Patterns of paleomobility in the ancient Antilles: an isotopic approach

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CHAPTER 3 ISOTOPE ARCHAEOLOGY

3.1 Introduction

In this chapter the basic principles of isotopic analysis of human remains are presented with a primary focus on those applications that are of most interest for studying human migrations and mobility from the archaeological record. Established biogeochemical methods for assessing human migration patterns based on the human skeleton can be divided into two broad analytical approaches. The first, elemental analysis, measures the absolute or relative concentrations of certain elements, such as strontium and barium, in skeletal tissues. The second, isotopic analysis, measures the isotopic composition (isotopic ratios), of specific elements within human tissues. This chapter covers the main principles and applications of isotope analyses to human provenance studies, with an emphasis on strontium isotope analyses of ancient human remains.

Strontium isotope ratios are the most commonly analyzed isotopes for archaeological provenance studies but oxygen and lead isotopes, and more recently sulfur, have also been utilized for these purposes. Carbon and nitrogen isotopes have been traditionally employed towards dietary studies. Unlike light stable isotopes such as carbon and nitrogen, heavy elements (with a higher atomic mass, such as strontium) do not undergo substantial fractionation during most naturally occurring physical and biochemical processes [although see (Knudson et al. 2010b)]. In other words, the mass differences between different isotopes of a given heavy element are much smaller than the mass differences between the different isotopes of lighter elements. This means that the isotopes of a heavy element tend to be less influenced by mass dependent effects. For strontium, even if some minor biofractionation would take place this would be corrected for via the application of a mass correction factor to the 'natural' ratio of $^{86}Sr/^{88}Sr$ = 0.1194. Thus, isotopic signatures of strontium within biogenic tissues are directly reflective of the local sources of biologically available Sr in a given ecosystem or

environment rather than biochemical processes, such as metabolic pathways. Therefore, analysis of strontium isotope ratios in biogenic tissues is an effective tool for provenance studies, while carbon and nitrogen isotopes are more informative of dietary patterns.

3.2 Strontium Isotopes

Atoms that have the same the same number of protons (*atomic number* or *Z*) but different numbers of neutrons (*neutron number* or *N*) are called isotopes. "The isotopes of an element have identical chemical properties and differ only in their masses" (Faure and Mensing 2005). Variations in the isotopic compositions of certain elements are the result of two distinct processes: "1. the spontaneous decay of the nuclei of certain atoms to form stable nuclei of other elements and the accumulation of these radiogenic daughter atoms in the minerals in which they formed and 2. the enrichment (or depletion) of certain stable atoms of elements of low atomic number in the products of chemical reactions as a result of changes in state such as evaporation and condensation of water during physical processes such as diffusion" (Faure and Mensing 2005:3). The first of these processes accounts for most variations in isotopic ratios amongst the heavier elements, such as strontium, and the latter accounts for much of the variation in isotopic ratios amongst lighter elements such as carbon, nitrogen and oxygen.

"Strontium is a member of the alkaline earths of group IIA, which consists of Be, Mg, Ca, Sr, Ba, and Ra. The ionic radius of Sr^{2+} (1.13 Å) is slightly larger than that of Ca^{2+} (0.99 Å), which it can replace in many minerals" (Faure and Mensing 2005:75). There are four naturally-occurring isotopes of strontium (^{84}Sr, ^{86}Sr, ^{87}Sr, and ^{88}Sr) with isotopic abundances of approximately 0.56, 9.87, 7.04, and 82.53 percent respectively (Faure and Mensing 2005:3). All four of these are stable, although ^{87}Sr is radiogenic, being produced by the radioactive decay of ^{87}Rb, with a half-life of ~4.88 X 10^{10} years (Faure and Mensing 2005:3). Globally, the ratio of $^{87}Sr/^{86}Sr$ is variable and it varies according to three main factors, "(1) the $^{87}Sr/^{86}Sr$ at time the rock crystallized, $t = 0$, (2) the $^{87}Rb/^{86}Sr$ ratio, which is directly proportional to the Rb/Sr ratio in most cases, and (3) the time *t* elapsed since formation." (Bentley 2006:137).

3.2.1 Strontium Isotopes in Natural Systems

The primary sources of strontium into any particular ecosystem can be divided into geological and non-geological sources. In most contexts, the geological source is the largest contributor of strontium into plant tissues (Bentley 2006:153; Bern et al. 2005) through the primary weathering of soil minerals. However, research into various environmental systems indicates that atmospheric contributions of strontium can be quite substantial (Aberg et al. 1998; Dijkstra et al. 2003; Kennedy and Derry 1995; Kennedy et al. 1998; Kennedy et al. 2002; Pozwa et al. 2000; Pozwa et al. 2002; Pozwa et al. 2004; Vitousek et al. 1999). Non-geological sources of Sr include both the atmosphere and hydrosphere and a significant non-geological source of strontium in coastal areas comes from the sea (Hodell et al. 2004; Montgomery et al. 2003). Marine effects can be derived from sea-spray or marine-derived strontium in rainwater (Chadwick et al. 1999; Kennedy et al. 1998; Whipkey et al. 2000), or through the direct or indirect consumption of marine resources by animal organisms, including humans (Bentley et al. 2005; Price and Gestsdóttir 2006; Wright 2005). Atmospheric effects derive from many types of precipitation including not only rainfall but also atmospheric dryfall, the settling of wind-blown particulate matter (Biscaye et al. 1974; Muhs et al. 1990).

3.2.2 Strontium Isotopes- Geology

The major oceans of the Earth contain mid-oceanic ridges where erupting lava flows deposit large quantities of volcanic rock, producing ridges which sometimes extend above sea level forming oceanic islands, such as Iceland. These volcanic processes produce mid-oceanic ridge basalts or MORB, and they generally have low but somewhat variable $^{87}Sr/^{86}Sr$ ratios (Faure and Mensing 2005). Rocks derived from older continental crust, which has not been subducted and re-melted into the mantle for many hundreds of millions or billions of years, tend to have elevated $^{87}Sr/^{86}Sr$ ratios. This is because there has been sufficient time for any extant ^{87}Rb to decay into ^{87}Sr, thereby producing measurable differences in the quantity of ^{87}Sr relative to ^{86}Sr. Owing to the long half-life

of ^{87}Rb, the effects of this process are most noticeable in extremely old materials, for example Pre-Cambrian cratons. However, since age is not the only variable influencing $^{87}Sr/^{86}Sr$ ratios, some older crustal materials do not have highly radiogenic $^{87}Sr/^{86}Sr$ ratios. Formations containing very low initial Rb concentrations (relative to Sr) will not substantially elevate $^{87}Sr/^{86}Sr$ ratios even over very long periods of time (Faure and Mensing 2005).

Oceans contain sediments derived from the continents and transported by various physical processes. The Sr isotope ratio of modern seawater has been consistently measured and appears highly homogeneous. This is due to the fact that the global mixing of seawater occurs on the order of decades whereas changes significant enough to alter the global seawater composition occur at much greater timescales (Hess et al. 1986; Richter et al. 1992; Veizer et al. 1999). Therefore, the modern marine strontium signature is calculated as $^{87}Sr/^{86}Sr$ (oceans) = 0.70918 ± 0.00001 (2σ) (Faure and Mensing 2005; Howarth and McArthur 1997; McArthur et al. 2001; Veizer et al. 1999). In other words, the Sr ratio of the oceans has not changed appreciably at the time scales that concern most archeologists (i.e., thousands of years).

