium isotope analysis in archaeology. Both physical and chemical changes to bones and teeth can occur in the post-depositional environment, including loss of biogenic $^{87}\text{Sr}/^{86}\text{Sr}$ signatures (Nelson et al. 1986; Sillen 1986; Price et al. 1992; Sillen and Sealy 1995; Hoppe et al. 2003). The degree of diagenetic contamination in any skeletal element is variable and depends on a number of factors including the length of interment, burial environment, and climate (Price et al. 1992; Nielsen-Marsh and Hedges 2000a, b). The type of material (tooth enamel vs. bone) and chemical element (strontium vs. carbon) analyzed also can affect the degree and rate of diagenetic processes over time (Schoeninger 1995).

Fortunately for strontium isotope studies, tooth enamel is largely resistant to diagenesis owing to its highly crystalline nature, low organic matter content, and lack of porosity (Koch et al. 1997; Hillson 2005). Numerous studies have demonstrated that biogenic enamel isotope signatures are retrievable from prehistoric samples with minimal treatment (Quade et al. 1992; Wang and Cerling 1994; Koch et al. 1997; Budd et al. 2000; Hoppe et al. 2003; Trickett et al. 2003). Bone, on the other hand, is much more prone to diagenetic alteration than tooth enamel, largely because of its high organic matter content (~30%), high porosity, and poorly crystalline structure. Upon interment, physical contaminants from the surrounding soil such as quartz, calcite, and clay can seep into the pore spaces of bone (Kyle 1986). Additionally, the post-mortem dissolution and recrystallization of bone mineral can affect strontium isotope levels in bone as well as the ratio of strontium to other elements such as calcium (Ca) and phosphorus (P) (Sillen 1981, 1989; Nelson et al. 1986; Price et al. 1992; Sandford 1992).

Over the last few decades, a number of studies have documented ways of measuring and monitoring diagenetic contamination in prehistoric materials and have established protocols to counteract diagenetic contamination in teeth and bone (Nelson et al. 1986; Sillen 1986; Price et al. 1992; Sillen and Sealy 1995; Nielsen-Marsh and Hedges 2000a, b; Hoppe et al. 2003). While the loss of biogenic isotope signatures in archaeological samples is at times unavoidable, the degree and effects of diagenetic contamination can be monitored and often ameliorated by appropriate sampling and cleaning procedures. (A more thorough discussion of these techniques can be found in Sects. 35.2.3.2 and 35.2.3.5.)

#### 35.2.2.2 Selecting a Sample: Human Teeth and Bone

Tooth enamel of the permanent dentition is laid down as a series of layers beginning just before birth, as is the case with the first molar crown, and continuing to approximately 14 years of age with the completion of the third molar crown (Table 35.1). Once formed, chemical signatures in teeth do not alter. $^{87}\text{Sr}/^{86}\text{Sr}$ values in different teeth, therefore, represent discrete growth periods in an individual’s childhood and adolescence.

Scholars generally select enamel samples from the first, second, and third permanent molars, as well as premolars, for radiogenic strontium isotope analysis. First molars begin their initial formation in utero (about 28–32 weeks after fertilization) (Hillson 1996) and are completed by around 3 years of age, while

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<th>Table 35.1 Approximate timing of dental crown and root formation of human permanent dentition</th>
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<td>Permanent tooth type</td>
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<tr>
<td>First incisor</td>
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<td>Second incisor</td>
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<tr>
<td>Canine</td>
</tr>
<tr>
<td>First premolar</td>
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<td>Second premolar</td>
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<td>First molar</td>
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<tr>
<td>Second molar</td>
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<tr>
<td>Third molar</td>
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Note that the age ranges here are approximate and that actual values may vary. Data compiled from Schour and Massler (1940) and Smith (1991).
second molars initially begin to form in an individual’s second or third year and crown completion takes place at 7 or 8 years (Schour and Massler 1940; Smith 1991). Premolars have fairly identical formation rates as second molars with the initiation of first and second premolar crowns beginning towards the end of the second year and continuing into the third year (Schour and Massler 1940; Smith 1991). First premolar crowns generally are completed by 6 years, followed closely by second premolar crowns at 7 years (Schour and Massler 1940; Smith 1991). Third molars are the most variable tooth in terms of their development (Hillson 1996), initially forming sometime between 7 and 13 years of age and crown completion occurring between 12 and 16 years. Third molars form well after the period of weaning and therefore are the least likely of all teeth to be affected by maternal strontium isotope signatures. On the other hand, not all individuals develop third molars (Garn et al. 1962) and therefore these teeth may not always be available for study. Additionally, by analyzing \(^{87}\text{Sr} / ^{86}\text{Sr}\) values in third molars in lieu of earlier forming teeth, archaeologists might overlook evidence for residential mobility that occurred earlier in childhood and adolescence.

Mandibular and maxillary dentitions have comparable development rates (Hillson 1996) such that isotope ratios from upper and lower teeth in the same dental position shoulder expected to yield highly similar isotope ratios. Similarly, chemical signatures are not expected to vary significantly among the different crown side surfaces (i.e., lingual, buccal, mesial, and distal) (Dolphin et al. 2005).

In compiling enamel samples for radiogenic strontium isotope analysis most researchers collect material from a single tooth rather than multiple teeth for any one individual. By doing so, archaeologists minimize their impact on the skeleton. At the same time, however, they likely underestimate the amount of mobility in prehistoric populations (Schweissing and Grupe 2003). A small number of researchers have addressed this problem by serially sampling teeth from a single individual. Schweissing and Grupe (2003), for example, collected enamel from each permanent tooth type from seven adult skeletons buried at the archaeological site of Neuburg/Donau in Bavaria. Their analysis revealed that six of the seven individuals migrated to the site at various stages in their childhoods, including one individual who appeared to have moved between at least two different regions in his life: once during childhood and another after 14 years of age. Importantly, in the case of this latter individual, evidence for multiple migrations would have remained undetected had the authors not sampled several teeth from the same skeleton. Similarly, Buikstra et al. (2004) documented several examples of early life migrations among skeletons from the Copan Acropolis. By comparing \(^{87}\text{Sr} / ^{86}\text{Sr}\) values from multiple teeth from single individuals the authors were able to determine around what age migrant individuals settled at Copan and, in at least one instance, detect multiple residence changes during an individual’s childhood (Fig. 35.3). Ultimately, the number of teeth analyzed from any one individual is subject to access and approval by curators and governmental agencies, and is likely to be guided by several factors including availability and integrity of dental material, cost of analysis, and the research question(s).

While it is feasible to differentiate between individuals of local and non-local origin using tooth enamel alone, many archaeological studies incorporate comparisons of strontium isotope signatures in tooth and bone from the same individual. Unlike tooth enamel, which forms early and does not chemically alter once laid down, human bone remodels throughout an individual’s lifetime (Tetelbaum 2000). The rate at which bones remodel depends upon the skeletal element and the type of bone analyzed. The diaphyses of long bones such as the tibia and femur take decades to remodel, while ribs replace their chemical constituents after only a few years (Jowsey 1961; Jowsey et al. 1965; Parfitt 1983; Eriksen 1986; Hill 1998). Similarly, different types of bone mineralize at different rates, such that dense cortical bone remodels slowly while trabecular bone remodels relatively rapidly (Mulhern and Van Gerven 1997, 2000).

