Journal of Archaeological Science 40 (2013) 2561–2569

Contents lists available at SciVerse ScienceDirect

Journal of Archaeological Science

journal homepage: http://www.elsevier.com/locate/jas

$^{87}Sr/^{86}Sr$ variability in Puerto Rico: geological complexity and the study of paleomobility

William J. Pestle [a,*], Antonio Simonetti [b], L. Antonio Curet [c]

[a] Department of Anthropology, University of Miami, Merrick Hall 102E, Coral Gables, FL 33124, United States
[b] Department of Civil & Environmental Engineering & Earth Sciences, University of Notre Dame, Notre Dame, IN 46556, United States
[c] Department of Anthropology, Field Museum of Natural History, 1400 S. Lake Shore Drive, Chicago, IL 60605, United States

ARTICLE INFO

Article history:
Received 26 October 2012
Received in revised form
20 January 2013
Accepted 24 January 2013

Keywords:
$^{87}Sr/^{86}Sr$
Caribbean
Puerto Rico
Paleomobility
Paleomigration

ABSTRACT

The temptation to use biogeochemical techniques to resolve issues of paleomigration is evident and well intentioned. Knowledge of radiogenic strontium isotope baselines in a region of interest is a *sine qua non* of such archaeological studies of paleomobility. Here, we present the first detailed study of baseline $^{87}Sr/^{86}Sr$ values for the island of Puerto Rico. The high degree of $^{87}Sr/^{86}Sr$ variability present in this corpus of modern Puerto Rican bedrock and terrestrial malacological samples (0.70406–0.70909) is a testament to the complex geology of that island. This diversity of $^{87}Sr/^{86}Sr$ values makes parsing issues of origin a difficult and highly contingent task. Given these complexities, regional studies seeking to assess paleomigration by such isotopic means should proceed with a great deal of caution.

1. Introduction

Over the past two decades, the analysis of radiogenic strontium isotope ratios ($^{87}Sr/^{86}Sr$) for the purpose of reconstructing ancient migration has become commonplace in archeology (Åberg et al., 1998; Bentley, 2006; Buzon et al., 2007; Ezzo et al., 1997; Frei and Price, 2012, Knudson et al., 2004; Price and Getsdóttir, 2006; Price et al., 2000; Thornton, 2011; Wright, 2005). The application of this technique is based on variation in strontium isotope ratios in the mineral fraction of human teeth and bones, which reflect the isotopic composition of the geological (and to lesser degree atmospheric) substrate from which an individual obtained food and water at the time of tissue mineralization. A difference between the $^{87}Sr/^{86}Sr$ signature of an individual's dental/skeletal tissues and that of the local geology is interpreted as being the result of movement/migration during that individual's lifetime. Knowledge of radiogenic strontium isotope baselines in a region of interest is thus a *sine qua non* of archaeological studies of paleomobility. $^{87}Sr/^{86}Sr$ values of bedrock vary according to geologic age and composition (Faure, 1986), and don't fractionate appreciably as the strontium moves through the trophic structure of an ecosystem, from rocks to plants to animals, including humans (Blum et al., 2000). With knowledge of the local/regional strontium isotope variation, it becomes possible, in instances of discrete geological variation, to make assessments of possible ancient human migration.

While in recent years there has been an increase in such studies relative to the insular Caribbean (Booden et al., 2008; Laffoon et al., 2012; Laffoon and de Vos, 2011; Valcárcel Rojas et al., 2011), the region's baseline $^{87}Sr/^{86}Sr$ values still require further review and refinement. Here, we present the first detailed study of baseline $^{87}Sr/^{86}Sr$ values for the island of Puerto Rico, with an eye toward building a database useful for future studies of prehistoric and historic movement and migration in this easternmost island of the Greater Antilles.

2. Strontium systematics

Strontium is an element that is typically found in rock, water, soil, plants, and animals (at the ppm level) and possesses four natural isotopes: ^{84}Sr, ^{86}Sr, ^{87}Sr, and ^{88}Sr, the relative abundances of which are ~0.56%, ~9.9%, ~7.0%, and ~82.6%, respectively. On the basis of its similar ionic radius and valence charge ($^{2+}$), Sr usually substitutes for Ca^{2+} in crystallographic lattice positions within Ca-bearing geological materials. ^{87}Sr is the sole 'radiogenic' isotope, produced by the slow radioactive decay of ^{87}Rb (Faure, 1986). The small relative mass difference between ^{86}Sr and ^{87}Sr (~4.5%)

---

* Corresponding author. Tel.: +1 312 446 6370.
E-mail address: w.pestle@miami.edu (W.J. Pestle).

0305-4403/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jas.2013.01.020

renders isotopic fractionation through biological processes insignificant (Faure and Powell, 1972). Strontium abundances and corresponding isotopic ratios of soils and groundwater are a function of the local ("background") geology. $^{87}Sr/^{86}Sr$ ratios reflect the average $^{87}Rb/^{86}Sr$ ratios of the rocks in a particular geographic area, which is mainly a function of the composition of the rocks (i.e., constituent minerals), and the absolute age of the rocks. As a result, older rocks characterized by elevated $^{87}Rb/^{86}Sr$ ratios have the highest present day $^{87}Sr/^{86}Sr$ ratios. For example, a geographic region consisting of old (>1 billion year old) granites, which are typically characterized by high Rb/Sr ratios will have higher $^{87}Sr/^{86}Sr$ ratios than areas with geologically younger rocks with low Rb/Sr, such as basalt or limestone of marine origin (Faure, 1986).

The bioavailable strontium present in soil and groundwater is incorporated into local plants and subsequently into animals foraging on that vegetation. Hence, the strontium isotopic composition of an individual's or animal's diet will be recorded in the corresponding hard tissues (Ericson, 1985). For example, it is well known that Sr commonly substitutes for Ca in the hydroxyapatite of teeth and bone (Nelson et al., 1986). Moreover, tooth enamel of permanent adult teeth forms during early childhood (generally the first 12 years of life, with the exception of the third molar) and is subsequently considered a metabolically inactive tissue because it does not undergo any further remodeling (Hillson, 2005). Thus, tooth enamel will reflect the $^{87}Sr/^{86}Sr$ composition of the bioavailable Sr present in the geographic area in which a person or animal lived while the tooth was forming. Minerals that may have formed during subsequent diagenetic alteration may be taken up by the surface of the tooth during life or after burial, though these materials seldom penetrate deep into the enamel (Budd et al., 2004; Price et al., 2004; Wright, 2005).

A number of other factors, including mineral variation within single rocks (Bentley, 2006:141), differential weathering (Borg and Banner, 1996; Chadwick et al., 1999), and the contribution of atmospheric strontium sources, in particular seaspray and oceanic-derived precipitation (Kennedy et al., 1998; Price and Getsdóttir, 2006; Vitousek et al., 1999; Whipkey et al., 2000), can influence bioavailable strontium isotope signatures. As a result, knowledge of both geological and bioavailable strontium signatures are necessary when establishing the baseline $^{87}Sr/^{86}Sr$ values for a region of interest. Novel geological and biological data are presented below, as are previously published data on potential atmospheric strontium sources.