The marine $^{87}Sr/^{86}Sr$ ratio does fluctuate over geological time but this fluctuation or variation has been resolved to the extent that an approximation of the $^{87}Sr/^{86}Sr$ signal of a marine carbonate or limestone deposit can be derived from an estimate of the time period of its formation or deposition (Hess et al. 1986; Howarth and McArthur 1997; McArthur et al. 2001; Richter et al. 1992; Veizer et al. 1999). The $^{87}Sr/^{86}Sr$ ratio of seawater fluctuated considerably throughout the Phanerozic, varying between ~0.7068-0.7092 (Faure and Mensing 2005). Episodes of global volcanic activity have been proposed as possible causes for temporary decreases in oceanic $^{87}Sr/^{86}Sr$ ratios throughout this time period (Faure and Mensing 2005).

3.2.3 Strontium Isotopes- Hydrology

The Sr composition of freshwater reservoirs, such as lakes and rivers, reflects a cumulative average of all sources of Sr to a particular drainage. These sources include

atmospheric deposition and especially the movement of sediment from upstream sources by erosion. Analysis of suspended and soluble loads of freshwater indicates that their Sr isotope signals may vary both spatially and temporally within a single drainage system (Bentley 2006). As soluble and non-soluble components of soils may possess different Sr isotope ratios, analyzing both may contribute to tracking variable sediment sources for particular drainages (Probst et al. 2000).

Sedimentation rates and sources are in turn affected by a number of variables, including precipitation rates, vegetation cover, slope, and land usage to name a few. In most cases, the Sr signals of transported sediments in streams and rivers are considered to be representative of the weighted average of all upstream sources (Bentley 2006). If a particular drainage system passes over several isotopically distinct regions, it will probably contain sediments which possess variable Sr isotope ratios. These variable sources can be partly tracked by sampling a drainage system along its course and observing potential changes in Sr ratios throughout the system. In fact, analyses of Sr ratios from rivers in Europe have been utilized as a proxy for the local signals of specific drainage basins, under the not unrealistic expectation that most organisms subsisting within a certain basin or portion thereof should share similar Sr isotope compositions (Bentley and Knipper 2005; Hodell et al. 2004; Hoogewerff et al. 2001; Voerkelius et al. 2010).

While this appears to be a useful approach in continental settings, the usefulness of it in insular settings can be limited by several factors. In the Caribbean, many islands lack substantial permanent river systems, especially some of the flatter and more arid limestone islands. The larger islands of the Greater Antilles and many of the volcanic islands with high topographic relief generally receive much greater quantities of annual precipitation, producing seasonal or even year round drainages (Woods 1989; Woods and Sergile 2001). However, it is often these same places that have attracted human settlement and agricultural and industrial development over the years, which in turn tends to bring about large scale and widespread modification of the landscape and potentially profound alterations to the local hydrological cycle. This does not mean that sampling of freshwater sources has no utility in insular Caribbean settings, and in fact in some places such work might prove quite fruitful (Pett-Ridge et al. 2009). The previously mentioned

concerns merely limit the possibility of relying heavily on hydrological data sources throughout the entire macro-region.

3.2.4 Strontium Isotopes- Atmosphere

Strontium from the atmosphere can be divided into broad two categories; strontium in water vapor and strontium in dryfall. Sea-spray caused by wind blowing across the surface of the oceans can transport seawater, and the Sr that it contains, into the atmosphere. Atmospheric water vapor can then be transported by currents considerable distances before precipitating back to the surface of the Earth. Dryfall refers to the falling of wind-blown particles back to earth's surface and can be quite substantial in certain regions of the world especially during the dry season and in areas that are downwind from large sources of wind-transported sediment.

A well-known example of this is the transport and deposition of large quantities of Saharan dust across the Atlantic. Large 'clouds' of Saharan dust can even be observed from space and under favorable weather conditions result in the movement of soil particles, sand, and dust across the surface of the planet (Goudie and Middleton 2001; Muhs et al. 1990). Although atmospheric sources are generally considered to be minor contributors of environmental strontium in most places, their relative contribution to overall Sr budgets is sometimes substantial (accounting for over 50% of the total measured strontium in some cases) especially in regions where soil minerals are highly weathered and/or in which the concentration of Sr in bedrock and soils is relatively depleted (Borg and Banner 1996; Kennedy et al. 1998; Vitousek et al. 1999).

The combination of multiple hydro-atmospheric processes will tend to alter or obscure strontium isoscapes (or isotope landscapes) derived solely from terrestrial or geological sources alone [herein I use the definition of isoscapes as “maps of the spatial isotopic variation of the material(s) of interest” (West et al. 2010:vi)]. This is particularly true in coastal and insular settings where the atmospheric and marine sources of Sr may contribute substantially to overall ecological (or biosphere) Sr budgets. However, non-terrestrial sources can be significant even in continental or interior settings (Gallet et al.

1998; Graustein and Armstrong 1983; Graustein 1989) and thus researchers should be cognizant of the complexities of the many potential processes and variable sources of Sr when constructing isoscapes of biologically available Sr in all contexts (Bataille et al. in press; Evans et al. 2009).

3.2.5 Strontium Isotope Ecology

Strontium often enters the food chain through the transport of weathered soil minerals into plant tissues. It then moves through the food chain as herbivores and omnivores consume plants and as carnivores consume them, and so on. Although there is a known reduction of strontium concentrations (relative to calcium) at each successive step in the food chain (Burton et al. 1999; Burton and Price 2000), this reduction does not alter the measured strontium isotope ratio. Therefore, it is possible to use simple mixing models to constrain the limits of Sr isotope ranges for given islands or regions of the Caribbean using geological Sr isotope data and that of the sea as the two main sources of Sr into local terrestrial ecosystems (Bentley 2006; Montgomery et al. 2007). In fact, any organism which subsists solely or primarily on local food resources (terrestrial and/or marine) is expected to have a Sr isotope value that falls somewhere between the marine and terrestrial values for that locale.

One of the main problems with associating a specific organism to a specific place using isotopes involves the complexities of catchments areas, places from where an organism derives most of its subsistence. The size of a catchment varies between and within species based on a wide range of factors such as: the mobility of the organism, the distribution of its primary food sources, seasonal changes in resource availability, and so on. Catchment sizes and food sources can also be highly variable between individuals of the same species, based on factors such as sex and age. These complications can be further exacerbated amongst humans where the consumption of food is conditioned by a wide variety of social and cultural processes, in particular the use and manipulation of food as a tool for social differentiation.

A few points about ecological considerations of dietary Sr require further elaboration. Life cycle changes in food sources influence Sr isotope compositions of diets if food sources change throughout an individual's life, a process which occurs amongst a wide array of species. Most mammals primarily consume their mother's milk for a certain period of time after birth. The Sr isotope composition of breast milk should be expected to be similar to the average Sr isotope composition of the mother's diet over the period of time that the milk was produced. Under certain circumstances the mother's body may draw on stores of minerals from her own tissues during the period of lactation in which case some of the Sr in her breast milk may be derived from Sr which was consumed much earlier than the period of lactation (perhaps by several years or even decades). During the process of weaning, young mammals begin to consume food sources other than mother's milk. However, the dietary breadth is not always equivalent between the juvenile and adult members of the same species, especially amongst humans. In other words, in some species, juveniles do not necessarily consume diets that are identical to their parents even after weaning has occurred (Katzenberg et al. 1996; Wright and Schwarcz 1999).

Variations in Sr isotope compositions occur between species residing in the same isotopic environment for a variety of reasons. If the entire catchment area for any two or more species falls within a single Sr isotope domain, or isotopically homogenous area, then all individuals within that area may be expected to have similar Sr signatures. However, in areas of greater isotopic (Sr) diversity, the catchment ranges of various species, or even individuals of the same species may overlap one or more distinct isotope domains. This obviously complicates the relationship between the isotopic compositions of bodily tissues and the isotopic composition of the Sr sources. In isotopically diverse landscapes, Sr concentrations and compositions may vary widely, not only between different food types but also between similar food types with distinct (biogeochemical) origins. It should also be kept in mind that distinct isotopic origins does not necessarily imply large spatial distances, as one food source may have a quite different Sr isotope composition from another nearby identical food source if there are distinct differences in the background Sr isotope budgets between them. These complications can be become further exacerbated by temporal or seasonal changes which may alter food resource acquisition patterns within and between species.