Rates of turnover for different bones have significant consequences for migration studies and should be taken into account when selecting bone samples for analysis (Price et al. 2002). In instances where migrants relocated to an area within months or a few years prior to death, \(^{87}\text{Sr} / ^{86}\text{Sr}\) values in their skeletons, particularly among those bones with long turnover rates, likely will reflect their last place of residence rather than their new locale simply because strontium turnover in bone takes time. On the other hand, individuals who have resided in a place for multiple years and who have eaten locally-grown foods during that
time will exhibit skeletal $^{87}\text{Sr}/^{86}\text{Sr}$ values closer to, or within, the local $^{87}\text{Sr}/^{86}\text{Sr}$ range. In cases where individuals have resided at a new location for a period of a few years, $^{87}\text{Sr}/^{86}\text{Sr}$ values among bones with rapid turnover likely will approximate local values while $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from bones with prolonged remodeling rates probably will fall somewhere between the $^{87}\text{Sr}/^{86}\text{Sr}$ range of their previous locale and that of their current place of residence. A small number of studies have analyzed multiple bones from a single individual with interesting results. For example, in their study of the Tyrolean Iceman colloquially known as “Ötzi,” Hoogewerff et al. (2001) documented different $^{87}\text{Sr}/^{86}\text{Sr}$ values in the Iceman’s rib and femur, potentially indicating that Ötzi travelled to multiple locales during the last years of his life.

While it may be tempting to select those bones with fairly rapid turnovers such as ribs or the ends of long bones for radiogenic strontium isotope analysis, these bones are composed primarily of trabecular bone, which is much more susceptible to diagenesis than compact, cortical bone (Lambert et al. 1982; Buikstra et al. 1989). Ultimately the selection of bone(s) for sampling should be guided not only by the research question and scope, but also by the integrity of the specimen and the overall risk of diagenetic contamination as assessed from the burial environment.

35.2.2.3 Sampling Strategies for Human Tooth Enamel and Bone

Tooth Enamel

Most strontium isotope analyses of archaeological human tooth enamel rely on the bulk sampling method, in which enamel is collected across a tooth’s buccal, lingual, mesial, or distal crown surface from the occlusal margin to the cemento-enamel junction (CEJ), indiscriminate of enamel growth phases. By collecting samples in this manner, scientists average out potential fluctuations in $^{87}\text{Sr}/^{86}\text{Sr}$ that may have accumulated over the course of a tooth’s mineralization, which as discussed in the previous section occurs at different times and rates in an individual’s life. The resulting $^{87}\text{Sr}/^{86}\text{Sr}$ enamel value, therefore, represents

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**Fig. 35.3** $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from tooth enamel and bone for two adult individuals buried at the site of Copan. Based on data from Buikstra et al. (2004), this figure illustrates that neither individual was born locally although $^{87}\text{Sr}/^{86}\text{Sr}$ values from their bones (plotted at 40 and 50 years respectively) indicate that they lived at Copan several years prior to their death. Note that residence change for Burial 95-1 likely would not have been detected using the third molar (M3) only. Similarly, potential evidence for multiple childhood migrations for Burial 95-2 is detectable only by sampling several of the individual’s teeth (i.e., M1, I2, and M3).
a bulk signature formed over a period of several months or years in an individual’s childhood or adolescence (Fig. 35.4).

The collection of enamel bulk samples for isotope analysis is relatively straightforward. The enamel surface of the tooth selected for sampling should be abraded using either a Dremel tool or dental drill fitted with a carbide burr to eliminate adhering materials and to remove surface enamel that is most susceptible to diagenetic alteration (Hillson 1996). Enamel samples then can be drilled into a fine powder or chunked and later ground to a powder using a sterilized mortar and pestle. Researchers should aim to collect approximately 5–20 mg of tooth enamel if possible. Although only a small amount of enamel is needed for actual analysis, some material may be lost during sample preparation.

As an alternative to collecting samples in bulk, some consideration has been given to the possibility of microsampling enamel growth layers using either microdrilling or laser ablation. Laser ablation, and to a lesser extent microdrilling, requires far smaller samples than the bulk method, making it well-suited for isotopic analyses of rare materials such as fossil specimens or extremely small faunal teeth (Copeland et al. 2008). Furthermore, microsamples from a single human tooth potentially can capture seasonal fluctuations in enamel isotope values in strontium and other elements, as has been demonstrated in isotope studies of non-human animals (Koch et al. 1995; Fricke and O’Neal 1996; Stuart-Williams and Schwarcz 1997).

Despite these proposed benefits, microsampling remains largely untested on human archaeological samples. In part, this is due to the fact that microsampling via microdrilling remains technically difficult and fairly specialized (Richards et al. 2009). In addition, comparisons of laser ablation (LA-) and traditional solution mode-multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) $^{87}\text{Sr}/^{86}\text{Sr}$ measurements from tooth enamel have shown that the former method is generally more inaccurate and imprecise than solution-based techniques, owing to potential isobaric interferences during laser analysis (Simonetti et al. 2008; Nowell and Horstwood 2009). In recent years, however, a small number of studies have addressed these deficiencies and demonstrated that LA-MC-ICP-MS data may be appropriate in some archaeological cases. Research by Horstwood et al. (2008), for example, indicates that with proper on-line interference correction routines and additional calibration against a set of reference materials, reliable LA-MC-ICP-MS-based $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from archaeological specimens potentially can be produced. While Horstwood et al. (2008) acknowledge that their analytical protocol appears to work well on samples with concentrations $>$300 ppm Sr, the authors note that laser ablation data from samples with concentrations $\leq$200 ppm Sr exhibit a much greater degree of inaccuracy and should used cautiously. Copeland et al. (2008, 2010) analyzed modern and fossil rodent teeth from the Sterkfontein Valley of South Africa using both LA-MC-ICP-MS and TIMS. While their LA-MC-ICP-MS-based enamel $^{87}\text{Sr}/^{86}\text{Sr}$ values were less precise than the corresponding TIMS values, the authors do argue that LA-MC-ICP-MS is sufficiently accurate to investigate geographic origins and residential mobility if the geologic units under study are considerably variable from one another.

Finally, it is still somewhat unclear whether the analysis of microsamples from different human enamel growth layers can actually capture seasonal variations in $^{87}\text{Sr}/^{86}\text{Sr}$. As noted by Montgomery and Evans (2006), enamel mineralization in human teeth occurs in a multidirectional pattern such that $^{87}\text{Sr}/^{86}\text{Sr}$ values likely are averaged out across a tooth’s surface rather than occurring in a well-ordered, sequential fashion across growth layers. Recent research (Dolphin et al. 2005; Richards et al. 2008), however, indicates that incremental shifts in $^{87}\text{Sr}/^{86}\text{Sr}$ values within a single human tooth may be detectable using laser ablation.

