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

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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 paleomobility by such isotopic means should proceed with a great deal of caution.

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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 archaeology (Åberg et al., 1998; Bentley, 2006; Buzon et al., 2007; Ezzo et al., 1997; Frei and Price, 2012; Knudson et al., 2004; Price and Gestsdóttir, 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%)...
renders isotopic fractionation through biological processes insig-
nificant (Fauve and Powell, 1972). Strontium abundances and cor-
responding 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 re-
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 for-
aging on that vegetation. Hence, the strontium isotopic composi-
tion of an individual’s or animal’s diet will be recorded in the
corresponding hard tissues (Ercison, 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 bio-
available Sr present in the geographic area in which a person or
animal lived while the tooth was forming. Minerals that may have
dformed 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 at-
mospheric strontium sources, in particular seaspray and oceanic-
derived precipitation (Kennedy et al., 1998; Price and Getsdötter,
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 in-
terest. 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 is-
land 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,
behind 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 Cor-
dillera 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 dif-
fering 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 sedi-
mentary rocks from the middle through the end of the Creta-
ceous (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 pro-
duced 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 lime-
stone 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 terrace 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 accord-
ing to age of formation and environment (marine versus subaerial).
Conversely, the more recent formations of marine limestones that
mantine the island are expected to exhibit more “enriched” \(^{87}\)Sr/\(^{86}\)Sr values
(>0.707), again with slight variations stemming from tem-
poral shifts in marine \(^{86}\)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 paleomi-
igration has been attempted. Indeed, it is only within the past five
years that any such studies have been realized in the Caribbean writ
large (Booeden 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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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, Masisel, 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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 for these study-wide SRU-bearing data, the present study was undertaken in order to systematically assess the range of $^{87}\text{Sr}/^{86}\text{Sr}$ variation on an insular scale. This effort is just one small part of a large bio-archaeological 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 straight-forward “mapping” of $^{87}\text{Sr}/^{86}\text{Sr}$ 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 (genre 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 $^{87}\text{Sr}/^{86}\text{Sr}$. 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 Gotsdottir, 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 food-stuffs on the $^{87}\text{Sr}/^{86}\text{Sr}$ values of modern rodents (Price et al., 2002:126), and 3) previous success in the use of snails for the characterization of local $^{87}\text{Sr}/^{86}\text{Sr}$ 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 Nu Plasma 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) 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 sample liquid, Sr-bearing aliquots were diluted 3:1 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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 Gotsdottir, 2006; Vitousek et al., 1999; Whippy et al., 2000). Marine enrichment of human $^{87}\text{Sr}/^{86}\text{Sr}$ 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, aerolized 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 ($^{87}\text{Sr}/^{86}\text{Sr}$ = 0.7157) strontium (Borg and Banner, 1996).

6. Results

Results of the $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ data for one of our terranes, T11. The other two failed samples were from the Tertiary/Cretaceous intrusive and Marine Tertiary volcanioclastic terranes, respectively. Finally, the $^{87}\text{Sr}/^{86}\text{Sr}$ results of two samples from the Quaternary nonvolcanioclastic terrane (T1) were omitted from consideration as their $^{87}\text{Sr}/^{86}\text{Sr}$ values (0.70595 and 0.70557) were markedly inconsistent with that unit’s geology, as
two broad groupings is statistically 
significant. Marine limestone is characterized by the highest average 
nonvolcaniclastic terranes have 
Unsurprisingly, the difference in the 
Table 1 
Sr/
T1 Quaternary Nonvolcaniclastic V1 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 0.70512 0.000154 
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 0.70868 0.00002 
Mean 0.70512 0.000154 
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 0.000042 
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 0.000147 
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 0.000042 
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 0.00007 

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 \(^{87}\text{Sr}/^{86}\text{Sr}\) values between 0.7065 
and 0.70909, with a mean of 0.70792 \pm 0.00085 whereas the volca-
niclastic terranes are more depleted, with \(^{87}\text{Sr}/^{86}\text{Sr}\) values be-
tween 0.70394 and 0.70686, and a mean of 0.70505 \pm 0.00090. 
Unsurprisingly, the difference in the \(^{87}\text{Sr}/^{86}\text{Sr}\) signatures of these 
methods is statistically significant (Student's t-test, 
P < 0.01).