3.2.6 Strontium Isotopes- Biology

Bioapatite, commonly expressed as $Ca_{10}(PO_4)_6(OH)_2$, is the primary mineral component of human bone and dental enamel (Burton 2008). Both the hydroxyl and phosphate molecules are often be replaced by other molecules or ions such as fluorine (F) or carbonate (CO_3) (Hillson 1996). Fully formed human dental enamel is approximately 97-99% calcium hydroxylapatite (White and Folkens 2005). Because strontium and calcium are somewhat chemically similar, strontium can and does replace calcium within the crystalline structure of bioapatite (Bentley 2006). Unlike bone, enamel does not undergo substantial remodeling after maturation and mineralization ceases and is highly resistant to diagenic alteration or contamination, and thus it preserves the biogenic signal of its formation and development (Budd et al. 2000; Koch et al. 1997; Montgomery et al. 2000; Nielsen-Marsh and Hedges 2000; Price et al. 2002; Trickett et al. 2003). As the time of formation and development of human teeth are fairly well-known (Hillson 1996), the analysis of Sr isotope ratios of dental enamel will provide information concerning the geochemical environment of an individual's youth, from several months *in utero* until approximately 10-14 years of age depending on the tooth type (White and Folkens 2005).

One of the main functions of the skeletal system is its role as a sink or reservoir for minerals from which the body can draw upon when needed (White and Folkens 1999). As such, the concentrations of many minerals and their constituent elements are generally elevated in skeletal tissues relative to other bodily tissues (Larsen 1997). This is fortunate for archaeological analysis in that these minerals and trace elements exist in sufficiently measurable quantities in the very types of biological tissues most resistant to decay (teeth and bones), and thus most likely to be preserved in the archaeological record.

There are distinct differences between the processes of growth, development, and maintenance in the structure and composition of bone and teeth that are relevant for biochemical analysis (Larsen 1997; Price 1989; White and Folkens 2005). Teeth consist of several main components: crown, root, and pulp chamber. The enamel crown is nearly completely mineralized after formation. Once growth and development cease, enamel is

considered an inert mineral tissue (Hillson 1996). Although some research has indicated that surficial enamel can undergo mineral exchange in both the oral and burial environment, internal core enamel appears to be highly resistant to these processes (Budd et al. 2000; Kohn et al. 1999; Lee-Thorp 2002).

Dentine is the portion of the tooth directly under the enamel crown and separated from it by the enamel-dentine junction, it is less mineralized than enamel but more so than bone (Hillson 1996). Because dentine interfaces with the pulp cavity of the root and its supply of nerves and blood vessels, it is somewhat more dynamic than enamel in that deposition and mineralization of dentine matrix may occur in response to stress to form secondary or tertiary dentine (Hillson 2005). However, it appears that this response is limited and that, unlike bone, primary dentine does not undergo substantial remodeling subsequent to initial formation and mineralization (Brudevold and Söremark 1967; Molleson 1988).

Bone represents the other major component of the skeletal system and differs from teeth in several key aspects. In general, bone is much more dynamic than dental enamel and unlike enamel it is considered a living tissue (White and Folkens 2005). Bone consists of two main phases; an inorganic mineral phase composed mostly of calcium hydroxylapatite, and an organic protein phase consisting mostly of collagen. This matrix contributes to bone's dynamic nature, allowing it the rigidity necessary for its structural and weight-bearing functions but also the elasticity to respond to stress and activity, as well as fulfilling its role as storage reservoir for various essential minerals. There are two main types of bone, cortical (dense) bone and cancellous or trabecular (spongy) bone. In general, the outer portions of most bones are comprised of a layer of dense cortical bone and the inner portions are comprised of a layer of spongy bone (Price 1989; White and Folkens 2005).

Cancellous bone is less dense and possesses greater surface area for interfacing with the circulatory system, for example within the distal ends of long bones. Because of this interface, the rates of bone accretion and absorption (remodeling) are generally much higher in spongy bone than in dense bone. Thus, bone remodeling is generally more active and rapid in spongy bone with turnover or replacement of the mineral structure occurring at various rates (Schweissing and Grupe 2003b). Owing in part to its greater

density, reduced access to the bloodstream, and somewhat different function, cortical bone turnover is much slower and can take as much as several decades for most mineral replacement to occur (Larsen 1997; Price 1989; White and Folkens 2005).

In summary, enamel is essentially an inert and completely mineralized tissue once it is fully formed, while bone, because it continues to interact with the body's metabolic and regulatory systems (although at reduced rates in older years) is dynamic and its mineral phase is continually replaced throughout an individual's life. The implications of these distinctions are that teeth preserve a record of an individual's biogeochemical environment during infancy and childhood, while bone contains the same information for an individual's last few years or decades of life, depending on the type of bone (Hoppe et al. 2003).

Because the timing of formation and development of different human teeth are relatively consistent and well known it is possible to analyze the isotopic signatures from various teeth from a given individual to roughly estimate his/her age at the time of migration, if this occurred during childhood or adolescence. Also, because the replacement of bone mineral may take several years it is possible that a recent migrant may not reflect a local signal in bone tissue. The existence of an individual with a nonlocal dental isotopic signal and a bone isotopic that is either non-local or intermediate between that of his/her dental enamel and the local signal may be suggestive of recent migration prior to death.

In fact, because different tissues form at different time periods and remodel at varying rates they reflect different time aspects of an individual's life history. Serial sampling refers to the collection and analysis of different types of tissues from the same individual, for example: different dental elements (deciduous versus permanent teeth, molars versus premolars), different portions of dental elements (enamel versus dentine, or even dental calculus), and different types of skeletal elements (cancellous versus cortical bone), or even non-skeletal tissues if present (hair, skin, soft tissue). By serial sampling and analysis of different tissues from the same individual, it is possible to address questions concerning the timing or age of various life history events or processes, such as the age at which an individual moved to a distinct isotopic setting, the age at which weaning occurred, and possibly combinations of processes such as changes in dietary

practices associated with migration/mobility (Grupe et al. 1999; Muller et al. 2003; Schroeder et al. 2009; Schweissing and Grupe 2003b).

3.3 Strontium Isotopes- Archaeology

It is the aforementioned specific characteristics of strontium isotope systems and their interactions within biogeochemical systems that explain their archaeological utility. This approach was originally proposed by Ericson (1985). The methodology, techniques, theory, and application of strontium isotope analyses to human provenance studies have been developed and refined over the last two decades as discussed below.

3.3.1 History of Sr Isotope Research in Archaeology

As with many advances in archaeological sciences, the basic methodological and theoretical foundations of isotopic studies were previously developed by several other related fields prior to their application to archaeology. Isotope archaeology probably owes its greatest debt to isotope geochemistry, a field that developed immensely throughout the twentieth century to address a wide array of research questions concerning the evolution and dating of geological structures within Earth and planetary sciences. Although the scale of this research is far beyond the scope of this chapter [see (Faure and Mensing 2005; Fry 2006) for extensive overviews of these topics]; it is important to note that enormous advancements in our ability to accurately and precisely measure a wide variety of isotopes within different types of sample materials had already been developed prior to the first application of isotopic analyses within archaeology. However, it is also necessary to credit applications of isotopic studies within the ecological sciences as the direct ancestor of isotopic archaeology. Successful applications of isotopic research into animal and plant biochemistry and wildlife ecology were borrowed upon heavily by archaeological scientists in the first applications of these methodological approaches to human remains from the archaeological record.