Fig. 35.4 Bulk sampling of enamel for $^{87}\text{Sr}/^{86}\text{Sr}$ isotope analysis from a human skeleton buried at the site of Ancón, Peru. The arrow points to the region of the tooth from which enamel was drilled.

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techniques. In their study of deciduous dentition from modern children living in the Solís Valley of Mexico, Dolphin et al. (2005) illustrated that significant intra-tooth variations in trace element concentrations between prenatally- and postnatally-formed enamel were detectable using LA-ICP-MS. Similarly Richards et al. (2008) used laser ablation plasma ionization mass spectrometry (LA-PIMMS) to document three distinct clusters of $^{87}\text{Sr} /^{86}\text{Sr}$ values from a single Neanderthal third molar, which appeared to correspond to distinct phases of enamel secretion (Fig. 35.5). Richards et al. (2008) interpret the variation in $^{87}\text{Sr} /^{86}\text{Sr}$ signatures over time as evidence for Neanderthal mobility. Assuming the results of Dolphin et al.’s (2005) and Richards et al.’s (2008) study are applicable to archaeological samples more broadly, then laser ablation ICP-MS methods potentially can capture variation in enamel $^{87}\text{Sr} /^{86}\text{Sr}$ at much finer temporal resolutions than previously achieved using the bulk sampling technique.

**Bone**

The collection of bone for radiogenic strontium isotope analysis also can be performed using a Dremel Multipro drill, generally outfitted with an inverted cone tip to remove bone powder or a diamond disk saw to remove bone chunks. Prior to removing the sample, the sample area should be gently abraded to remove surface contamination. Samples should be taken from cortical bone, if possible, as this type of bone is less susceptible to diagenetic alteration than trabecular bone. It is suggested that at least 50 mg to 1 g of bone be collected since some preparation protocols for bone are quite rigorous and can result in a partial loss of sample (Hoppe et al. 2003).

**A Cautionary Note**

We conclude our discussion on sample selection with a note of caution. It should be remembered that collecting samples for radiogenic strontium isotope analysis involves the permanent removal of enamel and bone from archaeological specimens, samples which are entirely consumed during purification and mass spectrometry. Although the amount of enamel and bone needed for isotopic analysis is quite small (less than 20 mg for traditional bulk sampling methods and even less for laser ablation techniques), there are ways of minimizing one’s impact on archaeological materials when selecting specimens for sampling. In the case of tooth enamel, researchers should attempt to sample teeth that are no longer embedded in the alveolar bone if possible as this reduces the risk of damage to the surrounding alveolus and bones of the skull. When selecting bone for analysis, researchers should avoid sampling intact bone if fragmentary bone is available for study, so as to minimize impact to the skeleton. Additionally, scientists should try to avoid removing

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**Fig. 35.5** Sequential sampling of a Neanderthal tooth using laser ablation. Numerical values represent the $^{87}\text{Sr} /^{86}\text{Sr}$ ratios for the individual laser-ablation pits. Reprinted from Richards et al. (2008), with permission from Elsevier.
enamel or bone samples from those teeth or parts of the skeleton that exhibit pathological lesions, cultural modifications, or other diagnostic markers as these features potentially can be used to reconstruct aspects of health, diet, growth, and socio-cultural practices among ancient populations (see e.g., Larsen 1997; Katzenberg and Saunders 2008). Finally, since multiple isotopic signatures can be retrieved from a single dental or bony element, scientists should formulate hypotheses and devise their sampling strategy so as to maximize the kinds of chemical information that can be retrieved from their samples while minimizing damage to the skeleton.

### 35.2.3 Laboratory Analysis

Radiogenic strontium isotope analysis is a well-established procedure and has long been used by geologists. Accordingly laboratory sample preparation procedures and analytical techniques have been thoroughly studied and routinely applied (see e.g. Faure and Mensing 2005). Here we briefly summarize preparation protocols relevant for archeological studies and provide some information on mass spectrometric analyses.

#### 35.2.3.1 Preparation Protocols for Isotopic Analysis of Bedrock, Soil, and Groundwater

To determine the strontium isotopic composition of environmental samples that represent the local provenance signature, care must be given to collect relevant and representative samples. As discussed earlier, such samples may include bedrock, soil, groundwater, river/lake water or vegetation, as well as archaeozoological and archaeobotanical materials (the seawater signature is well established and does not need to be analyzed). Several sub-samples from different, randomly spread locations relevant to the study should be collected and homogenized. Archaeobotanical remains should be carefully cleaned of dust by ultrasonication in milliQ water and acetone several times, while archaeozoological materials should be prepared for analysis following the protocols outlined for human samples below.

Homogenized samples are dissolved in preparation for strontium separation for mass spectrometry. Many sample dissolution methods have been used ranging from an acid pre-leach followed by partial dissolution in weak acid (McArthur et al. 1993) to a total dissolution in mixed strong acids (Hein et al. 1993). The main purpose of the pre-leaching is to minimize contamination by non-target components which may adhere to the sample surface. Such cleaning should be included if diagenetic contamination is expected (Bailey et al. 2000). The pristine, clean, representative target samples should then be fully dissolved. The exact procedure for sample dissolution will depend on the properties of the sample. For example, pure carbonate rocks may be digested by relatively dilute acetic acid (Montanez et al. 1996), while bulk silicate rocks and soil are typically dissolved in a mixture of hot concentrated HNO₃ and HF acids followed by drying and repeat treatment with HCl and HNO₃ (Billings and Adams 1964). Organic substances like vegetation are either oxidized in hot HNO₃ or ashed for 4 h in a muffle furnace at 550°C and then dissolved in a mixture of 6 N HNO₃ and HCl (Porder et al. 2003), followed by removal of any residue through filtration. After dissolution, samples are dried down and then re-constituted in a minimal amount of acid (typically HCl or HNO₃) in preparation for strontium separation by ion chromatography. Water or other fluids containing strontium are filters and the appropriate volume containing enough strontium for analysis is dried down and re-constituted as above. All reagents used for dissolution must be trace metal clean or distilled to lower strontium blanks.

#### 35.2.3.2 Preparation Protocols for Isotopic Analysis of Archaeological Human Remains and Modern Fauna

Pretreatment protocols for archaeological specimens will vary depending on the element analyzed. As discussed earlier, tooth enamel is largely resistant to diagenetic contamination while archaeological bone is highly susceptible to post-mortem alteration. As a result, pretreatment protocols for tooth enamel are relatively straightforward and less intensive than that required for archaeological bone specimens.

While various preparation methods exist for archaeological tooth enamel, the general consensus is that
sequential rinses or an overnight bath in weak (≤1.0 N) acetic acid remove most diagenetic carbonates in enamel (Sillen 1986; Hoppe et al. 2003). It should be noted that samples can lose up to 70% of their weight depending on the specific preparation protocol followed (Hoppe et al. 2003); thus, care should be taken in determining the number of rinses enamel powder should be subjected to. Once acetic acid has been removed from enamel samples, the residual powder should be rinsed with distilled/deionized water and sonicated three times for at least 5 min. Samples can be left overnight to dry or dried down using a desiccator, hot plate, or oven (below 50°C).