3. Puerto Rico: geography, geology, and geochemistry

Puerto Rico is the smallest (ca. 9100 km^2) and easternmost island of the Greater Antilles. The island is roughly rectangular in outline, with a maximum east–west length of approximately 180 km, and a north–south extent of just under 65 km. To the west, across the Mona Passage, lies the much larger (nearly 76,500 km^2) island of Hispaniola, while to the east are the Virgin Islands and, beyond that, the Lesser Antilles which curve southwards in a long arc terminating near the mouth of the Orinoco River in northern Venezuela. The island's topography is dominated by several mountain ranges with peaks in excess of 1000 m: the Uroyán Mountains and Cerro del las Mesas near the west coast, the Cordillera Central, which runs east-west along the island's long axis, the Sierra de Cayey in the southeast, and the Sierra de Luquillo in the island's northeast (Picó, 1974:26). These tall mountain peaks, dramatically eroded through the millennia by forces of rain and wind, present a diverse dissected terrain with a great deal of biotic (particularly floral) diversity.

As with the larger islands of the Greater Antilles, Puerto Rico presents a more diverse geological history than do the smaller and more homogenous islands of the Lesser Antilles, with a palimpsest of igneous, metamorphic, and sedimentary rocks of radically differing ages. The geological history of Puerto Rico stretches back nearly 150 million years (Krushensky and Schellekens, 1998), and consists of roughly seven phases (Bawiec, 1998; Cox, 1985):

(1) Early (126 mya) sedimentary, volcanic, and tectonic events in southwestern Puerto Rico forming the Bermeja Complex (Mattson, 1960),
(2) Overlaying of the Bermeja Complex by volcanic and sedimentary rocks from the middle through the end of the Cretaceous (roughly 112–65 mya),
(3) Intrusion of plutons from 125 to 65 mya,
(4) Uplifting and erosion from the middle to late Eocene (approximately 45–30 mya),
(5) Late Eocene (40–30 mya) intrusive volcanic activity in the island's center and northeast,
(6) Uplift, erosion and formation of marine limestone deposits during the Oligocene and Miocene (34–5 mya),
(7) Arching, uplift, and erosion in the period 5 mya to present.

These complex and sometimes simultaneous processes produced an island of intricate and multifaceted geologic character, which the U.S. Geological Survey (Bawiec, 1998) has divided into 151 map units and twelve terranes[1] (Fig. 1).

While it is generally correct to characterize this geological makeup as, "consisting of coastal alluvial plains skirted by limestone hills with interior central cordilleras primarily containing mixed complexes of metamorphic, intrusive and volcanic deposits of Jurassic to Miocene age", (Laffoon et al., 2012:2373), such a characterization obscures important variations in geological/depositional environment and age of formation/deposition. More importantly, the terrane map of Puerto Rico lays bare the non-discrete distribution of particular geological units; e.g., intrusive terranes of Cretaceous age appear in southwest, west-center, and southeast of the island, and the entirety of the island is encircled by nonvolcaniclastic terranes of Quaternary age. This stands in sharp contrasts with the geology of other areas (e.g., the Maya region), where geological age increases or decreases in a unidirectional manner and where strontium isotope studies of paleomigration have been particularly successful (e.g., Hodell et al., 2004).

Based on these geological data and the basic knowledge of strontium isotope systematics outlined above, we can make certain predictions about $^{87}Sr/^{86}Sr$ values in various parts of Puerto Rico. In general, it is expected that rocks from the island's volcaniclastic terranes will be characterized by depleted $^{87}Sr/^{86}Sr$ signatures (<0.706), and that these signatures will vary significantly according to age of formation and environment (marine versus subaerial). Conversely, the more recent formations of marine limestones that mantle the island are expected to exhibit more 'enriched' $^{87}Sr/^{86}Sr$ values (>0.707), again with slight variations stemming from temporal shifts in marine $^{87}Sr/^{86}Sr$ values at the time of their formation (Capo and DePaolo, 1990; McArthur et al., 2001).

4. Previous $^{87}Sr/^{86}Sr$ studies in the Caribbean and statement of problem

To date, no strontium isotope study of Puerto Rican paleomigration has been attempted. Indeed, it is only within the past five years that any such studies have been realized in the Caribbean writ large (Booden et al., 2008; Laffoon and de Vos, 2011; Schroeder

---

[1] Map units, "having affinities based upon lithologic rock type, depositional environment, and (or) age of deposition", (Bawiec, 1998).

