Isotopes, isoscapes, and the search for geographic origins: unrealized potential or unrealisitc expectations

Laffoon, J.E.; Sonnemann, T.F.; Kalayci, T.; Lambers, K.; Klinkenberg, V.

DIGITAL ARCHAEOLOGY
Promises and Impasses

51

ANALECTA PRAEHISTORICA LEIDENSIA

edited by
TUNA KALAYCI, KARSTEN LAMBERS
& VICTOR KLINKENBERG

DIGITAL ARCHAEOLOGY
Promises and Impasses

edited by

TUNA KALAYCI, KARSTEN LAMBERS
AND VICTOR KLINKENBERG

ANALECTA
PRAEHISTORICA
LEIDENSIA 51

Published by Sidestone Press, Leiden
www.sidestone.com

Series: Analecta Praehistorica Leidensia
Series editors: V. Klinkenberg, R. van Oosten, R. Jansen and C. van Driel-Murray

Lay-out & cover design: Sidestone Press

ISBN 978-94-6426-227-8 (softcover)
ISBN 978-94-6426-228-5 (hardcover)
ISBN 978-94-6426-229-2 (PDF e-book)

ISSN 0169-7447 (Print)
ISSN 2665-9573 (Online)

DOI 10.59641/f48820ir

Universiteit Leiden

Contents

Kalaycı, T., K. Lambers and V. Klinkenberg. 2023. "Digital Archaeology: Promises and Impasses". *Analecta Praehistorica Leidensia* 51. Leiden: Sidestone Press, pp. 57-75. DOI: 10.59641/f48820ir.

Isotopes, Isoscapes, and the Search for Geographic Origins: Unrealized Potential or Unrealistic Expectations?

Jason E. Laffoon and Till F. Sonnemann

1 Introduction

Isotope analysis has become an increasingly popular and utilized tool to investigate geographic origins in archaeology. Since the first application of isotope methods to assess human migrations and mobility over thirty years ago (Ericson 1985), these methods have developed to become one of the most commonly employed approaches to the study of human (and animal) paleomobility in archaeological research. Traditionally, most applications of this method have focused on simply distinguishing between locals and nonlocals. Over the last decade, however, there has been a concerted effort to develop spatially explicit databases of baseline (bioavailable) strontium and oxygen isotope variation. When applied at large scales these data can be used to generate maps or predictive models (isoscapes) of spatial isotopic variation in order to assess individual geographic origins (Bowen 2010; West *et al*. 2010). These isoscapes have been developed for different isotope systems, various sample materials, and many areas of the world at multiple scales from regional, to sub-continental, to global (*e.g.*, Adams *et al*. 2019; Bataille and Bowen 2012; Bataille *et al*. 2012; 2018; 2020; Britton *et al*. 2020; Funck *et al*. 2021; Hedman *et al*. 2018; Hobson *et al*. 2012; Laffoon *et al*. 2012; Lugli *et al*. 2021; Scaffidi and Knudson 2020). Nevertheless, to date, there has been insufficient research testing the reliability of these isoscapes for their intended purpose of determining individual origins. Despite some preliminary evidence of the efficacy of such isoscapes for archaeological and forensic human provenance research (Bataille *et al*. 2021; Colleter *et al*. 2021; Ehleringer *et al*. 2010; Font *et al*. 2015a; Laffoon *et al*. 2017), much more systematic testing of the validity of these models, using individuals of known origin, is needed before these can be more broadly and effectively applied to archaeological studies of migration and mobility, generally, and of individual origins in particular.

In this paper, we assess the reliability of extant Circum-Caribbean strontium and oxygen isoscapes using isotope data obtained from modern individuals of known childhood origins. The current study represents a follow up to a previous study that used dual isotopes and isoscapes (strontium and oxygen) to test and validate this approach. In this previous study (Laffoon *et al*. 2017), one modern individual of known origin from Caracas (Venezuela), and two (nonlocal) archaeological individuals recovered from sites on Trinidad and Cuba were tested. The results of this previous study demonstrated that: many areas within the broader macro-region (Circum-Caribbean) could be eliminated as possible places of origin based on a single isotope value; potential places of origin were further narrowed by using two isotopes proxies; the approach accurately identified

Jason E. Laffoon
Faculty of Archaeology
Leiden University
PO Box 9514
2300 RA Leiden
The Netherlands
j.e.laffoon@arch.leidenuniv.nl

Till F. Sonnemann
Bonn Center for Digital Humanities
University of Bonn
till.sonnemann@uni-bonn.de

Caracas as a possible place of origin for the modern individual; and the suspected geographic origins of the archaeological individuals (based on other lines of evidence) were consistent with the possible origins based on the dual-isoscape approach (Laffoon *et al*. 2017). The original study, however, only included one (modern) known origin individual, which is insufficient to robustly test the isoscape approach. The current study includes further testing of the isoscape method based on six additional modern, known-origin individuals including four individuals with just strontium isotope data, and two more with both strontium and oxygen isotope data. The results of the current study demonstrate considerable variation in the efficacy of the isoscape approach for constraining childhood origins and illustrate areas where the predictive models may need to be improved before they can be more widely utilized for the accurate and precise identification of individual geographic origins in archaeological and forensic research.