Archaeological bone samples should be subjected to more rigorous preparation procedures than those established for enamel, although it should be emphasized that no one pretreatment protocol has been found that completely separates biogenic and diagenetic strontium in bone (Koch et al. 1997; Hoppe et al. 2003; Trickett et al. 2003). Many studies have adopted Sillen’s (1986) solubility profile method (or a version thereof), in which powdered bone is subjected to a series of washes in 0.1 N buffered acetic acid (pH = 4.5). Following acid treatment, bone samples should be rinsed and sonicated three or more times using distilled/deionized water for several minutes. Given the large number of rinses (Sillen’s original 1986 study consisted of 24 acid washes), it is recommended that powdered bone samples initially weigh between 30 and 50 mg, as significant amounts of sample can be lost during preparation. As an alternative to Sillen’s method, Knudson et al. (2004) recommend sonication of chunked bone in deionized water for 30 min, followed by sonication in 5% acetic acid for 30 min, and a second aliquot of 5% acetic acid for 5 min. Once rinsed, bone samples can be ashed and powdered.

Since diagenesis is not an issue when analyzing bones and teeth from modern fauna, enamel and/or bone from these specimens require minimal pretreatment prior to analysis. Mechanical cleaning is recommended for all modern samples, and if necessary, samples can be left overnight in H₂O₂ (30%) to remove organics. Cleaned samples should then be sonicated with distilled water and dried down. All dried samples (both archaeological and modern tooth enamel and bone) can then be dissolved in either HCl or HNO₃ in preparation for strontium separation by ion chromatography.

### 35.2.3.3 Strontium Separation

Prior to mass spectrometric analysis strontium has to be purified to reduce mass interference (isobaric interference) by other elements and compounds (specifically ⁸⁷Rb) and to maximize ionization efficiency (residual Ca decreases the ionization efficiency of strontium and the stability of the ion beam). This is done using ion chromatography by passing the sample solution through columns filled with cation-exchange resin (Hart and Brooks 1974). About 1 µg of strontium is typically prepared for isotope analysis. The most commonly used separation technique involves cation-exchange chromatography using resin (AG 50W-X8) in HCl medium. This resin retains strontium more strongly than singly charged alkali metals including major elements such as Mg and trace elements such as Rb (Hart et al. 1974). However the separation from Ca and Al can be compromised when these are present in higher concentrations relative to Sr. Use of longer columns can alleviate some of these problems (Birck 1986). Columns are conditioned with 3–5 column volumes of 0.75 N HCl and the sample (dissolved in a small amount of 0.75 N HCl) is loaded. The cations are eluted with 1.8 or 2 N HCl (depending on the acid strength used for calibration). The strontium fraction is collected and this fraction is dried down in preparation for mass spectrometry. Columns should be calibrated routinely to ensure clean separation and efficient recovery of Sr.

Strontium can also be extracted from nitric acid media using crown ether in octanol, sorbed on an inert substrate (Sr-Spec, Eichrom® non-ionic ester polymer resin, 100–150 mesh) (Horwitz et al. 1991, 1992). This method is more selective for strontium and strontium does not break through from the columns until about 30 column volumes of 3 M HNO₃. Strontium is eluted using 0.05 M HNO₃. Using Sr-Spec is very effective in separating rubidium and calcium from Sr, although barium is not as easily removed and longer columns or stronger acid should be used for barium-rich samples (Pin and Bassin 1992). Strontium blanks of the Sr-Spec columns are typically higher than those of the AG 50W-X8 resin, particularly when using columns repeatedly, thus care should be taken to monitor blanks or to avoid re-use of columns (Pin and Bassin 1992). All reagents used for strontium separation should be trace metal grade or
distilled to ensure low blanks, and procedural blanks should be monitored routinely.

### 35.2.3.4 Instrumentation

Several options are available for mass spectrometric analysis including solid-source thermal ionization mass spectrometry (TIMS), solution quadrupole-based ICP-mass spectrometry (ICP-QMS) (Vanhaecke et al. 1999), or multi collector ICP-mass spectrometry (MC-ICPMS) (Waigh et al. 2002). The MC-ICPMS could also be coupled to laser ablation (LA-MC-ICPMS) (Christensen et al. 1995; Vroon et al. 2008). TIMS has been used for longer than the other instruments and is the most routinely used procedure for strontium isotope analyses. Although using the TIMS is time consuming (each analysis could take 1–2 h) it requires less strontium compared to other solution-based analyses and has the highest reproducibility and precision.

For TIMS, following strontium purification, the samples are dried down, re-dissolved in 1% HNO₃, and loaded with 0.1 M H₃PO₄ on degassed Re or Ta filaments. Samples as small as 20 ng of strontium can be routinely analyzed using TIMS although larger samples are desirable if sample is available. The isobaric interferences of $^{87}$Rb are determined by monitoring $^{85}$Rb and applying corrections as needed. All $^{87}$Sr/$^{86}$Sr values are corrected for mass fractionation in the instrument using $^{86}$Sr/$^{88}$Sr = 0.1194 (this correction is also referred to as normalization). NIST SRM 987 strontium carbonate standard is always analyzed along with samples, and the value of this standard is used to determine long-term external precision as well as for comparison of data between laboratories. The accepted value of the SRM 987 standard is 0.71024 and data is either corrected for offset from this value or the value obtained for SRM 987 is reported along with the data. Typical internal precision on the mass spectrometer is around 0.000010, and the external precision is approximately ±0.000020 (Oslick et al. 1994) (2σ standard deviation).

Techniques for radiogenic strontium isotope analysis on a double-focusing multiple collector inductively coupled plasma mass spectrometer (MC-ICPMS) can be done on unspiked or spiked samples (Fortunato et al. 2004). Measurement protocol includes standard–sample–standard bracketing methods using the isotopic reference material (SRM 987) with wash solutions between sample or standard introductions. This permits internal correction of the isotope ratios $^{87}$Sr/$^{86}$Sr and $^{84}$Sr/$^{86}$Sr with $^{88}$Sr/$^{86}$Sr and lowering of memory effects from preceding samples. Using MC-ICPMS can yield accurate and precise results similar to those obtainable by TIMS and the procedure is faster (about four samples per hour). This procedure, however, requires a larger amount of strontium (at least 300 ng per sample) in comparison to TIMS. Moreover, backgrounds should be measured to correct for interferences on masses 84 and 86 derived from small amounts of krypton in the argon supply in the plasma, and stable, residual memory strontium and rubidium signals from sample material on the torch and cones. Simultaneous ionization of all elements during plasma-based ionization also proves to be a disadvantage during radiogenic strontium isotope analysis as extra chemical clean-up steps, beyond those necessary for TIMS analyses, are required to remove potential isobaric interference. LA-MC-ICPMS strontium isotope ratio measurements of various geological and biological samples allow analyses of small samples and fine spatial resolution with minimal preparation. However significant deviations in the $^{84}$Sr/$^{86}$Sr ratio and the radiogenic $^{87}$Sr/$^{86}$Sr ratio from values obtained by analysis of strontium chemically separated from the sample matrix have been reported (Vroon et al. 2008). The precise reasons for this remain unclear but likely reflect a combination of isobaric interferences from Ca dimers and Ca argides and doubly charged REE, as well as disruption of mass bias effects due to differential loading of the plasma.