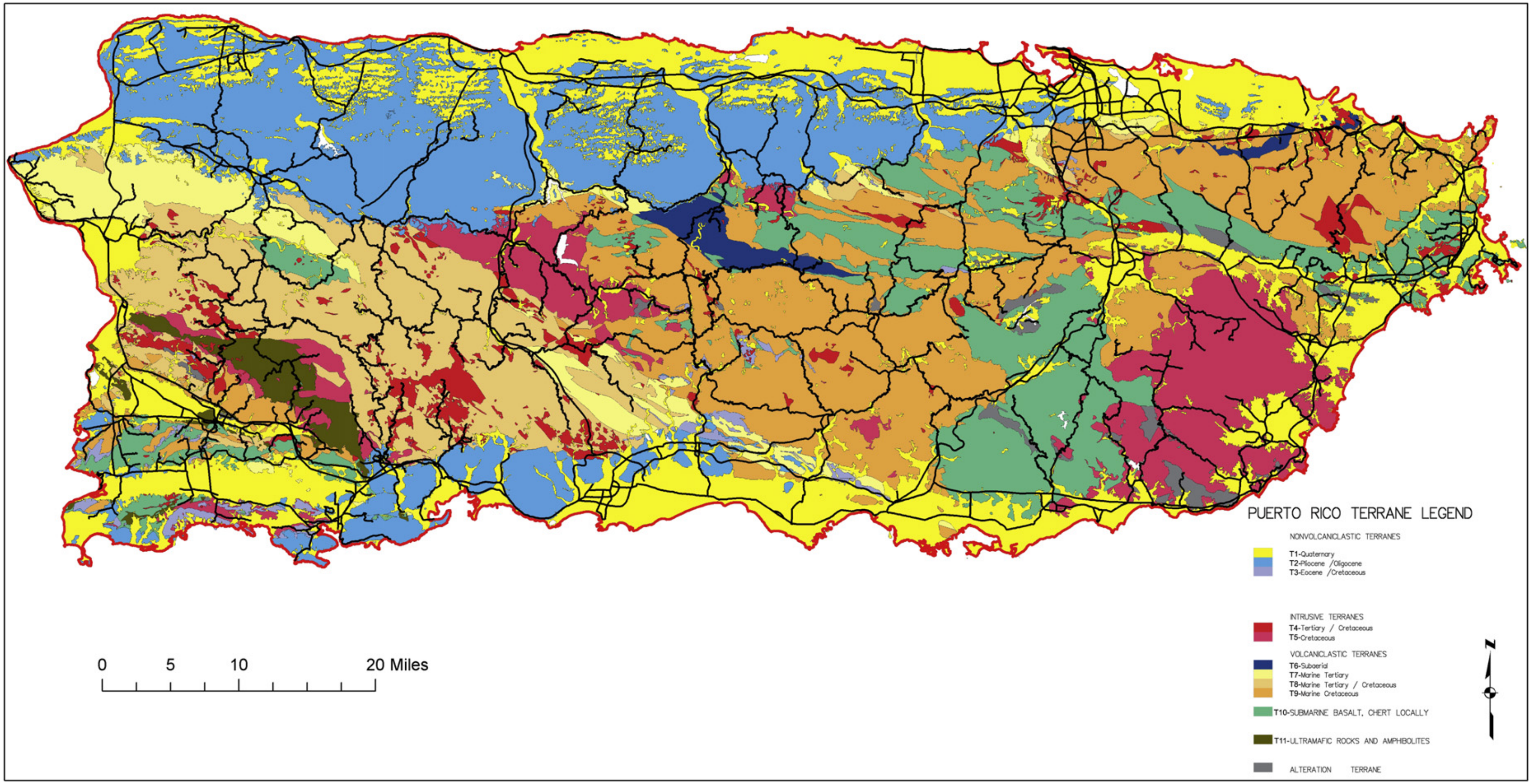

Fig. 1. Geological terrane map of Puerto Rico, adapted from Bawiec (1998).

et al., 2009; Valcárcel Rojas et al., 2011). Notably, all four of these studies identified non-local individuals (or at least putatively non-local individuals) in their respective study samples, affirming the dynamism of life in the prehistoric and historic Caribbean. At present, however, very little work aimed at establishing *baseline* $^{87}Sr/^{86}Sr$ variation within Puerto Rico has been attempted. While Laffoon et al. (2012) included a small number of samples (18) from Puerto Rico in their study of baseline bioavailable strontium variation across the Caribbean, nine of these eighteen samples came from just one site, Maisabel, leaving the other nine samples to characterize the remainder of the island. Furthermore, based on the noted findspots, this subset of nine samples overlooked significant portions of the island's geology. Finally, by reducing the complex geology of Puerto Rico to only two components (volcanic, intrusive, and ultramafic rocks *versus* Pliocene–Quaternary marine limestone), and by presenting an average $^{87}Sr/^{86}Sr$ value for the entire island (Laffoon et al., 2012:Figure 3), the data reported by Laffoon et al. (2012) would appear to inaccurately homogenize a complex isotopic landscape, something that the authors of that study would never do consciously.

In light of this dearth of baseline Sr isotope data for Puerto Rico, and ultimately as a means of combating any impression of island-wide simplicity or homogeneity, the present study was conceived in order to systematically assess the range of $^{87}Sr/^{86}Sr$ variation on an insular scale. This effort is just one small part of a large bioarchaeological study begun some seven years ago, and which to date has produced publication focusing on radiocarbon and light element (C&N) stable isotope analysis of human skeletal remains (Pestle, 2010a, 2010b; 2010c; Pestle and Colvard, 2012), and recently has realized genetic studies of these same remains. The working hypothesis of the strontium isotope portion of this larger study is that due to the complexity of Puerto Rican geology and the influence of other strontium sources any straightforward "mapping" of $^{87}Sr/^{86}Sr$ will be highly problematic. Assessment of this hypothesis requires a large sample set that is truly representative of the island's geologic diversity. Furthermore, it is our contention, subject to validation, that due to the non-discrete nature of Puerto Rico's geology, identification of place of origin for archaeological humans will be speculative and contingent.

5. Materials and methods

Based on the U.S. Geological Survey assessment of geological terranes presented in Bawiec (1998), the geology of Puerto Rico was divided into eleven analytical units (Fig. 1, Table 1). The "Alteration Terrane", the twelfth U.S.G.S. division, was excluded from this first iteration of baseline assessment given its inherent heterogeneity and small surface area (<1% of the island's landmass) (Bawiec, 1998). In July and August of 2010, two of the authors (WP and AC) collected bedrock samples from each of these eleven terranes noting sample location and altitude in a handheld GPS unit equipped with a barometric altimeter. In total, 47 geological samples were collected from these eleven terranes.

In addition, in six instances, terrestrial snail shells (genus *Caracolus*) were also collected from the immediate area of the geological samples. These paired rock-snail samples were collected in order to assess and account for differences between geological and bioavailable $^{87}Sr/^{86}Sr$. Snails, rather than the more typically employed mammalian fauna (Bentley and Knipper, 2005; Evans and Tatham, 2004; Ezzo et al., 1997; Knudson et al., 2004; Price and Getsdóttir, 2006; Price et al., 2000; Sjogren et al., 2009; Thornton, 2011), were employed for these purposes in the present case because of: 1) the depauperate terrestrial mammalian fauna of ancient Puerto Rico, 2) the potential influence of non-local foodstuffs on the $^{87}Sr/^{86}Sr$ values of modern rodents (Price et al., 2002:126), and 3) previous success in the use of snails for the characterization of local $^{87}Sr/^{86}Sr$ values (Price et al., 2002:126). All samples were air dried and stored in sterile sample bags for shipment to the University of Notre Dame for analysis.

Samples for Sr isotope analysis were prepared at the Department of Civil and Environmental Engineering and Earth Sciences, University of Notre Dame and analyzed using a NuPlasma II MC-ICP-MS instrument located at the MITERAC ICP-MS facility, University of Notre Dame. Rock samples were crushed and pulverized using traditional sample preparation methods. The rock powders were subsequently digested in 15 ml Savillex© Teflon vials using an HF:HNO_3 (4:1) acid mixture and placed on a hot plate at ~120 °C for 48 h. Digested samples were then uncapped and dried on a hot plate (~120 °C). The dried aliquots were then re-dissolved in concentrated HNO_3, vials were then recapped and place on a hot plate (~120 °C) and allowed to flux for 24 h; this step was repeated twice. All dried samples were dissolved in 3 ml of 0.75 N HCl and then loaded (0.25 ml aliquot) onto 10 cm ion exchange columns containing 1.42 ml of 200–400 mesh AG50W-X8 resin. Chemically separated, Sr-bearing aliquots of 5 ml of 2.5 N HCl each were collected into Teflon vials and then left to dry overnight on a hot plate (~100 °C). Subsequent to ion chromatographic treatment of the samples, the Sr-bearing aliquots were diluted in a 2% HNO_3 solution (~1.5 ml) and aspirated into the ICP torch using a desolvating nebulizing system (DSN-100 from Nu Instruments Inc., Wrexham, UK). Strontium isotope data were acquired in static, multicollection mode using five Faraday collectors for a total of 400 s, consisting of 40 scans of 10 s integrations. Accuracy and reproducibility of the analytical protocol were verified by the repeated analysis of a 150 ppb solution of the NIST SRM 987 strontium isotope standard during the course of this study; this yielded an average value of 0.71023 ± 0.00001 (2σ standard error; $n = 11$ analyses).