As previously stated, the first archaeological application of Sr isotope analysis to provenance studies appeared the mid-1980's. In the next few years, very little research in this vein was published until the early to mid-1990's when these methods began to be systematically applied to archaeological skeletal materials from the Americas and Europe. In the last ten years or so, many more researchers have analyzed and published Sr isotope research on archeological materials from various regions of the world. The last two decades have also witnessed a broad expansion of the types of research questions addressed by, and the theoretical aspects of, Sr isotope archaeology. Some of the main developments within this burgeoning field revolve around the way in which locals and nonlocals are identified, how local Sr isotope ranges are defined, and the extent and ways in which Sr isotope data are interpreted vis-à-vis other archaeological and isotopic data as discussed in further detail in the following sections [see reviews in (Bentley 2006; Montgomery 2010; Pollard 2012; Price et al. 2002)].

3.3.2 Determining the Local Sr Isotope Signature

Most archaeological applications of strontium isotope provenance studies rely on determining the isotopic range of a given site or region from which the sampled skeletal material is derived [see reviews in (Beard and Johnson 2000; Bentley et al. 2004; Price et al. 2002). One advantage of biogenesis is its averaging effect, which tends to homogenize local or intra-regional variation causing individuals and organisms to possess broadly similar ratios despite micro-level Sr isotope heterogeneity such as in rock and soil minerals and/or varying dietary contributions (Price et al. 2002; Sealy et al. 1991; Sillen et al. 1995; Sillen et al. 1998). This allows for a local range of Sr isotope variation to be defined for a given locality and any isotope ratios observed within a skeletal assemblage that do not fall into this range are then identified as ‘nonlocal’. In this sense, nonlocal simply refers to an individual sample possessing an $^{87}Sr/^{86}Sr$ ratio that is outside of the range of local Sr isotope variation. Various methods and approaches for defining and estimating the local range are utilized, each with its own advantages and disadvantages

(Bentley et al. 2004; Price et al. 2002). Since these are an essential component of the overall methodological approach used in this project, they merit further discussion.

One method for determining if an individual is local or not, is to directly compare the strontium isotope ratios from enamel with that of bone for each individual (Price et al. 1994a). Isotope ratios of bone samples are considered to be directly reflective of the local range, under the assumption that people often die and are interred in the general vicinity of where they lived at the end of their lives. Therefore, 'locals' should have quite similar enamel and bone signals owing to lifelong residence at or near the place of birth, while nonlocals can possess enamel signatures that vary significantly from their own bone signatures. There are several potential problems with the application of this approach. For example, recent migrants whose bones have not had time to normalize or equilibrate to the local signature will complicate assessments of the local range. Although probably uncommon, certain mortuary customs involving long-distance, post-mortem transport of remains will result in similar difficulties (Keegan 2009). Lastly, and perhaps more importantly, bone is much more susceptible to diagenesis or post-mortem contamination than dental enamel and thus ratios obtained from bone may not only reflect biogenic Sr but also contributions of Sr from the burial environment (Budd et al. 2000; Chiaradia et al. 2003; Hoppe et al. 2003; Lee-Thorp 2002; Sillen and Sealy 1995).

Owing in part to these concerns, some researchers have attempted to develop statistical approaches for distinguishing between locals and nonlocals using human enamel Sr isotope data alone (Bentley et al. 2002; Ezzo et al. 1997; Grupe et al. 1997; Price et al. 1994b; Price et al. 2001; Wright 2005). These approaches rely on several assumptions; 1) that most individuals within a burial population are probably locals, in that they have spent their entire lives in and around the site or at least within the region in question, and 2) that if migrants or nonlocals are present they will comprise a minority of the individuals in any given burial population. If these assumptions can be upheld then the local range can be estimated through statistical assessment of the human enamel Sr isotope data. For example, the local range can be defined as the mean of all isotopic ratios of analyzed samples plus or minus two times the standard deviation (mean ± 2 s.d.). Any individual whose enamel ratio falls outside of this range is then considered to be nonlocal. Such a local range estimate is more likely to underestimate rather than overestimate the

number of non-locals within a burial population (Bentley et al. 2004). One benefit of this approach is that it requires only one tooth per individual and does not necessitate the collection and analysis of bone samples, which may be absent, unavailable, or contaminated by diagenetic processes in many cases.

Determinations of local isotopic ranges can be derived from non-skeletal isotopic ratios as well. This is accomplished by analyzing samples of bedrock, soil, or water which are collected in and around a site and are thus presumably broadly similar to the ratios obtained from the skeletal elements of local individuals (Evans et al. 2009; Evans et al. 2010; Evans et al. 2012; Frei and Frei 2011; Hodell et al. 2004; Voerkelius et al. 2010; Wright 2005). Problems with this method include the tendency for some of these types of samples, especially whole rocks and bulk soil samples, to reflect much greater isotopic variation or heterogeneity than signatures from bones and teeth (Bentley et al. 2004; Price et al. 2002). This is probably a result of the aforementioned process of 'bio-averaging', whereby skeletal elements tend to possess signals with reduced variance compared to the possible variation of consumed resources. In addition, dietary contributions of Sr from outside the geological region will not be represented in these samples. Lastly, because of inherent differences in the weathering and solubility of various types of rocks and minerals, biological signatures may vary from those taken directly from underlying bedrock, especially if the local geology is complex and heterogeneous. In fact, this lack of correlation between geological and biological samples from the same region has been demonstrated in multiple settings (Bentley et al. 2004; Kennedy and Derry 1995; Kennedy et al. 1998; Laffoon and Hoogland 2009; Price et al. 2002; Price and Gestsdóttir 2006; Vitousek et al. 1999).

To overcome some of the limitations of the previous approaches, Price and colleagues have recommended using local faunal samples to determine local ranges of 'biologically available' strontium (Price et al. 2002). This is based on the premise that although the lithosphere is the ultimate source of strontium, it is the Sr composition of the biosphere that is most relevant for archaeologists. The choice of type of fauna varies depending on the geographical, temporal, and cultural setting (Bentley 2006; Evans et al. 2009; Price et al. 2002) but species with relatively restricted catchment ranges are required in order to be reflective of the locally available Sr signature. Migratory species

are obviously excluded because their tissues are potentially representative of a much wider range of Sr sources. Suitable species include those that are relatively sedentary, those that obtain most if not all of their dietary requirements locally, those with suspected catchment ranges approximating those of humans, or species with similar dietary behavior as humans. Small terrestrial mammals have been suggested as possibly ideal in this regard (Price et al. 2002). Cattle, pigs, dogs, rodents, land snails, and others have been used or suggested in various contexts because they meet some or all of these criteria (Ericson, 1989; Knudson et al. 2004; Schweissing and Grupe 2003b; Sealy et al. 1991; Sillen et al. 1998). In addition, archaeological materials are preferable to modern ones, owing to the possibility that the latter may be contaminated by modern pollutants or fertilizers (Borg and Banner 1996; Price et al. 2002).

The bioavailable approach has several benefits including; 1) the utilization of the averaging tendencies of biological organisms, which reduces potential isotopic heterogeneity inherent in non-biological materials; 2) it does not require the use of human bone tissue; 3) local faunal materials are generally more abundant, accessible, and available than human bone; 4) assuming that fauna are local, this method reduces questions of locality and context (circularity) which plague approaches that are based on human Sr isotope data alone. Nonetheless, there are some limitations to the bioavailable approach including: the availability of appropriate samples; unknown Sr catchment represented by some proxy samples; the possibility of movement of animals or their remains, and large investments of time and resources required for extensive coverage (Bataille et al. in press; Evans and Tatham 2004; Evans et al. 2009).