It is also unclear whether this is an instrument- or technique-specific problem and great care should be taken when attempting strontium isotope analyses by in situ LA-MC-ICPMS techniques (García-Ruiz et al. 2008; see also Section “Tooth Enamel,” this chapter, for a related discussion).

### 35.2.3.5 Monitoring for Diagenesis

The identification and removal of diagenetic contamination are crucial steps in radiogenic strontium isotope analysis of archaeological materials. As discussed above, the elimination of post-depositional contaminants from bone and enamel can be achieved through mechanical cleaning at the time of sample collection...
and through chemical cleaning prior to sample dissolution. Even with these pretreatment procedures in place, however, it is difficult to be sure that all diagenetic contamination has been successfully removed. Several methods for monitoring diagenetic contamination have been established (Sillen 1989; Price et al. 1992; Nielsen-Marsh and Hedges 2000a, b), and a few techniques are briefly summarized here.

Changes to the mineralogy and crystallinity of enamel and bone can be monitored using either X-Ray Diffraction (XRD) or Fourier Transform Infrared Spectroscopy (FTIR). In general, crystallinity is higher in altered apatite than in pristine apatite (Sillen 1989; Shemesh 1990). The degree of crystallinity can be measured by calculating the crystallinity index (CI). On an infrared spectrum, the CI is the extent of phosphate peak splitting at 565–605 cm\(^{-1}\) and is measured by calculating the relative depth of the valley between the two peaks (Shemesh 1990). Samples that exhibit CI's above the range established for fresh bone, 2.8–3.1 (Weiner and Bar-Yosef 1990; Wright and Schwarcz 1996; White et al. 1998; Garvie-Lok et al. 2004), likely have been altered.

Diagenetic contamination in enamel and bone also can be detected by comparing levels of trace elements in archaeological samples with those in modern specimens (Kohn et al. 1999; Hoogewerff et al. 2001; Price et al. 2002). The ratio of calcium to phosphorus (Ca/P), for example, is commonly used to monitor potential alteration in archaeological materials. Modern, unaltered bone contains approximately 37% Ca by weight and 17% P, with a Ca/P ratio of about 2 (Kyle 1986). Ca/P ratios much greater than 2, therefore, indicate contamination. Additionally, scientists have measured carbonate/phosphate ratios (C/P) in archaeological specimens (Nielsen-Marsh and Hedges 2000a), as well as the concentration of uranium (U) in archaeological bone samples (Kohn et al. 1999; Knudson and Price 2007; Conlee et al. 2009) in order to monitor diagenetic contamination.

### 35.3 Applications

The number and diversity of strontium isotope studies in archaeology have increased dramatically over the last two decades. The technique primarily has been used to differentiate between individuals and fauna of local and non-local origin, although a few studies have adopted the method as a means of identifying and sourcing non-local building materials, artifacts, and foodstuffs. While not exhaustive, the present section highlights some of the major applications of radiogenic strontium isotope analysis in archaeology to date (Table 35.2).

#### 35.3.1 \(^{87}\text{Sr}/^{86}\text{Sr} \) Analyses of Archaeological Humans

The majority of strontium isotope applications have focused on analyses of human skeletons from archaeological sites. While \(^{87}\text{Sr}/^{86}\text{Sr} \) values in human bones and teeth have been used primarily to identify immigrants and track the degree of residential mobility in the past, archaeologists have applied \(^{87}\text{Sr}/^{86}\text{Sr} \) data to address broader socio-cultural questions pertaining to imperial strategies and colonization, marital residence patterns, ethnicity and identity, and violence and warfare.

Ancient complex societies adopted various political strategies to manage their vast territories, including the establishment of colonies. Traditionally, archaeologists have used the presence of foreign, imperial-style ceramics, textiles, architecture, and burial practices as proxies for population movement and imperial control. With the introduction of radiogenic strontium isotope analysis to archaeology, scientists have been able to directly document the presence of foreigners at ancient sites and, thus, explore colonization models more effectively.

A number of such studies has been conducted in the Andean region of South America, including Kelly Knudson and colleagues’ work (Knudson et al. 2004; Knudson and Price 2007; Knudson 2008) on residential mobility within the Tiwanaku polity during the Middle Horizon (550–1000 AD). In a pioneering study, Knudson et al. (2004) identified a number of migrants at the eponymous capital of Tiwanaku in Bolivia, as well as two immigrants, likely from Tiwanaku, at the site of Chen Chen in Peru. The latter site had been interpreted as a Tiwanaku colony based on the presence of Tiwanaku-style artifacts, but prior to Knudson et al.’s work, no definitive evidence for Tiwanaku migration was available.

Similar research has been carried out by Slovak (2007) and Slovak et al. (2009) on the nature of Wari
<table>
<thead>
<tr>
<th>Region</th>
<th>Time period</th>
<th>Additional isotopes used</th>
<th>Primary organism(s) or materials</th>
<th>Site(s), country</th>
<th>References</th>
</tr>
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<tbody>
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<td>Human</td>
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<td></td>
<td>Neolithic</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>Neufahrn/Donaun, Bavaria</td>
<td>Bentley et al. (2003)</td>
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<td></td>
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<td>West Herskott, England</td>
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<td></td>
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imperial influence at the site of Ancón on Peru’s Central Coast. While some scholars have suggested that the site functioned as an important Wari outpost, other researchers have argued that Ancón remained free of Wari political and economic control. While the results of Slovak et al.’s (2009) radiogenic strontium isotope study do not resolve the debate entirely, the presence of a migrant, likely from the Wari heartland, coupled with Wari-style objects at the site suggests that the identification of Ancón as a Wari colony is at the very least plausible.

Radiogenic strontium isotope research on Late Horizon (1400–1532 AD) sites in the Andes has shed light on Inka political strategies as well, including the state-mandated resettlement of individuals into mitima communities (D’Altroy 2005). Andrushko et al. (2009), for example, noted a dramatic increase in the number of migrants at the Inka site of Chokepukio, Peru during the Late Horizon, which the authors partially attribute to coerced migrations resulting from Inka imperial labor policies. On the other hand, a multi-isotopic study by Turner et al. (2009) has shown that the famed site of Machu Picchu likely did not function as a mitima colony composed of relocated colonists from a common geographic area or areas, but rather was maintained as a private estate for the Inka emperor by elite retainers whose geographic origins were widely dispersed.

In the Old World, strontium isotope data also has been used to investigate colonization. Buzon et al. (2007), for example, examined radiogenic strontium isotope signatures among individuals buried at the site of Tombos in ancient Nubia during the Egyptian New Kingdom Period – a time when the Nubian Kingdom was subsumed under Egyptian rule. As noted by the Buzon et al. (2007), the nature and extent of Egyptian control over Nubia remains unclear; however, strontium isotope data indicates that colonial rule at Tombos may have been administered not only by individuals of foreign origin (i.e., Egyptians) but by local Nubians as well.