As discussed above, strontium available to plants and animals can be derived from sources other than bedrock geology. To account for these sources, reference was made to previously published values for atmospheric and dietary strontium sources. In insular settings, sea-spray and sea-derived precipitation, both of which have an enriched $^{87}Sr/^{86}Sr$ signature equivalent to their marine source (0.7092, (Capo and DePaolo, 1990; McArthur et al., 2001)), serve as extremely important sources of bioavailable strontium (Kennedy et al., 1998; Price and Getsdóttir, 2006; Vitousek et al., 1999; Whipkey et al., 2000). Marine enrichment of human $^{87}Sr/^{86}Sr$ could also be the result of the habitual consumption of marine foodstuffs, a dietary predilection that would appear to be the case at least in certain periods of Puerto Rican prehistory. Furthermore, in Puerto Rico, as in the Caribbean more generally, aerosolized dust from the Sahara Desert, which can be extremely abundant during the summer months (Prospero et al., 1981), presents another potential source of highly enriched ($^{87}Sr/^{86}Sr = 0.7157$) strontium (Borg and Banner, 1996).

6. Results

Results of the $^{87}Sr/^{86}Sr$ analysis are listed in Table 1. Five of the 47 geological samples (10.6%) failed to generate high quality (precise) Sr isotope ratios due to low ion signals. Three of these five failed samples are from the ultramafic/amphibolite unit, a type of rock that is generally deficient or poor in strontium (Stueber and Murthy, 1966:1244). As a consequence, we present no $^{87}Sr/^{86}Sr$ data for one of our terranes, T11. The other two failed samples were from the Tertiary/Cretaceous intrusive and Marine Tertiary volcaniclastic terranes, respectively. Finally, the $^{87}Sr/^{86}Sr$ results of two samples from the Quaternary nonvolcaniclastic terrane (T1) were omitted from consideration as their $^{87}Sr/^{86}Sr$ values (0.70595 and 0.70557) were markedly inconsistent with that unit's geology, as

Table 1
^{87}Sr/^{86}Sr values of Puerto Rican geological samples grouped by geological terrane.

Terrane	Age	Type	Sample #	Latitude	Longitude	Altitude (m)	^{87}Sr/^{86}Sr	2σ
T1	Quaternary	Nonvolcaniclastic	Y1	17.99	−66.03	45.2	0.70904	0.00007
			Y1	17.98	−66.67	61	0.70909	0.00002
			Y3	18.47	−66.21	−5.6	0.70906	0.00003
			Y4	18.01	−66.72	12.1	0.70862	0.00004
						Mean	**0.70895**	**0.00022**
T2	Pliocene/Oligocene	Nonvolcaniclastic	B1	18.01	−66.74	83.1	0.70859	0.00007
			B2	18.33	−66.95	239.8	0.70822	0.00001
			B3	18.37	−66.69	263.5	0.70849	0.00004
			B4	18.37	−66.47	93.1	0.70824	0.00002
			B5	18.33	−66.95	241	0.70815	0.00001
			B6	18.37	−66.69	256.4	0.70844	0.00006
						Mean	**0.70836**	**0.00018**
T3	Eocene/Cretaceous	Nonvolcaniclastic	V1	17.97	−66.94	53.2	0.70736	0.00002
			V2	18.00	−66.31	43	0.70753	0.00003
			V3	18.09	−66.59	238.7	0.70734	0.00002
			V4	17.96	−66.94	57.4	0.70701	0.00003
			V5	17.97	−66.92	7.1	0.70707	0.00004
			V6	17.96	−66.95	94.1	0.70706	0.00004
			V7	17.98	−67.06	45.4	0.70670	0.00002
			V8	18.06	−66.51	93.2	0.70650	0.00001
						Mean	**0.70707**	**0.00035**
T4	Tertiary/Cretaceous	Intrusive	R1	18.06	−66.63	166.4	0.70481	0.00001
			R2	18.10	−66.51	142	0.70500	0.00002
			R3	18.37	−65.89	49.6	0.70536	0.00002
						Mean	**0.70506**	**0.00028**
T5	Cretaceous	Intrusive	P1	18.20	−66.63	399.8	0.70457	0.00001
			P2	18.33	−66.47	142.7	0.70394	0.00002
			P3	17.98	−67.04	30.5	0.70686	0.00002
						Mean	**0.70512**	**0.00154**
T6	Subaerial	Volcaniclastic	DB1	18.30	−66.49	352	0.70445	0.00001
			DB2	18.30	−66.49	363.1	0.70406	0.00002
			DB3	18.30	−66.50	386.2	0.70407	0.00001
			DB4	18.28	−66.51	363.3	0.70453	0.00001
			DB5	18.36	−65.95	60.5	0.70461	0.00002
						Mean	**0.70434**	**0.00026**
T7	Marine Tertiary	Volcaniclastic	C1	18.32	−67.15	118.9	0.70665	0.00002
			C2	18.12	−66.66	479.1	0.70583	0.00001
			C3	18.10	−66.64	298.8	0.70611	0.00002
						Mean	**0.70620**	**0.00042**
T8	Marine Tertiary/Cretaceous	Volcaniclastic	T1	18.12	−66.61	348.7	0.70451	0.00001
			T2	18.21	−66.79	622.9	0.70412	0.00002
			T3	18.22	−67.16	53.4	0.70683	0.00004
						Mean	**0.70515**	**0.00147**
T9	Marine Cretaceous	Volcaniclastic	O1	18.25	−66.52	593.8	0.70440	0.00003
			O2	18.23	−66.53	565.5	0.70422	0.00001
			O3	18.00	−66.24	55.1	0.70499	0.00001
			O4	18.03	−66.44	118.8	0.70507	0.00002
						Mean	**0.70467**	**0.00042**
T10	–	Submarine Basalt	G1	18.00	−66.19	91.6	0.70597	0.00004
			G2	17.99	−66.16	94.7	0.70569	0.00002
			G3	18.04	−67.14	54.2	0.70465	0.00001
						Mean	**0.70544**	**0.00070**

compared to those other samples from that terrane. It seems likely that these samples were taken from intrusive rock formations not represented on the USGS terrane map.

Broken down by broad geological type (Fig. 2), the island's nonvolcaniclastic terranes have ^{87}Sr/^{86}Sr values between 0.7065 and 0.70909, with a mean of 0.70792 ± 0.00085 whereas the volcaniclastic terranes are more depleted, with ^{87}Sr/^{86}Sr values between 0.70394 and 0.70686, and a mean of 0.70505 ± 0.00090. Unsurprisingly, the difference in the ^{87}Sr/^{86}Sr signatures of these two broad groupings is statistically significant (Student's *t*-test, $p < 0.01$).

Assessing the ten terranes individually, the most enriched ^{87}Sr/^{86}Sr values were found in the three nonvolcaniclastic marine limestone units (T1, T2, and T3). T1, which is composed of Quaternary marine limestone is characterized by the highest average ^{87}Sr/^{86}Sr value (0.70895 ± 0.00022); this average value approaches the ^{87}Sr/^{86}Sr of modern seawater (0.7092). T2 and T3, terranes that are composed of successively older marine limestone formations (dating to the Pliocene/Oligocene and Eocene/Cretaceous, respectively) have increasingly depleted average ^{87}Sr/^{86}Sr values of 0.70836 ± 0.00018 and 0.70707 ± 0.00035, respectively.