Assessing the ten terranes individually, the most enriched 
\(^{87}\text{Sr}/^{86}\text{Sr}\) values were found in the three nonvolcaniclastic marine 
limestone units (T1, T2, and T3). T1, which is composed of Quar-
ternary marine limestone is characterized by the highest average 
\(^{87}\text{Sr}/^{86}\text{Sr}\) value (0.70895 \pm 0.00022); this average value 
approaches the \(^{87}\text{Sr}/^{86}\text{Sr}\) of modern seawater (0.7092). T2 and T3, 
terranes that are composed of successively older marine lime-
stone formations (dating to the Pliocene/Oligocene and Eocene/
Cretaceous, respectively) have increasingly depleted average 
\(^{87}\text{Sr}/^{86}\text{Sr}\) values of 0.70836 \pm 0.00018 and 0.70707 \pm 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 \(^{87}\text{Sr}/^{86}\text{Sr}\) value of T4, which is Tertiary/
Cretaceous in age, is 0.70506 \pm 0.00028, whereas the presumably 
older (Cretaceous) T5, which would be expected to have a more 
depleted \(^{87}\text{Sr}/^{86}\text{Sr}\) signature, was found to average 
0.70512 \pm 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 

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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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ average values, although a fair degree of variability and overlap are present. T7, which dates to the Tertiary, has the most enriched average $^{87}\text{Sr}/^{86}\text{Sr}$ signature of 0.70620 ± 0.00042, the Tertiary/Cretaceous T8 has a more depleted average $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.70515 ± 0.00147, and the pure Cretaceous unit T9 has the lowest average $^{87}\text{Sr}/^{86}\text{Sr}$ signature at 0.70467 ± 0.00042. The final terrane, T10, which consists of submarine basalts, possesses an average $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ signatures of paired bedrock/snail shell samples ranged from an enrichment of 0.00164 to a depletion of 0.00007 from geological to bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$, with the latter value being at
Table 2

<table>
<thead>
<tr>
<th>Sample pairing</th>
<th>Terrane</th>
<th>Material</th>
<th>(^{87/86}\text{Sr})</th>
</tr>
</thead>
<tbody>
<tr>
<td>T2(1)</td>
<td>T2</td>
<td>Geological</td>
<td>0.70822</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Snail</td>
<td>0.70815</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(\Delta)</td>
<td>0.00007</td>
</tr>
<tr>
<td>T2(2)</td>
<td>T2</td>
<td>Geological</td>
<td>0.70849</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Snail</td>
<td>0.70844</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(\Delta)</td>
<td>0.00005</td>
</tr>
<tr>
<td>T6</td>
<td>T6</td>
<td>Geological</td>
<td>0.70461</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Snail</td>
<td>0.70501</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(\Delta)</td>
<td>-0.00040</td>
</tr>
<tr>
<td>T9</td>
<td>T9</td>
<td>Geological</td>
<td>0.70507</td>
</tr>
<tr>
<td></td>
<td></td>
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<tr>
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<td>(\Delta)</td>
<td>-0.00003</td>
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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 \(^{87/86}\text{Sr}\) 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, Modern 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 \(^{87/86}\text{Sr}\) 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 \(^{87/86}\text{Sr}\) depleted volcaniclastic terranes not represented in the data set of Laffoon et al., and/or b) the influence of \(^{87/86}\text{Sr}\) enriched atmospheric and marine sources on the \(^{87/86}\text{Sr}\) 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 \(^{87/86}\text{Sr}\) values than has been reported in previous studies.

This theme of broad baseline variation is further corroborated when \(^{87/86}\text{Sr}\) 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 encompasses almost entirely the total variation in \(^{87/86}\text{Sr}\) values reported across the entire insular Caribbean (Fig. 4). The only \(^{87/86}\text{Sr}\) 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 \(^{87/86}\text{Sr}\) values.

The high degree of \(^{87/86}\text{Sr}\) 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 \(^{87/86}\text{Sr}\) isotopic studies to track paleomobility in the insular Caribbean.
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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 stratigraphic sources would serve to make the “mapping” of $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ values noted in Puerto Rico, any notion of average or “typical” $^{87}\text{Sr}/^{86}\text{Sr}$ signature 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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ 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 outcrops, 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, S85) 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 isotopic studies of paleomigration. That two islands have different $^{87}\text{Sr}/^{86}\text{Sr}$ 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 $^{87}\text{Sr}/^{86}\text{Sr}$ signature falls outside the range of local bioavailable strontium isotopic values can something meaningful be said about non-local origins.

Finally, 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” $^{87}\text{Sr}/^{86}\text{Sr}$ 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, all using the same baseline $^{87}\text{Sr}/^{86}\text{Sr}$ signature as the more local “exotic” outcropping. Ultimately, the diversity of $^{87}\text{Sr}/^{86}\text{Sr}$ 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 DNA).

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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