Montgomery et al. (2005) used both strontium and lead isotopes to explore the origins of potential immigrants at an Anglian cemetery in England during the fifth to seventh centuries AD, a time period that partially overlaps with the mass migration of Anglo-Saxons into Britain. While the geographic origins of the individuals identified as non-local based on their $^{87}\text{Sr}/^{86}\text{Sr}$ signatures in Montgomery et al.’s (2005) study could not be assigned definitely to Angeln, the authors’ results highlight the potential of strontium isotope applications to elucidate the nature and extent of colonization and mass migrations in the archaeological record.

Finally, extensive evidence for human migration has been documented via radiogenic strontium isotope analysis during the early and late Neolithic periods of Europe (see e.g., Bentley et al. 2003, 2004; Price et al. 2004, 2006a, b; Nehlich et al. 2009). Price et al. (2004), for example, employed radiogenic strontium isotope analysis to determine whether the spread of Bell Beaker culture throughout many parts of Europe during the Neolithic/Bronze Age transition resulted from the migration of foreign peoples into these areas, or from the importation of Bell Beaker-style artifacts into communities by local peoples. The results of their study, which included skeletons from Bell Beaker graves in Austria, the Czech Republic, Germany, and Hungary, demonstrate that 51 of 81 individuals analyzed (61.7%) were born elsewhere. While Price et al.’s (2004) data does not entirely resolve the question of whether migration or diffusion was responsible for the rapid and widespread expansion of the Bell Beaker culture at this time, their work underscores the considerable degree of human mobility that did occur during the Neolithic and provides an alternative line of evidence for evaluating the Bell Beaker phenomenon.

Radiogenic strontium isotope analysis also has yielded potential evidence for exogamous marriage practices in the past, particularly among females. Grupe et al. (1997) and Price et al. (1998) documented a disproportionate number of migrant females at Bell Beaker period (2500–1900 BC) sites in Bavaria, which they interpret as tentative evidence for exogamy. Similarly Schweissing and Grupe (2003) noted a similar trend among southern Bavarian populations during the Roman-period occupation. In this latter case study, approximately 56% of females analyzed (or 10 out of 18 women) yielded non-local $^{87}\text{Sr}/^{86}\text{Sr}$ enamel signatures compared to 37% of males (or 15 of 41 men). These results potentially suggest that females were married into the community at a greater rate than men. Elsewhere in Europe, Bentley et al. (2004) documented significantly more female immigrants than male immigrants at Early Neolithic cemeteries in Germany, which may indicate that larger number of females married into low-lying agricultural communities during this time period.
In some cases, strontium isotope data from human skeletons has been used to elucidate the relationship between ethnic identity and geographic origin. Price et al. (2000), for example, analyzed $^{87}\text{Sr}/^{86}\text{Sr}$ values from a sample of individuals buried in various ethnic enclaves or *barrios* at the site of Teotihuacan in Mexico and discovered that the majority of people appear to have been born non-locally. In this instance, then, the presence of foreign cultural elements such as architecture, pottery, burial practices, and food preparation techniques within the *barrios* appear to have resulted (at least in part) from foreign peoples living there. In a similar vein, Bentley et al. (2003) noted a significant correlation between burial location and $^{87}\text{Sr}/^{86}\text{Sr}$ signatures at the site of Vaihingen, Germany. A greater percentage of non-local individuals (14 of 24), as determined by their $^{87}\text{Sr}/^{86}\text{Sr}$ values, were buried in the Neolithic ditch encircling Vaihingen than in the settlement itself (5 of 22 individuals) (Bentley et al. 2004). These findings potentially suggest that non-local individuals were viewed as socially and/or ethnically different and buried distinctly (Bentley et al. 2003). Finally, in an innovative study by Price et al. (2006a, b), $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from five individuals buried at a colonial cemetery in Campeche, Mexico indicate that they likely were born in West Africa. Importantly, four of these individuals appeared ethnically distinct prior to isotope analysis based on the type of dental modification they exhibited. Price et al.’s (2006a, b) study not only documents some of the earliest representatives of the pre-eighteenth century slave trade, but also highlights a correlation between biological indicators of identity, i.e., place of origin, and sociocultural indicators of identity, i.e., dental modification practices.

On the other hand, radiogenic strontium isotope data may not always accord well with sociocultural data. Expanding on research produced in an earlier pilot study (Price et al. 1994; Ezzo et al. 1997), Ezzo and Price (2002), noted that ethnic identities among individuals buried at Grasshopper Pueblo, Arizona often were blurred and not necessarily correlated with strontium isotope signatures. Similarly, Knudson and Torres-Rouff (2009) documented a complex relationship between isotope signatures and cultural identity among individuals buried at the site of Caspana in northern Chile during the Late Intermediate Period (1100–1400 AD). Based on potential ethnic markers such as cranial modification and burial styles, the Caspana individuals were distinct from neighboring groups in the region, as might be expected among a migrant community; however all but one of the individuals at Caspana was born and raised locally.

Finally, the use of strontium isotope data has been applied to investigations of violence and warfare in the archaeological record. The identity of trauma victims has long been of interest to archaeologists and physical anthropologists but has been relatively difficult to ascertain; radiogenic strontium isotope analysis has the potential to provide archaeologists with data regarding the geographic origins of these individuals in the past. Price et al. (2006a, b) analyzed $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from 22 of the 34 human skeletons uncovered in a Neolithic-period mass grave in Talheim, Germany. The results of their study indicate that the majority of the individuals were born locally, and lend support to the authors’ (Price et al. 2006a, b) interpretation that the burials probably represent members of a single community who were killed by another group. In a similar vein, recent applications of radiogenic strontium isotope analysis to victims of trophy head taking in the Andes, a practice in which the skull of an individual was obtained either through decapitation or by its removal from a corpse, has produced interesting and exciting results. In a study by Tung and Knudson (2008), 3 of 5 trophy heads found at the Wari site of Conchopata in Peru were identified as non-local using radiogenic strontium isotope analysis. The authors suggest that one possible interpretation for these results is that the non-local trophy heads were foreign enemies who had been taken captive and subsequently decapitated. On the other hand, strontium isotope data from Conlee et al. (2009) and Knudson et al. (2009) have established that trophy head taking among the ancient Nasca of southern Peru was a localized phenomena, in which trophy head victims likely were culled from local populations in the region.

### 35.3.2 $^{87}\text{Sr}/^{86}\text{Sr}$ Analyses of Archaeological Fauna

In addition to examining human mobility in the past, scholars have analyzed strontium isotope data from fauna to better understand animal migration patterns. While the technique has been widely applied to extant animal populations (van der Merwe et al. 1990; Koch...
et al. 1992, 1995; Chamberlain et al. 1997), the present section highlights radiogenic strontium isotope studies of archaeological fauna.