3.3.3 Complications and Limitations of Strontium Isotope Analysis

Having summarized the basic methods and premises of strontium isotope analysis it is necessary to review some of the complications and limitations of this analytical tool. First, as is the case with most types of biochemical analysis of human remains, this method is destructive. This fact alone limits the application of this technique for certain archaeological materials. Most techniques for isotope analysis rely on sample processing

protocols involving the complete or partial destruction of teeth and bone elements. Other techniques, such as the direct laser ablation of solid materials, as opposed to dissolution and chemical separation have been applied to both trace element (Cucina et al. 2005; Speakman et al. 2002) and isotope analysis (Balter et al. 2008; Copeland et al. 2011; Montgomery et al. 1999; Prohaska et al. 2002; Richards et al. 2008). These methods may permit analyses that are minimally destructive but as yet these have not been fully or adequately developed to obtain measurements that are as reliable as traditional wet chemistry procedures [see discussions in (Copeland et al. 2008; Copeland et al. 2010; Horstwood et al. 2008; Nowell and Horstwood 2009; Simonetti et al. 2008)]. Moreover, recent technological developments suggest that Sr isotopes can be determined on 0.1 ng of Sr equivalent to ~ 0.1 mg of enamel making the conventional approach essentially non-destructive (Koornheef et al. 2012).

Another widespread problem in most types of biochemical analyses of human remains is the potential for contamination. Ultimately, isotopic signals that are the result of biogenic and not diagenic processes are those that are of interest for most provenance studies (Budd et al. 2000; Chiaradia et al. 2003; Hoppe et al. 2003; Lee-Thorp 2002; Sillen and Sealy 1995). As previously mentioned, enamel is considered highly resistant to diagenesis and there is a general consensus that existing protocols to remove potential contaminants from enamel are sufficient in most cases (Budd et al. 2000; Hedges 2002; Hoppe et al. 2003; Lee-Thorp and Sponheimer 2003; Trickett et al. 2003). On the other hand, bone's relative resistance or susceptibility to diagenic processes varies widely depending on several factors, including; the type of bone (cortical or cancellous); preservation conditions; soil pH and moisture levels; and the protocols used to mitigate these concerns (Hoppe et al. 2003). Lastly, although biogenic signals can be preserved for considerable time in both bone and teeth, even enamel is susceptible to diagenesis in very ancient (paleontological) samples or under extremely poor conditions for preservation (Hoppe et al. 2003; Sealy et al. 1995). Fortunately, even if bone (or tooth) contamination were to take place, the effects of the diagenesis would generally be that nonlocals would be misidentified as locals (and not vice versa) causing an underestimation of migrants/migration (Price et al. 2002; Price et al. 2006).

Another potentially substantial limitation of isotope studies of migration is the potential lack of geological and isotopic variability. In the absence of spatial variability, no amount of movement or migration will be discernible with these methods. In large regions with similar geology or widespread geological homogeneity, there may be insignificant isotopic variation to identify nonlocals relative to locals. Since there is an essential spatial parameter to both geological/isotopic variation and to migration, areas of broader or more widespread homogeneity will have fewer identifiable nonlocals and potentially greater numbers of isotopically local, 'internal' migrants. Nevertheless, long-distance migrants would still be identifiable as long as they originated from a region that was isotopically distinct. Extreme heterogeneity of background isotopic variation may be similarly problematic, particularly when occurring on scales that are discordant with the distances that people usually migrate (Hodell et al. 2004). However, owing to the previously mentioned effects of bio-averaging, excessive heterogeneity of isotopic variation seems less problematic under most circumstances.

The lack of ability to identify, or more accurately the tendency to misidentify a nonlocal as such, represents a common limitation of isotope studies. As previously mentioned, the underestimation of nonlocals or migrants can also be caused by diagenesis; the lack of isotopic variation; the inability to identify local, internal, or short-distance migrants; statistical overestimation of local ranges; and the isotopic heterogeneity of baseline samples. However, the underestimation of nonlocals would seem to have fewer severe interpretive consequences than, and would generally be considered preferable to, the overestimation of nonlocals. The possibility of overestimation of the number of nonlocals is also intriguing considering the large number of nonlocals identified in some archaeological populations (Bentley et al. 2002; Bentley et al. 2003; Bentley et al. 2004).

Marine-derived strontium can influence the strontium isotope ratios in human teeth and bones in direct and indirect ways. In coastal and island settings, marine Sr can be deposited directly into terrestrial ecosystems via sea spray, rainwater, or even the use of kelp as fertilizer (Capo et al. 1998; Montgomery et al. 2007; Sealy et al. 1991; Whipkey et al. 2000). From there the marine Sr can enter the local groundwater and soils and can be incorporated into plant tissues and from there passed on to the local

(terrestrial) food chain. Marine-derived strontium can also enter terrestrial food webs through the direct consumption of marine resources (e.g., seafood and sea salt) by consumers (Burton and Price 1999; Ericson 1985; Price and Gestsdóttir 2006; Sealy et al. 1991; Wright et al. 2010). However, the degree to which the consumption of marine resources can influence the $^{87}Sr/^{86}Sr$ ratios of human skeletal materials is not entirely clear. On the one hand, Sr concentrations in the sea are quite high and some types of seafood (such as seaweed, shellfish, and sea salt) also can possess high Sr concentrations (Burton and Price 1999; Montgomery et al. 2007; Wright 2005). On the other hand, although the concentration of strontium in bones and teeth is to some extent dose dependent, it is also influenced by the concentration of calcium in the diet and by trophic level effects (Burton and Wright 1995; Burton et al. 1999; Burton et al. 2003). As Burton and Price have noted (1999:235) "While seawater is high in strontium, marine diets are not". In general, amongst omnivores, plants are expected to be the largest contributors to dietary Sr, owing to the much higher levels of Sr in plant tissues relative to meat (Burton and Price 2000; Elias 1980).

Thus, substantial consumption of marine-derived Sr, both indirectly and directly, can produce a 'marine effect' where biogenic $^{87}Sr/^{86}Sr$ ratios in the tissues of terrestrial consumers are influenced by the $^{87}Sr/^{86}Sr$ ratio of seawater. The potential of this so-called marine effect to complicate the interpretation of Sr isotope results has been noted for several regions of the world (Burton and Price 1999; Ericson 1985; Evans et al. 2010; Montgomery et al. 2007; Price and Gestsdóttir 2006; Shaw et al. 2010; Slovak et al. 2009; Wright et al. 2010). Luckily, because the Sr isotope ratio of modern seawater is well documented (McArthur et al. 2001) and thus it is possible to estimate and assess the so-called 'marine effect' and its influence. In fact, some researchers have found that the $^{87}Sr/^{86}Sr$ of human skeletal material from island and coastal populations, reflect a mixture of terrestrial and marine-derived strontium, with $^{87}Sr/^{86}Sr$ values falling somewhere between these two end members (Montgomery et al. 2007; Montgomery et al. 2005; Price and Gestsdottir 2006; Wright, 2005). Furthermore, several independent methods are available to monitor and quantify possible marine influences on the $^{87}Sr/^{86}Sr$ ratios of terrestrial organisms. For example, $^{87}Sr/^{86}Sr$ ratios in terrestrial plant tissues can provide a means for assessing the relative contributions of marine Sr (via sea spray or rainfall) and

bedrock weathering to the bioavailable $^{87}Sr/^{86}Sr$ of local ecosystems. Additionally, both stable isotope analysis (carbon and nitrogen) and the analysis of trace elements (such as barium concentrations) can contribute to assessments of direct marine resource consumption.