The two intrusive terranes, T4 and T5, while entirely distinct from the marine limestones of T1–T3, are less clearly differentiated from one another and, in the case of T5, show a high degree of variability. The average ^{87}Sr/^{86}Sr value of T4, which is Tertiary/Cretaceous in age, is 0.70506 ± 0.00028, whereas the presumably older (Cretaceous) T5, which would be expected to have a more depleted ^{87}Sr/^{86}Sr signature, was found to average 0.70512 ± 0.00154. The variability in Sr isotope ratios for samples in this terrane (which range in individual values from 0.70394 to 0.70686) could be attributed to slight differences in the modal

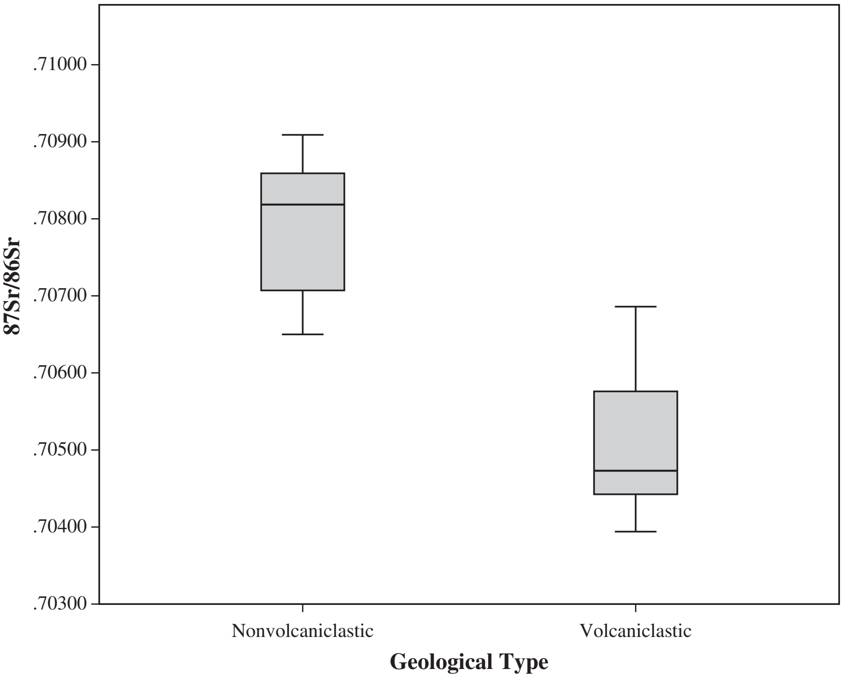

Fig. 2. ^{87}Sr/^{86}Sr values of Puerto Rican geological samples grouped by broad geological type.

abundances of constituent minerals within the rock units, or to variable degrees of weathering and alteration.

Turning next to the volcaniclastic terranes, rocks of T6, the subaerial volcaniclastic unit had the lowest average ^{87}Sr/^{86}Sr of any of the analyzed terranes at 0.70434 ± 0.00026. In the case of the marine volcaniclastic terranes (T7, T8, and T9), increasing geological age results in increasingly depleted ^{87}Sr/^{86}Sr average values, although a fair degree of variability and overlap are present. T7, which dates to the Tertiary, has the most enriched average ^{87}Sr/^{86}Sr signature of 0.70620 ± 0.00042, the Tertiary/Cretaceous T8 has a more depleted average ^{87}Sr/^{86}Sr of 0.70515 ± 0.00147, and the pure Cretaceous unit T9 has the lowest average ^{87}Sr/^{86}Sr signature at 0.70467 ± 0.00042. The final terrane, T10, which consists of submarine basalts, possesses an average ^{87}Sr/^{86}Sr signature of 0.70544 ± 0.00070, a value that overlaps substantially with a number of the marine and intrusive volcaniclastic terranes.

As seen in Fig. 3 and Table 2, comparison of the measured ^{87}Sr/^{86}Sr signatures of paired bedrock/snail shell samples ranged from an enrichment of 0.00164 to a depletion of 0.00007 from geological to bioavailable ^{87}Sr/^{86}Sr, with the latter value being at

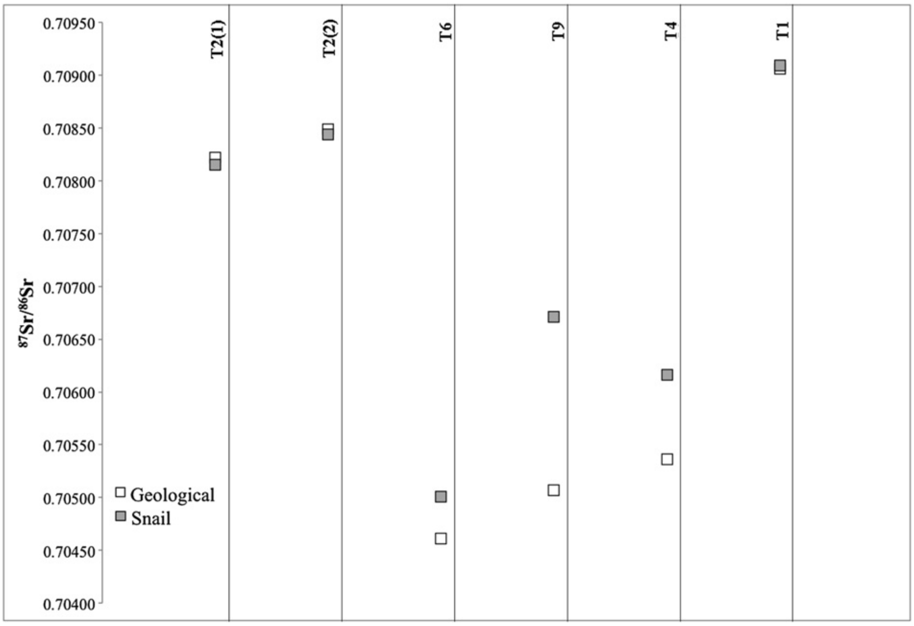

Fig. 3. ^{87}Sr/^{86}Sr values of paired geological-malacological (snail shell) samples.

Table 2
$^{87}Sr/^{86}Sr$ values of paired geological–malacological (snail shell) samples.