Hoppe et al.’s (1999) investigation of mammoth and mastodon mobility in the American Southeast during the late Pleistocene was one of the earliest applications of $^{87}\text{Sr}/^{86}\text{Sr}$ to prehistoric faunal populations. Hoppe et al.’s (1999) research demonstrated that neither mammoth nor mastodons engaged in large-scale migrations (>700 km), and highlighted significant variability in the extent and frequency of mobility between the two animal species. Subsequent radiogenic strontium isotope studies on the origins and migration patterns of Pleistocene- and Holocene-era North American fauna have been carried out by other scholars, including Porder et al. (2003), Hoppe (2004), and Feranec et al. (2007).

While the above investigations have improved researchers’ understanding of prehistoric animal behavior, radiogenic strontium isotope ratios from ancient fauna also have been used to elucidate human activity in the past. Towers et al. (2009), for example, recently investigated the origins of cattle and aurochs buried in two Bronze Age barrows in England. Hundreds of cattle remains had been found associated with human burials within the barrows, but little was known about the animals’ origins or the purpose(s) of their placement in the tombs. Radiogenic strontium isotope data from a sub-sample of the archaeological fauna indicated that while most animals were raised locally, two individuals had been born elsewhere, suggesting that long-distance exchange was undertaken by human populations at the time (Towers et al. 2009).

Radiogenic strontium isotope analyses of archaeological fauna also can shed light on ancient animal husbandry practices. Balasse et al. (2002) determined strontium isotope ratios from two archaeological sheep and one archaeological cow (along with C and N isotope ratios from these and other archaeological fauna) to investigate the plausibility of a seasonal mobility model for Late Stone Age pastoralists in South Africa. Their findings suggest that ancient fauna were not herded between the coast and hinterland seasonally, but rather stayed within one region for their entire lives or migrated at some point (rather than seasonally) during their lifetimes. In a more recent multi-isotopic study of ancient herding practices, Thornton et al. (2010) tentatively reconstructed prehistoric herding practices among human groups in the Osmore Valley, Peru using $^{87}\text{Sr}/^{86}\text{Sr}$ signatures (along with $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values) from archaeological camelid bone. Although radiogenic strontium isotopes were unable to differentiate between camelids potentially herded in mid- to lower-elevation zones, $^{87}\text{Sr}/^{86}\text{Sr}$ values were sufficiently different to distinguish between camelids pastured in the highland puna region and those herded in middle and lower elevation habitats.

### 35.3.3 $^{87}\text{Sr}/^{86}\text{Sr}$ Analyses of Building Materials, Artifacts, and Food

Similar to radiogenic strontium isotope studies of human and faunal remains, the origins of artifacts, archaeological building materials, and ancient foodstuffs have been traced using $^{87}\text{Sr}/^{86}\text{Sr}$ analysis. For example, Freestone et al. (2003) reconstructed the potential origins and raw materials used to create sixth to eleventh century AD glass found in the Eastern Mediterranean region by determining $^{87}\text{Sr}/^{86}\text{Sr}$ values from four production sites in Israel and Egypt. Similarly, Henderson et al. (2005) utilized radiogenic strontium isotopes (along with oxygen and lead isotopes) to trace the potential geologic sources of Syrian glass from the eighth to ninth centuries AD. Elsewhere in the Old World, radiogenic strontium isotope analysis has been applied to shell beads from the Paleolithic La Madeleine child burial in France to determine their origin (Vanhaeren et al. 2004). $^{87}\text{Sr}/^{86}\text{Sr}$ results indicate that at least some of the shells were collected from faraway beaches rather than nearby outcrops. The authors (Vanhaeren et al. 2004) argue that prehistoric artisans likely preferred beach shells to shells found in neighboring Miocene outcrops because the former type was morphologically more compatible with Paleolithic bone needle technology.

The origins of textiles, particularly those made from plant materials, can be elucidated using radiogenic strontium isotope analysis. Benson et al. (2006), for example, employed $^{87}\text{Sr}/^{86}\text{Sr}$ analysis (as well as oxygen isotopes) to successfully trace the origins of raw materials used in textile manufacture in the American Southwest. In a more recent study, the feasibility of radiogenic strontium isotope analysis to track the origins of ancient woolen textiles was recently tested by Frei et al. (2009) using modern sheep hair from Scandinavian specimens. The authors determined that
with appropriate pretreatment procedures, $^{87}\text{Sr}/^{86}\text{Sr}$ values from hair potentially can be used to source woollen materials in the archaeological record (Frei et al. 2009).

In addition to artifacts, the sources of various building materials have been identified via radiogenic strontium isotope analysis. At various archaeological sites in the Mediterranean, Gale et al. (1988) used sulfur and strontium isotopes to trace the geologic sources of gypsum – a material used in internal construction during Mycenaean times. Similarly, Brilli et al. (2005) determined $^{87}\text{Sr}/^{86}\text{Sr}$ values for eight Mediterranean quarry areas which were in use during classical times. While some of the quarries exhibited overlapping $^{87}\text{Sr}/^{86}\text{Sr}$ values, the authors demonstrate that radiogenic strontium isotope data, in tandem with other geochemical and petrographic applications, can be used to determine the potential provenance of architectural and sculptural marble artifacts (Brilli et al. 2005). At Chaco Canyon, New Mexico, Reynolds et al. (2005) determined $^{87}\text{Sr}/^{86}\text{Sr}$ data for multiple specimens of ponderosa pine used in the construction of buildings at various Great Houses. Reynolds et al.’s (2005) results indicate that Chaco’s ancient inhabitants sourced their timber from a number of different outcrops, many of which were located at considerable distances from archaeological sites.

Finally, radiogenic strontium isotope analysis has been used to investigate the origins of food stuffs in the archaeological record. Recently, Benson et al. (2009) and Benson (2010) have determined $^{87}\text{Sr}/^{86}\text{Sr}$ values for maize found at sites in Chaco Canyon and Aztec Ruin, New Mexico, which in turn have allowed the authors (Benson et al. 2009; Benson 2010) to speculate on changing trade relationships in the ancient American Southwest.

### 35.4 Future Directions

Radiogenic strontium isotope analysis in archaeology has come a long way in the last 25 years, but more work remains to be done. The distribution of radiogenic strontium isotope applications thus far has been limited to several key areas in space and time, including the Neolithic in central Europe, Great Britain, and Southeast Asia; the Late Stone age in South Africa; early colonization of the Pacific; the Pleistocene/Holocene transition in the central and Southeastern U.S.; and the development of complex societies in the American Southwest, Mesoamerica, and the Middle though Late Horizons in the Andes of South America. Many areas and time periods, including the hominin fossil record, remain relatively unexplored using radiogenic strontium isotope analysis, although the few studies that have been conducted indicate enormous promise.