Another potential limitation of isotope analyses is that if nonlocals originate from regions (regardless of how distant) with similar isotopic signals as the local population or area, they will be misidentified as locals (false negatives). In addition, and perhaps more significantly, the isotope data itself usually cannot be used to identify a specific origin in the absence of other lines of evidence, in part because so many regions throughout the world share similar isotope ranges. Isotope results can be used to eliminate proposed or hypothesized places of origin derived from other lines of evidence and to generate hypotheses, which is how these results are most often applied (Knudson et al. 2005; Price et al. 2000; White et al. 1998; White et al. 2002). Additionally, isotope approaches are also limited to the recognition of first-generation migrants only and not their descendants (e.g., second, third, or later generation migrants) (Price et al. 2006).

In summary, acknowledgement of the limitations of traditional archaeological approaches to the study of human migrations is one of the primary reasons for the development and gaining popularity of biochemical and isotope methods. The results of several of these studies have revealed that a substantial proportion of some populations are migrants and that their associated materials (grave goods) and mortuary contexts were not necessarily good indicators of migratory behavior or differential origins. Since the analysis of material culture may be a relatively poor predictor of migration in certain cases, it would seem logical to attempt to assess and explore the possibility of migration even in the absence of material culture indicators. In other words, it is possible that migration may be evident, important, identifiable, analyzable and relevant to the general aims of archaeological research in many contexts in which there is no material culture evidence for its existence.

Nonetheless, isotope approaches alone can only identify first generation migrants and in certain cases contribute to investigations of geographic origins and cultural affinities. Therefore, such approaches are most effectively employed in conjunction with other analytical approaches such as those focusing on for example: material culture,

settlement data, bioarchaeology, paleodemography, and mortuary evidence. Often, it is only from a broad-based, multi-disciplinary approach that meaningful patterns can be elucidated.

3.3.4 Archaeological Applications of Sr Isotope Analysis to Human Migrations

Analysis of strontium isotope ratios from human skeletal remains is the most commonly employed biochemical method for assessing patterns of human mobility and migrations and for exploring the geographic origins of individuals and groups. This method has been applied to address a wide array of research questions in an enormous range of archaeological contexts.

1) In Europe: to assess patterns of mobility for Linearbandkeramik (Bentley et al. 2012; Bentley et al. 2003; Bentley et al. 2002; Bentley et al. 2004; Price 2001; Price et al. 2006b; Richards et al. 2008b), Bell Beaker (Grupe et al. 1997; Price et al. 1994a, 1998), and other Neolithic (Chiaradia et al. 2003; Giblin 2009; Haak et al. 2008; Latkoczy et al. 1998; Nehlich et al. 2009; Oelze et al. 2012a; Oelze et al. 2012b; Sjögren et al. 2009; Smits et al. 2010), and Roman-period (Schweissing and Grupe 2003a; Schweissing and Grupe 2003b), and medieval (Aberg et al. 1998) populations of central, northern and western Europe; to identify the origins of "Otzi" the iceman (Hoogewerff et al. 2001; Müller et al. 2003); to investigate migrations of Neolithic and Roman peoples in England (Budd et al. 2004; Chenery et al. 2010; Eckardt et al. 2009; Evans et al. 2006a; Evans and Tatham 2004; Evans et al. 2006b; Evans et al. 2012; Leach et al. 2010; Montgomery et al. 2000; Montgomery et al. 2005; Montgomery et al. 2007; Müldner et al. 2011); to explore Norse (Viking-age) migrations into Iceland (Price and Gestsdóttir 2006), Ireland (Knudson et al. 2012), and Scotland (Montgomery et al. 2003; Montgomery and Evans 2006); to test proposed population movements to Crete (Nafplioti 2008); and to examine Neanderthal mobility in Greece (Richards et al. 2008)

2) In North America: to track migration and mobility amongst prehistoric populations from the Western (Ericson 1985), Southwestern (Ezzo et al. 1997; Ezzo and Price 2002; Price et al. 1994b), Southern (Quinn et al. 2008), and Midwestern (Beehr

2011; Price et al. 2007a) United States; to identify migrants at the ancient sites of Teotihuacán, Mexico (Price et al. 2000) and Xuenkal (Tiesler et al. 2010) in Mexico and Tikal (Wright 2005) and Kaminaljuyú (Wright et al. 2010) in Guatemala; to test hypotheses concerning the origins of individuals, elites and the supposed 'founder' of the Maya city of Copan, Honduras (Buikstra et al. 2004; Price et al. 2010); to examine the origins of sacrificial victims at various sites across Mesoamerica (Price et al. 2007b; White et al. 2007) [see also (Price et al. 2008)]; and to test the provenance and natal origins of possible African migrants from colonial cemeteries in Campeche, Mexico (Cucina et al. 2005; Price et al. 2006), the New York African burial ground (Goodman et al. 2004; Jones et al. 2003) and from slave burials in Barbados (Schroeder et al. 2009) and St. Martin (Schroeder et al. 2012); and in the Caribbean to examine prehistoric residential mobility on the island of Guadeloupe (Booden et al. 2008).

3) In South America: to trace migration and mobility patterns from various Wari, Tiwanaku, and Chirabaya affiliated sites in Peru and Bolivia (Conlee et al. 2009; Knudson et al. 2004; Knudson and Buikstra 2007; Knudson and Price 2007; Knudson 2008; Knudson and Blom 2009; Slovak et al. 2009); to determine the origins of the Juch'uypampa cave mummies from southern Bolivia (Knudson et al. 2005); to investigate the origins of Nazca (Knudson et al. 2009) and Wari (Tung and Knudson 2008, 2010) trophy heads; to examine population heterogeneity and the life histories of individuals in the Atacama desert of northern Chile (Knudson et al. 2010a; Knudson and Torres-Rouff 2009; Torres-Rouff 2003); to explore the origins of Inka groups at Machu Picchu (Turner et al. 2009) and other sites in the Cuzco Valley (Andrushko et al. 2009; Andrushko et al. 2011); and patterns of residential mobility amongst a prehistoric coastal population in Brazil (Bastos et al. 2011).

4) In Africa: to examine mobility and dietary patterns amongst Australopithecines and other early hominins in South Africa (Copeland et al. 2011; Sillen et al. 1995; Sillen et al. 1998); to study patterns of mobility and kinship amongst Holocene skeletons from southwestern Libya (Tafuri et al. 2006); to test migration models for Nubian populations in the Nile Valley (Buzon et al. 2007) and to investigate identity and life histories of Khoisan and historical-era groups (Sealy et al. 1995) and shipwrecked slaves from coastal cemeteries in South Africa (Cox and Sealy 1997).

5) In Asia: to test hypotheses concerning relationships between cultural practices and population mobility amongst the Jomon in Japan (Kusaka et al. 2009; Kusaka et al. 2011); and to examine spatial and temporal variation in patterns of residential mobility amongst hunter-gatherer, Neolithic, Bronze Age, and Roman populations from Siberia (Haverkort et al. 2008), Arabia (Gregoricka 2011), Jordan (Perry et al. 2008), Turkey (Welton 2010), Malaysia (Valentine et al. 2008), and Thailand (Bentley et al. 2005; Bentley et al. 2007b; Bentley et al. 2009).

6) In Oceania: to study Lapita migrants and migrations on Vanuatu (Bentley et al. 2007a) and the Bismarck Archipelago, Papua New Guinea (Shaw et al. 2010; Shaw et al. 2011; Shaw et al. 2009).