Sample pairing	Terrane	Material	$^{87}Sr/^{86}Sr$
T2(1)	T2	Geological	0.70822
		Snail	0.70815
		Δ	0.00007
T2(2)	T2	Geological	0.70849
		Snail	0.70844
		Δ	0.00005
T6	T6	Geological	0.70461
		Snail	0.70501
		Δ	−0.00040
T9	T9	Geological	0.70507
		Snail	0.70671
		Δ	−0.00164
T4	T4	Geological	0.70536
		Snail	0.70616
		Δ	−0.00080
T1	T1	Geological	0.70906
		Snail	0.70909
		Δ	−0.00003

the level of the analytical uncertainty associated with individual measurements. The average offset between geological and snail samples is a relative enrichment of 0.00046 ± 0.00067. This enrichment versus geological baseline values is likely the result of the uptake by these snails of comparatively enriched bioavailable strontium derived from oceanic or atmospheric sources. The difference in $^{87}Sr/^{86}Sr$ signatures of these geological and biological samples was essentially negligible (within the associated analytical uncertainties) in three of the six sets of paired samples, and only slightly higher (fourth significant digit) in two further instances. In fact, only in one case (the paired sample from T9, Marine Cretaceous Volcaniclastic) was the observed offset of such a magnitude (0.00164) as to affect interpretation. These findings generally confirm the utility of snails as a suitable proxy for local baseline geological $^{87}Sr/^{86}Sr$ values although they also confirm that other strontium sources (sea spray, atmospheric dust) may have a meaningful impact on local strontium isotope systems.

The values generated from this study (range, 0.70394 to 0.70909, mean 0.70626 ± 0.00164) are significantly different (Student's *t*-test, $p < 0.01$) from the Puerto Rican data presented by Laffoon et al. (2012:Appendix), which range from 0.70523 to 0.70916 and average 0.70787 ± 0.00131 (Fig. 4). This difference is likely the result of some combination of: a) the inclusion in the present study of $^{87}Sr/^{86}Sr$ depleted volcaniclastic terranes not represented in the data set of Laffoon et al., and/or b) the influence of $^{87}Sr/^{86}Sr$ enriched atmospheric and marine sources on the $^{87}Sr/^{86}Sr$ signatures of the biological samples that make up Laffoon et al.'s dataset. Of significant importance, however, is the fact that the results reported here attest to a greater degree of baseline variability in $^{87}Sr/^{86}Sr$ values than has been reported in previous studies.

This theme of broad baseline variation is further corroborated when $^{87}Sr/^{86}Sr$ results from this study are compared to a broad corpus ($n = 261$) of insular Caribbean values (range, 0.70480–0.70928, mean 0.70817 ± 0.00091; Laffoon et al., 2012: Appendix). Not only are the means of the two populations not significantly different (Student's *t*-test, $p = 0.339$) but, more crucially, the variation defined by the samples from Puerto Rico encompass almost entirely the total variation in $^{87}Sr/^{86}Sr$ values reported across the entire insular Caribbean (Fig. 4). The only $^{87}Sr/^{86}Sr$ compositions found in the islands of the Caribbean that are not present in the baseline samples from Puerto Rico emanate from areas composed of extremely recent marine limestones, and/or reflect the influence of seaspray or marine precipitation on bioavailable $^{87}Sr/^{86}Sr$ values.

The high degree of $^{87}Sr/^{86}Sr$ variability present in this corpus of modern Puerto Rican bedrock and terrestrial malacological samples is a testament to the complex geology of the island. As discussed below, this underlying geological complexity calls into question the efficacy of using $^{87}Sr/^{86}Sr$ isotopic studies to track paleomobility in the insular Caribbean.

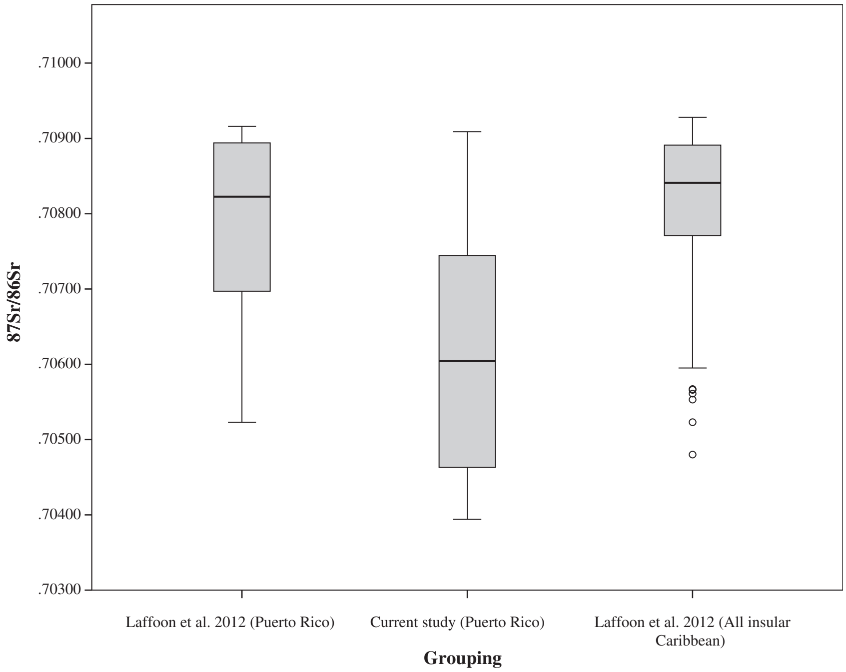

Fig. 4. $^{87}Sr/^{86}Sr$ values from current study of Puerto Rico (center column) as compared to values reported by Laffoon et al. (2012) for Puerto Rico (left) and entire insular Caribbean (right).

7. Discussion and conclusion

The temptation to use biogeochemical techniques to resolve issues of paleomigration is evident and well intentioned. To do so, however, requires detailed knowledge of appropriate baseline values and the sophisticated interpretation thereof. While geological circumstances in certain areas of the world (e.g. the Maya region (Hodell et al., 2004), or Scandinavia (Frei and Price, 2012) may have sufficient discrete geological variation to permit "mapping" of baseline $^{87}Sr/^{86}Sr$ values, thereby facilitating determination of the point-of-origin of non-local individuals, not all areas of interest are as accommodating. As discussed above, it was our hypothesis that the complexity of Puerto Rican geology and the influence of other strontium sources would serve to make the "mapping" of $^{87}Sr/^{86}Sr$ on the island, if not in the insular Caribbean more broadly, highly problematic. We contend that the results of the present study strongly support such a hypothesis and should serve as a strong note of caution for isotopic studies of paleomigration in this region. Three main takeaway points flow from the present work.

First, given the enormous variability in baseline $^{87}Sr/^{86}Sr$ values noted in Puerto Rico, any notion of average or "typical" $^{87}Sr/^{86}Sr$ signatures for this, or perhaps any, island ought to be dismissed. At best, such averages are of little utility, and at worst they might give the impression of a degree of simplicity and homogeneity that flies in the face of the broad diversity attested to by the data. Caribbean islands, and in particular the more geologically complex Greater Antilles, would not appear to be appropriate as units of analysis for isotopic studies of paleomigration. That two islands have different average $^{87}Sr/^{86}Sr$ signatures is largely meaningless for the study of paleomigration if the ranges of their bioavailable strontium isotopic values overlap. Only in those instances where an individual's $^{87}Sr/^{86}Sr$ signature falls outside the range of local bioavailable strontium isotopic values can something meaningful be said about non-local origins.