Sillen et al. (1995), for example, measured $^{87}\text{Sr}/^{86}\text{Sr}$ values from ~1.8 ma *Homo* sp. and *Australopithecus robustus* skeletons at the site of Swartkrans in South Africa, and (as discussed earlier) Richards et al. (2008) determined $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from a 40,000 ya Neanderthal tooth found in Greece. Additionally, Sillen et al. (1998) compiled $^{87}\text{Sr}/^{86}\text{Sr}$ isotope data for the Sterkfontein Valley of South Africa using water, soil, and plant samples and compared these values to $^{87}\text{Sr}/^{86}\text{Sr}$ from Pleistocene-era vertebrates. All three studies have shed light on hominin diet and potential mobility patterns, although future applications of radiogenic strontium isotope analysis to fossil hominids may be hindered by the rare nature of the specimens and the potential for diagenetic contamination. To this end, the recent work by Copeland et al. (2008, 2010) that demonstrates the non-destructive nature of sampling via laser-ablation should prove critical.

In those regions of the world where radiogenic strontium isotope analysis has been applied, more work should be done to define local $^{87}\text{Sr}/^{86}\text{Sr}$ signatures. Ideally, such reconstructions would entail the collection of samples from exposed bedrock, soil, plants, and water along with small fauna and archaeological human remains from the region. Hodell et al.’s (2004) study of the Maya region, which involved the collection of 216 samples of rock, soil, water, and plants from Mexico, Honduras, and Guatemala, provides a good model for other researchers to follow (Fig. 35.6), as does Hedman et al.’s (2009) recent research on bio-available $^{87}\text{Sr}/^{86}\text{Sr}$ values for the Midwestern United States. In this latter study, local $^{87}\text{Sr}/^{86}\text{Sr}$ signatures were established using the remains of nearly 50 ancient fauna from 14 archaeological sites in Illinois, Iowa, Indiana, and Missouri (Fig. 35.7). Both Hodell et al. (2004) and Hedman et al.’s (2009) research demonstrate the effectiveness and utility of collecting numerous and varied environmental samples from broad geographic regions and major geologic zones.

As discussed in the previous section, the ability of radiogenic strontium isotope analysis to source food
stuffs at ancient sites primarily has been used to examine archaeological models of prehistoric trade, exchange, and technology. Radiogenic strontium isotopes from ancient foodstuffs, however, also can be used to monitor the reliability of archaeological human $^{87}\text{Sr}/^{86}\text{Sr}$ values. The effectiveness of $^{87}\text{Sr}/^{86}\text{Sr}$ data as an indicator of migration depends upon the premise that ancient humans primarily ate locally-grown foods; however even foods that appear to have been cultivated locally may have been brought in from elsewhere or treated with foreign additives during food preparation (see e.g. Wright 2005). Analyzing $^{87}\text{Sr}/^{86}\text{Sr}$ values from food found in archaeological contexts (including those resources presumed to have been grown or raised locally), would better refine archaeological and isotopic interpretations.

The inclusion of additional isotopic tracers to radiogenic strontium isotope applications in archaeology also can facilitate interpretation of trends observed, and already has been alluded to in this chapter. Briefly, carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) isotope analyses of human remains can trace the relative contributions of $\text{C}_4$, $\text{C}_3$, and marine-based foods in an individual’s overall diet (DeNiro and Epstein 1978, 1981; Schoeninger and DeNiro 1984; Schwarcz and Schoeninger 2011) and thus can be used to identify potential dietary biases that might affect strontium isotope signatures. Similarly, oxygen isotope data in human tissue ($\delta^{18}\text{O}$),
which is dependent on a variety of climatic factors (Gat 1980; Yurtsever and Gat 1981; White et al. 1998), provides an alternative, independent marker of migration.

Finally, new research by Knudson et al. (2010) demonstrates that the relationship between $^{88}\text{Sr}$ and $^{86}\text{Sr}$ (expressed as $\delta^{88}\text{Sr}/^{86}\text{Sr}$ and reflecting mass dependent fractionation) also can be significant to archaeological studies. According to Knudson et al. (2010), $\delta^{88}\text{Sr}/^{86}\text{Sr}$ varies by trophic level, such that in terrestrial ecosystems $\delta^{88}\text{Sr}/^{86}\text{Sr}$ decreases as one moves from bedrock and soils to points further along the food web, while in marine environments $\delta^{88}\text{Sr}/^{86}\text{Sr}$ is highest in seawater and lowest in carnivorous fish and marine mammals. Ideally, then, it should be possible to distinguish among different components in ancient diets by examining $\delta^{88}\text{Sr}/^{86}\text{Sr}$ data in human bone and tooth mineral. While still in its incipient stages, Knudson et al.'s research using $\delta^{88}\text{Sr}/^{86}\text{Sr}$ data revealed dietary variability among archaeological Andean populations, and holds enormous promise for future archaeological studies.

35.5 Conclusion

The contributions of radiogenic strontium isotope analysis to archaeology thus far have been many. The provenience of artifacts, architectural elements,
and food stuffs can be sourced using this method, and the social and cultural implications of such findings explored more readily and reliably by archaeologists. Similarly, $^{87}\text{Sr}/^{86}\text{Sr}$ data from the human skeleton potentially provides researchers with direct evidence for an individual’s geographic origin and a means to measure the degree of residential mobility within his or her lifetime. These results, in turn, have been used to reexamine models of migration, colonization, marital patterns, warfare, ethnicity, and cultural identity in past societies.

Despite these successes, however, the interpretation of $^{87}\text{Sr}/^{86}\text{Sr}$ data is hardly straightforward. While the technique is often employed as a way of differentiating between migration and diffusion in the archaeological record (see e.g., Knudson et al. 2004; Price et al. 2004; Slovak et al. 2009), the presence of foreign individuals at sites, even when accompanied by foreign-style artifacts, should be viewed cautiously. In most cases, even with evidence for migration, scholars cannot rule out the possibility that other social processes such as trade and exchange or competition between elites in a local community for exotic goods may have played an equally significant part in instigating widespread cultural change. Similarly, there may be any number of reasons to explain why an individual relocated to a site during his or her lifetime. Simple radiogenic strontium isotope models that would equate foreigners with colonizers, for example, can be just as problematic as earlier archaeological approaches that equated archaeological cultures with peoples (see e.g., critique in Jones 1997).

In addition to these theoretical challenges, various aspects of strontium isotope methodology remain problematic. For example, no single pretreatment protocol (or series of protocols) is used exclusively to prepare archaeological samples for analysis (see Sects. 35.2.3.1 and 35.2.3.2 and references therein), nor is there a consensus as to whether biogenic $^{87}\text{Sr}/^{86}\text{Sr}$ values can be retrieved from archaeological bone (Sillen 1986; Koch et al. 1997; Hoppe et al. 2003; Trickett et al. 2003). Similarly, no one method appears to be universally adequate for establishing a “local” $^{87}\text{Sr}/^{86}\text{Sr}$ signature (Price et al. 2002; Wright 2005), nor is there an absolute agreement on the reliability and comparability of LA-MS-ICP-MS-based $^{87}\text{Sr}/^{86}\text{Sr}$ data and traditional solution-based TIMS data (see e.g., Horstwood et al. 2008; Simonetti et al. 2008; Nowell and Horstwood 2009; Copeland et al. 2010). Current and future radiogenic strontium isotope research surely will help to resolve some of these issues; in the interim, $^{87}\text{Sr}/^{86}\text{Sr}$ analysis remains a powerful tool with which to critically examine the archaeological record.

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**References**