3.3.5 Other Applications of Sr Isotope Analyses

In addition to the aforementioned applications of Sr isotope analyses, this methodological approach is widely utilized in various other fields such as forensics, animal ecology, and (within archaeology) artifact provenance studies. Forensic applications of isotopic analyses (Ehleringer et al. 2010; Pye 2004) rely on the same basic parameters and principles as archaeological applications, and vary mainly in that they can be applied to more recent human remains (Beard and Johnson 2000) and thus benefit from the higher likelihood of preserved soft tissues, or even to living subjects (Font et al. 2012). Forensic Sr isotope applications are fairly recent and are primarily limited by problems associated with the global movement of agricultural commodities and food products in modern times. The consequences of these tendencies are that many individuals consume an enormous array of foods of highly variable and unknown origins and that both the types of foods and their origins may change significantly throughout a person's life. All of which tends to obscure and complicate one of the basic underlying premises of the Sr isotope approach, i.e., the assumed relationship between an individual's biogenic Sr isotope signal and the Sr isotope signal of their geographic origin(s).

The application of isotopic analyses, including Sr, to determine the origins of modern food and water has become quite widespread in recent years (Voerkelius et al.

2010). This is particularly true in Europe where issues of food origins and the ability to independently assess authenticity claims have led to attempts to source a wide variety of consumer products, from wine and cheese, to meat and honey (Camin et al. 2007; Schellenberg et al. 2010). In addition, the possibilities of isotopic analyses have been explored recently from a law enforcement perspective. This is partly due to the enormous potential of provenience studies (including isotope approaches) to independently test the suspected origins of illicit drugs and even illegally harvested timber (Galimov et al. 2005; Kagawa et al. 2008; West et al. 2009). Other forensic applications of isotope analyses for the purposes of law enforcement, include the sourcing of confiscated raw ivory and goods manufactured from ivory (van der Merwe et al. 1990; Vogel et al. 1990).

Applications of isotopic analyses within the field of animal ecology, and specifically in reference to animal migrations and mobility, have also greatly expanded in recent years [e.g., (Hobson and Wassenaar 2008; Hobson et al. 2010) and references therein]. The underlying principles are generally the same as archaeological applications although most ecological research in this vein is focused on examining the migration patterns of living organisms, often based on the analysis of tissues samples that can be acquired without killing the organism, e.g., feathers, fur, claws, horns, antlers, and eggshell. It should be noted however, that some applications of isotopic provenance studies have been directed towards paleontological contexts, including analyses of samples from Australopithecines (Copeland et al. 2011; Sillen et al. 1995) and extinct species of mammoth and mastodon (Hoppe et al. 1999; Hoppe 2004).

To date most archaeological applications of Sr isotope analyses have been applied to human remains to study various aspects of human movement. The same method can also be applied to any material or artifact that has sufficiently preserved quantities of biogenic strontium. For archaeological samples, the main restriction derives from our confidence (or lack thereof) in the authenticity of the sampled Sr, in other words whether or not the Sr being measured is biogenic or diagenic. The point being that a wide variety of archaeological materials are potentially suitable for Sr isotope analysis, either alone or in conjunction with various other traditional methods for the sourcing of artifacts and materials. Although limited in scope to date several recent studies have shown that strontium isotope analyses can be applied to provenance studies of non-skeletal materials

such as wood, maize, carbonized grains, glass, shell, and wool (Benson et al. 2003; Degryse et al. 2010; English et al. 2001; Frei et al. 2009; Heier et al. 2009; Henderson et al. 2005; Shackleton and Elderfield 1990; Vanhaeren et al. 2004).

3.4 Light Stable Isotopes

Early research into the use of stable isotopes for dietary reconstruction dates to the late 1970's and early 1980's (DeNiro and Epstein 1978; DeNiro 1985; Schoeninger and DeNiro 1982, 1984; Schoeninger 1985; van der Merwe and Vogel 1978; Vogel and van de Merwe 1977). These studies clearly demonstrated a relationship between the isotopic composition of an animal's tissues and the isotopic composition of its diet. Archaeological applications of these methods soon followed and initial research provided promising results for the future of paleodietary studies. Subsequent field and laboratory studies however, highlighted the existence of a number of complex and complicating factors (Ambrose 1990, 1991; Ambrose and Norr 1993; Ambrose and Katzenberg 2000). The last thirty years have witnessed a tremendous amount of research involving archaeological applications of stable isotope analyses to paleodietary reconstruction, including advances and developments in methodological and theoretical aspects of these perspectives (Ambrose 1990; Ambrose et al. 1997; Ambrose et al. 2003; Balasse et al. 2002; Keegan and DeNiro 1988; Le Huray et al. 2006; Richards et al. 2002; Schoeninger 2009; Schroeder et al. 2009; Schwarcz et al. 2010; Thornton et al. 2011). Carbon and nitrogen isotope analyses are now routinely applied to archaeological human remains for the purpose of elucidating past dietary practices, albeit with a greater appreciation of the scope and limitations of these approaches. A brief summary of the principles of oxygen and carbon isotopes analyses are provided below and highlight issues that are relevant for migration research in the Caribbean.

3.4.1 Carbon Isotopes

There are three naturally occurring isotopes of carbon (^{12}C, ^{13}C, and ^{14}C) with relative proportions of 98.9%, 1.1%, and <0.0001%, respectively. ^{14}C is radiogenic being produced by the radioactive decay of ^{14}N when subjected to cosmic radiation in the atmosphere. ^{14}C isotope analysis has a long history within archaeology for the purpose of absolute dating of organic materials since the first development of the method by Libby and colleagues in 1949 (Arnold and Libby 1949). It is important to note that in order to properly interpret radiocarbon data arrays, the isotope ratios of ^{13}C /^{12}C are routinely analyzed when samples are submitted for ^{14}C dating. This means that it is possible to obtain relevant information on paleodietary practices in conjunction with radiocarbon data, often at little additional expense. Because of the principles of mass dependent fractionation, the ratio of ^{13}C /^{12}C is affected by a number of natural chemical and physical processes and thus varies between the environment and the body tissues of different organisms (Katzenberg 2008). ^{13}C /^{12}C ratios are often reported as delta (δ) values in parts per thousand (‰) relative to an international standard (Pee Dee Belemnite, a marine carbonate fossil).

Carbon isotope analyses of enamel and bone apatite and bone collagen have been successfully applied to paleodietary studies for several decades. Carbon isotope values from collagen ($\delta^{13}C_{co}$) and apatite ($\delta^{13}C_{ap}$) reflect different aspects of consumption, with $\delta^{13}C_{co}$ primarily reflecting sources of dietary protein and $\delta^{13}C_{ap}$ reflecting an average of whole diet (Ambrose and Norr 1993; Krueger and Sullivan 1984; Lee-Thorp et al. 1989). Carbon isotope analyses can also be conducted on either bone or teeth, with the former providing information on long term dietary patterns and the latter providing information concerning dietary consumption patterns during childhood. Carbon isotope values of human skeletal carbonate ($\delta^{13}C_{ca}$) reflect the isotope composition of dietary sources of carbon. Carbon is incorporated into plant tissues via three distinct photosynthetic pathways (C_3, C_4, and CAM) and plants utilizing C_3 versus C_4 pathways possess distinct $\delta^{13}C$ signals in their tissues (Bender 1971; Katzenberg 2008). In the Caribbean, C_3 plants have more negative $\delta^{13}C$ values, averaging approximately -25.5‰ and C_4 plants have less negative $\delta^{13}C$ values of around -9.5‰ (Pestle 2010). Owing to the effects of fractionation

there is an offset in $\delta^{13}C$ between an individual's diet (the consumed) and an individual's skeletal tissues (the consumer). The degree of fractionation between enamel or bone $\delta^{13}C_{ap}$ values and diet is somewhat variable ranging from ~9.6‰ in rats to as high as 14.6‰ for cattle (Ambrose and Norr 1993; Howland et al. 2003; Passey et al. 2005). As $\delta^{13}C_{ap}$ is generally reflective of whole diet, and strongly influenced by the consumption of fats and carbohydrates, it can be used to infer the relative contributions of different categories of plant resources (C_4 versus C_3) to an individual's diet.