Second, as isotopically similar or identical geological formations exist in multiple locales in Puerto Rico, the assessment of baseline strontium variability by geological sub-regions seems ill advised. While such an approach works well in places like the Yucatan, where the underlying geology consists of more-or-less spatially discrete bands of temporally and isotopically distinct marine limestone outcroppings, it is not well suited to a situation like the Caribbean, where the same rock types appear in multiple distinct locations. As Hodell et al., (2004:585) observe, "the existence of geologic terrains with distinct strontium isotopic signatures makes the Maya area of Mesoamerica an especially attractive place to apply this technique." Lacking such distinct and discrete geological terrains/terranes, the conclusion that rocks of different age and type differ in their $^{87}Sr/^{86}Sr$ signatures is of little utility for the purposes of archaeological inference.

Third, this situation is made more complex by the finding that the degree of $^{87}Sr/^{86}Sr$ variability seen in the bedrock geology of Puerto Rico more-or-less completely subsumes the variation in $^{87}Sr/^{86}Sr$ found across the insular Caribbean. The implication of this finding is that for any potential human $^{87}Sr/^{86}Sr$ value, there are a host of non-unique places of origin, from many different islands, all of which would "solve" the isotopic question at hand. Such a state of affairs casts significant doubt on our ability, at least using $^{87}Sr/^{86}Sr$ alone, to "map" points of origin for individuals possessing isotopic signatures distinct from their place of burial.

Ultimately, based on the results presented here, it would appear that $^{87}Sr/^{86}Sr$ analysis of paleomigration in a Puerto Rican, and perhaps broader Caribbean, context could hope to do little more than identify individuals with $^{87}Sr/^{86}Sr$ values distinct from their place of death and burial. While it may be tempting to divide a sample of individuals into "local" and "non-local" categories, given the findings presented herein, both of these classifications would seem to be highly contingent. An individual found in one of the many Precolumbian sites dotting the Puerto Rican coastal plains and possessing an enamel $^{87}Sr/^{86}Sr$ value consistent with the local geology could have been born and raised at that site, or, indeed, could have hailed from any other area of the island (or, indeed, other islands) possessing marine limestone geology of comparable age. On the basis of Sr isotope ratios, individuals from the opposite coasts of Puerto Rico will appear identical. Conversely, a putatively "non-local" individual found at the same site could originate from any of the myriad geological formations within, or external to, Puerto Rico that would still be entirely consistent with their enamel $^{87}Sr/^{86}Sr$ value.

Given the complexity of the Puerto Rican geological regime, the very meanings of terms like "local" and "non-local" are rendered more-or-less meaningless. Indeed, as a consequence of the non-discrete geology of the island, someone with a "non-local" $^{87}Sr/^{86}Sr$ value of their enamel could have been born and raised in closer proximity to the site of their death than a presumed "local". Equally, someone from a site five miles away would appear just as "exotic" (isotopically) as someone from an entirely differently island possessing the same baseline $^{87}Sr/^{86}Sr$ signature as the more local "exotic" outcropping. Ultimately, the diversity of $^{87}Sr/^{86}Sr$ values in Puerto Rico makes parsing such issues of origin a difficult and highly contingent task. Given these complexities, any studies seeking to assess paleomigration by such isotopic means should proceed with a great deal of caution, and look toward employing multiple mobility measures (including, for instance, oxygen isotopes or aDNA).

Acknowledgments

Portions of this material are based upon work supported by the National Science Foundation under Grant Number BCS-0612727. Our thanks are due to Joshua Torres for his assistance with mapping and GIS and to Gordon Ambrosino, Barney Nashold, Mariela Declet, Daniel Curet, and Carmen and Hernan Rivera for their help with sample collection.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jas.2013.01.020.

References

Åberg, G., Fosse, G., Stray, H., 1998. Man, nutrition and mobility: a comparison of teeth and bone from the Medieval era and the present from Pb and Sr isotopes. The Science of the Total Environment 224, 109–119.

Bawiec, W.J., 1998. Geologic terranes of Puerto Rico. In: Bawiec, W.J. (Ed.), U.S Geological Survey Open-file Report 98-38: Geology, Geochemistry, Geophysics, Mineral Occurrences and Mineral Resource Assessment for the Commonwealth of Puerto Rico. U.S. Geological Survey, Washington, D.C.

Bentley, R.A., 2006. Strontium isotopes from the Earth to the archaeological skeleton: a review. Journal of Archaeological Method and Theory 13, 135–187.

Bentley, R.A., Knipper, C., 2005. Geographical patterns in biologically available strontium, carbon and oxygen isotope signatures in prehistoric SW Germany. Archaeometry 47, 629–644.

Blum, J.D., Taliaferro, E.H., Weisse, M.T., Holmes, R.T., 2000. Changes in Sr/Ca, Ba/Ca, and $^{87}Sr/^{86}Sr$ ratios between trophic levels in two forest ecosystems in the Northeastern USA. Biogeochemistry 49, 87–101.

Booden, M.A., Panhuysen, R.G.A.M., Hoogland, M.L.P., de Jong, H.N., Davies, G.R., Hofman, C.L., 2008. Tracing human mobility with $^{87}Sr/^{86}Sr$ at Anse Á la Gourde, Guadeloupe. In: Hofman, C.L., Hoogland, M.L.P., van Gijn, A.L. (Eds.), Crossing the Borders: New Methods and Techniques in the Study of Archaeological Materials from the Caribbean. University of Alabama Press, Tuscaloosa, pp. 214–225.

Borg, L.E., Banner, J.L., 1996. Neodymium and strontium isotopic constraints on soil sources in Barbados, West Indies. Geochimica et Cosmochimica Acta 60, 4193–4206.

Budd, P., Millard, A., Chenery, C., Lucy, S., Roberts, C., 2004. Investigating population movement by stable isotope analysis: a report from Britain. Antiquity 78, 127–141.

Buzon, M.R., Simonetti, A., Creaser, R.A., 2007. Migration in the Nile Valley during the New Kingdom period: a preliminary strontium isotope study. Journal of Archaeological Science 34, 1391–1401.

Capo, R.C., DePaolo, D.J., 1990. Seawater strontium isotopic variations from 2.5 million years ago to the present. Science 249, 51–55.

Chadwick, O.A., Derry, L.A., Vitousek, P.M., Huebert, B.J., Hedin, L.O., 1999. Changing sources of nutrients during four million years of ecosystem development. Nature 397, 491–497.

Cox, D.P., 1985. Geology of the Tanama and Helecho Porphyry Copper Deposits and Vicinity, Puerto Rico. U.S. Geological Survey Professional Paper 1327. U.S. Geological Survey, Washington, D.C.

Ericson, J.E., 1985. Strontium isotope characterization in the study of prehistoric human ecology. Journal of Human Evolution 14, 503–514.

Evans, J.A., Tatham, S., 2004. Defining 'local signature' in terms of Sr isotope composition using a tenth- to twelfth-century Anglo-Saxon population living on a Jurassic clay-carbonate terrain, Rutland, UK. In: Pye, K., Croft, D.J. (Eds.), Forensic Geoscience: Principles, Techniques and Applications. Geological Society, London, pp. 237–248.