For this study, carbon isotopes in dental carbonate were analyzed to assess intra- and inter-regional differences in sources of dietary carbon and to help constrain the possible origins of individual immigrants at multiple scales. A large body of carbon and nitrogen bone isotope data from indigenous populations of the Caribbean has been developed over the last few decades (Laffoon and de Vos, 2011; Norr, 2002; Pestle, 2010; Stokes, 1998). At a macro-regional level, $\delta^{13}C_{ap}$ values display somewhat limited variation and indicate relatively modest contributions of C_4 plants to prehistoric Antillean diets. At archipelagic or insular scales, spatial patterning of bone collagen $\delta^{13}C$ and $\delta^{15}N$ values indicates some regional variation in dietary practices within the Antilles possibly as a result of biogeographic principles (Stokes 1998, 2005). For example, there is limited overlap in $\delta^{13}C_{co}$ and $\delta^{15}N$ clusters between the Greater and Lesser Antilles. The higher $\delta^{13}C_{co}$ and $\delta^{15}N$ values amongst the populations of the Lesser Antilles reflect a greater reliance on marine protein resources (owing to the relative paucity of terrestrial fauna on these smaller islands) relative to the Greater Antilles. Despite some degree of regional clustering of bone apatite ($\delta^{13}C_{ap}$) in the Antilles, the spatial patterning is much less distinct than the general pattern for collagen ($\delta^{13}C_{co}$) with a higher degree of overlap in the ranges of values between most islands and regions (Pestle 2010; Stokes 1998).

3.4.2 Oxygen Isotopes

Oxygen isotopes of skeletal tissues are primarily influenced by the isotopic composition of consumed water but are also influenced by the water in consumed food, food itself, as well as thermophysiology (Daux et al. 2008; Longinelli 1984; Luz et al. 1984; Podlesak

et al. 2008). The primary source of consumed water for most ancient populations is expected to be locally obtained drinking water which should correlate with meteoric and groundwater sources in most Amerindian contexts in the Antilles. Globally, meteoric $\delta^{18}O$ varies spatially primarily according to geographical and climatic variables, with $\delta^{18}O$ generally decreasing with increasing altitude, latitude, and distance from the coast or source of atmospheric water vapor (Bowen and Wilkinson 2002; Gat 1996). At smaller spatial scales, in tropical regions, rainfall regimes are primary factors influencing the spatial variation of $\delta^{18}O$ in meteoric water (Dansgaard 1964). Although there are broad similarities in rainfall regimes throughout the tropics, there is also substantial variation in the geographic and climatic conditions that influence $\delta^{18}O$ variation and clear spatial patterning in $\delta^{18}O$ exists at regional and local scales.

Oxygen isotope analysis of human skeletal tissues has also shown potential for archaeological investigations of human migration and residential mobility in various regions of the world (Bentley and Knipper 2005; Budd et al. 2004; Knudson 2009; Montgomery et al. 2005; Prowse et al. 2007; Schwarcz et al. 1991; White et al. 1998; Wright et al. 2010). $\delta^{18}O$ measurements can be conducted on the carbonate (CO_3) and/or phosphate (PO_4) components of bioapatite, which have been shown to produce different but comparable results in unaltered samples (Chenery et al. 2012). The direct comparison of $\delta^{18}O_c$ and $\delta^{18}O_p$ values requires either the conversion of one type of oxygen isotope value to the other or a conversion to drinking water oxygen isotope values. Although several formulae for converting these values have been proposed recently, there is still some debate about their application, accuracy, and compounded errors (Bell et al. 2010; Chenery et al. 2012; Millard and Schroeder 2010; Pollard et al. 2011). Additionally, oxygen isotope analyses of human skeletal tissues can be conducted on either bone or enamel. Owing to the much higher susceptibility of bone to diagenesis, bone oxygen isotope results should be used with caution. Enamel is considered much more resistant to diagenetic contamination than bone and as such enamel isotope results are considered to be more reliable (Hedges 2002; Hoppe et al. 2003; Lee-Thorp and Sponheimer 2003; Trickett et al. 2003). Enamel and bone isotope data also provide different information concerning provenance. In humans, dental enamel of most permanent teeth mineralizes during childhood (Hillson 1996) and does not undergo substantial remodeling throughout

life and is therefore reflective of the biochemical environment of an individual's childhood origin. Bone continuously remodels so that bone isotope values reflect the last years or decades of life and are less informative of natal origins.

Oxygen isotope analysis of enamel carbonate has been successfully applied to human remains from Barbados to distinguish between first generation (African) and later generation (Barbadian) slaves at the colonial period cemetery of Newton Plantation (Schroeder et al. 2009). At this site, first generation (forced) migrants from Africa possessed clearly distinct $\delta^{18}O$, and in some cases distinct $\delta^{13}C$ and dental modification practices, from locally born slaves. Owing to broadly shared environmental and climatic parameters influencing the spatial patterning of oxygen isotopes within the Antilles, we expected relatively limited variation in $\delta^{18}O$ amongst the indigenous populations of the region. For example, all of the sites in this study are within a few kilometers of the coast, at or near sea level. These similarities in geographic setting essentially eliminate two of the primary sources of variation in $\delta^{18}O$ of rainwater, namely distance from coast and altitude. Another possible, but unlikely, source of variation between our sample populations is latitude, which ranges from approximately 10° North from the most southerly site in Trinidad to 21° North at the most northerly site in Cuba.

Temporal variation at multiple scales, from seasonal or annual fluctuations in the amount of rainfall, to long-term climate change, represents another source of $\delta^{18}O$ variation. Paleoclimate research has documented long-term changes in environmental and climatic conditions in the Caribbean over the last several thousand years (Beets et al. 2006; Curtis et al. 2001; Hodell et al. 1991; Peros et al. 2007) although the degree of variation in $\delta^{18}O$ represented by these proxy data over the time period represented in our study (~ AD 600-1600) appears minimal relative to other potential sources of variation. Differences in rainfall regimes probably represent one of the largest sources of variation in meteoric $\delta^{18}O$ within the Antilles. Prevailing winds are generally from the northeast, and on the islands with high topographic relief there is substantial orthographic precipitation producing generally wetter conditions on the northeast coasts (for example the El Yunque rainforest on Puerto Rico) relative to the southwest coasts of these islands.

3.5 Summary

Traditionally, isotope analyses of human remains have been divided into paleodietary applications utilizing carbon and nitrogen isotopes and paleomobility applications utilizing strontium and/or oxygen isotopes. Increasingly, integrated approaches based on multiple isotope analyses have been systematically applied to human skeletal populations from different regions of the world (Bentley et al. 2007a; Knudson and Price 2007; Montgomery et al. 2005; Price et al. 2010; Sealy et al. 1995; Turner et al. 2009; Wright et al. 2010) to assess various aspects of individual identity. These multi-isotope approaches are deemed highly complementary because 1) as dietary patterns often display spatial variation, they can also contribute to assessments of geographic origins (Dupras and Schwarcz 2001; Müldner et al. 2011); and 2) dietary practices can change as the result of migrations or movements between diverse cultural or ecological areas (Knudson et al. 2010a; Sealy et al. 1995). Although, most applications of carbon and nitrogen isotope analyses to human remains in the Caribbean have explicitly focused on paleodietary reconstructions, several recent studies have begun to utilize $\delta^{13}C$ and $\delta^{15}N$ data to investigate issues of identity and individual origins (Schroeder et al. 2009; Varney 2007). This study utilizes a multiple isotope approach that is primarily based on strontium isotope analysis of dental enamel from a large number of human (and biosphere) samples, supplemented by oxygen and carbon isotope analyses of dental enamel from a subset of the total human sample population. These isotope results are also interpreted in the context of published carbon and nitrogen isotope data generated from prehistoric human bone material in the Caribbean.