Ezzo, J.A., Johnson, C.M., Price, T.D., 1997. Analytical perspectives on prehistoric migration: a case study from East-Central Arizona. Journal of Archaeological Science 24, 447–466.

Faure, G., 1986. Principles of Isotope Geology. John Wiley, New York.

Faure, G., Powell, J.L., 1972. Strontium Isotope Geology. Springer-Verlag, New York.

Frei, K.M., Price, T.D., 2012. Strontium isotopes and human mobility in prehistoric Denmark. Archaeological and Anthropological Sciences 4, 103–114.

Hillson, S., 2005. Teeth, second ed. Cambridge University Press, Cambridge.

Hodell, D.A., Quinn, R.L., Brenner, M., Kamenov, G., 2004. Spatial variation of strontium isotopes (^{87}Sr/^{86}Sr) in the Maya region: a tool for tracking ancient human migration. Journal of Archaeological Science 31, 585–601.

Kennedy, M.J., Chadwick, O.A., Vitousek, P.M., Derry, L.A., Hendricks, D.M., 1998. Changing sources of base cations during ecosystem development, Hawaiian Islands. Geology 26, 1015–1018.

Knudson, K.J., Price, T.D., Buikstra, J.E., Blom, D.E., 2004. The use of strontium isotope analysis to investigate Tiwanaku migration and Mortuary Ritual in Bolivia and Peru. Archaeometry 46, 5–18.

Krushensky, R.D., Schellekens, J.H., 1998. Geology of Puerto Rico. In: Bawiec, W.J. (Ed.), U.S Geological Survey Open-file Report 98-38: Geology, Geochemistry, Geophysics, Mineral Occurrences and Mineral Resource Assessment for the Commonwealth of Puerto Rico. U.S. Geological Survey, Washington, D.C.

Laffoon, J.E., Davies, G.R., Hoogland, M.L.P., Hofman, C.L., 2012. Spatial variation of biologically available strontium isotopes (^{87}Sr/^{86}Sr) in an archipelagic setting: a case study from the Caribbean. Journal of Archaeological Science 39, 2371–2384.

Laffoon, J.E., de Vos, B.R., 2011. Diverse origins, similar diets: an integrated isotopic perspective from Anse à la Gourde, Guadeloupe. In: Hofman, C.L., van Duijvenbode, A. (Eds.), Communities in Contact: Essays in Archaeology, Ethnohistory & Ethnography of the Amerindian Circum-Caribbean. Sidestone Press, Leiden, pp. 187–204.

Mattson, P.H., 1960. Geology of the Mayagues area, Puerto Rico. Geological Society of America Bulletin 71, 319–361.

McArthur, J.M., Howarth, R.W., Bailey, T.R., 2001. Strontium isotope stratigraphy: LOWESS Version 3: best fit to the marine Sr-isotope curve for 0–509 Ma and accompanying look-up table for deriving numerical age. The Journal of Geology 109, 155–170.

Nelson, B.K., DeNiro, M.J., Schoeninger, M.J., DePaolo, D.J., Hare, P.E., 1986. Effects of diagenesis on strontium, carbon, nitrogen, and oxygen concentration and isotopic concentration of bone. Geochimica et Cosmochimica Acta 50, 1941–1949.

Pestle, W.J., 2010a. Bone chemistry and paleodiet at the ceremonial center of Tibes. In: Curet, L.A., Stringer, L.M. (Eds.), Tibes: People, Power, and Ritual at the Center of the Cosmos. University of Alabama Press, Tuscaloosa, pp. 209–230.

Pestle, W.J., 2010b. Chemical, elemental, and isotopic effects of acid concentration and treatment duration on ancient bone collagen: an exploratory study. Journal of Archaeological Science 37, 3124–3128.

Pestle, W.J., 2010c. Diet and Society in Prehistoric Puerto Rico, an Isotopic Approach. Department of Anthropology, University of Illinois at Chicago.

Pestle, W.J., Colvard, M., 2012. Bone collagen preservation in the Tropics: a case study from ancient Puerto Rico. Journal of Archaeological Science 39, 2079–2090.

Picó, R., 1974. The Geography of Puerto Rico. Aldine, Chicago.

Price, T.D., Burton, J.A., Bentley, R.A., 2002. The characterization of biologically available strontium isotopes for the study of prehistoric migrations. Archaeometry 44, 117–135.

Price, T.D., Getsdóttir, H., 2006. The first settlers of Iceland: an isotopic approach to colonisation. Antiquity 80, 130–144.

Price, T.D., Knipper, C., Grupe, G., Smrcka, V., 2004. Strontium isotopes and prehistoric human migration: the Bell Beaker period in central Europe. European Journal of Archaeology 7, 9–40.

Price, T.D., Middleton, W.D., Manzanilla, L., 2000. Immigration and the ancient city of Teotihuacan in Mexico: a study using strontium isotope ratios in human bone and teeth. Journal of Archaeological Science 27, 903–913.

Prospero, J.M., Glaccum, R.A., Nees, R.T., 1981. Atmospheric transport of soil dust from Africa to South America. Nature 289, 570–572.

Schroeder, H., O'Connell, T.C., Evans, J.A., Shuler, K.A., Hedges, R.E.M., 2009. Trans-Atlantic slavery: isotopic evidence for forced migration to Barbados. American Journal of Physical Anthropology 139, 547–557.

Sjogren, K.-G., Price, T.D., Ahlstrom, T., 2009. Megaliths and mobility in south-western Sweden. Investigating relationships between a local society and its neighbors using strontium isotopes. Journal of Anthropological Archaeology 28, 85–101.

Stueber, A.M., Murthy, V.R., 1966. Strontium isotope and alkali element abundances in ultramafic rocks. Geochimica et Cosmochimica Acta 30, 1243–1259.

Thornton, E.K., 2011. Reconstructing ancient Maya animal trade through strontium isotope (^{87}Sr/^{86}Sr) analysis. Journal of Archaeological Science 38, 3254–3263.

Valcárcel Rojas, R., Weston, D.A., Mickleburgh, H.L., Laffoon, J.E., van Duijvenbode, A., 2011. El Chorro de Maíta. A diverse approach to a context of diversity. In: Hofman, C.L., van Duijvenbode, A. (Eds.), Communities in Contact: Essays in Archaeology, Ethnohistory & Ethnography of the Amerindian Circum-Caribbean. Sidestone Press, Leiden, pp. 225–252.

Vitousek, P.M., Kennedy, M.J., Derry, L.A., Chadwick, O.A., 1999. Weathering versus atmospheric sources of strontium in ecosystems on young volcanic soils. Oecologia 121, 255–259.

Whipkey, C.E., Capo, R.C., Chadwick, O.A., Stewart, B.W., 2000. The importance of sea spray to the cation budget of a coastal Hawaiian soil: a strontium isotope approach. Chemical Geology 168, 37–48.

Wright, L.E., 2005. Identifying immigrants to Tikal, Guatemala: defining local variability in strontium isotope ratios of human tooth enamel. Journal of Archaeological Science 32, 555–566.