Placing this chapter within the broader aims of this volume, exploring promises and impasses of digital archaeology, isoscapes are increasingly popular tools for investigating origins in widely diverse research fields. These include sourcing and authentication studies in food science (Schellenberg *et al.* 2010), tracing the illicit trade in elephant ivory (Van der Merwe *et al.* 1990) and illegal drugs (Galimov *et al.* 2005), tracking animal migrations in wildlife ecology (Hobson *et al.* 2010), determining the origins of human remains from forensic contexts (unidentified individuals, mass graves) (Chesson *et al.* 2018), and exploring human migrations in archaeology (Lee-Thorp 2008). This increased popularity can be attributed to several factors: such as the empirical nature of isotope methods; the increased throughput, reduced costs and improved accuracy of isotope measurements; the ability to apply them to a broad range of materials and research questions; and perhaps most importantly their vast potential for answering one of the most fundamental of questions: ‘where does someone (or something) come from?’. To date, however, the vast potential of isoscape approaches for researching origins has yet to be realized in part due to the fact that the science is quite young and fast developing, because large-scale reliable isoscapes are not yet available for many regions of the world, and due to the fact that extant isoscapes have not yet been systematically tested to validate that they are actually effective tools for such purposes. This case study represents an explicit attempt to test the validity of current Circum-Caribbean isoscapes for the purpose of constraining human geographic origins.

2 Isotope principles

Isotope analysis has a long history of applications in archaeological research. Analysis of the radioactive isotopes of carbon (carbon-14) forms the basis of the most commonly employed absolute dating method in archaeology. Stable isotope analyses have also been widely used for dietary studies (carbon and nitrogen) and for climate (oxygen) and environmental (carbon and oxygen) reconstructions (Lee-Thorp 2008). Isotope analysis of human remains was first proposed and applied to the study of human mobility using strontium isotopes (Ericson 1985) and later oxygen isotopes (Fricke *et al*. 1995; Schwarcz *et al*. 1991). Over the last several decades strontium and oxygen isotopes have been increasingly applied in archaeological research, to the extent that they now form, in addition to aDNA, one of the most commonly utilized analytical methods in paleomobility studies (Makarewicz and Sealy 2015).

Strontium isotope analysis is based on the following principles (see review in Bentley 2006). Of the four naturally occurring isotopes of strontium, three are stable (^{84}Sr, ^{86}Sr, and ^{88}Sr), and one is radiogenic (^{87}Sr) produced by the decay of ^{87}Rb with a half-life of roughly 4.88×10^{10} years (Faure and Mensing 2005). The amount of ^{87}Sr in rocks and minerals is thus a function of the amount of ^{87}Rb (relative to strontium) in the parent material and time, and the ratio of interest is reported as ^{87}Sr/^{86}Sr. Strontium isotopes display geographic variation based on the age and lithology of underlying bedrock (Faure and Mensing 2005). Due to chemical weathering and leaching, strontium derived from bedrock enters soil and groundwater, is taken up by plants and enters the food web (referred to as biosphere or bioavailable strontium). As strontium is chemically similar to calcium, the former readily replaces the latter in various minerals including hydroxyapatite, the main mineral component of bones and teeth (Bentley 2006). Therefore, in vertebrate animals, including humans, strontium tends to be concentrated in the skeletal system. Whereas bones remodel throughout life, the crowns of teeth are comprised of dental enamel which does not undergo remodelling and thus retains the biochemical and isotopic signatures of the period during which it formed. The enamel of human deciduous (baby) teeth forms and mineralizes *in utero* until infancy, while enamel of permanent teeth forms at different periods per tooth type, ranging from early infancy (first molars) to adolescence (third molars), although most tooth crowns form primarily during childhood (Hillson 1996). Therefore, human teeth preserve a permanent isotopic record, or biogeochemical passport, of the location where they formed as these will not change even after subsequent migration to a new location.

The principles of oxygen isotope analysis of human teeth for paleomobility studies reflect both similarities and differences with the strontium isotope method. Whereas both methods are primarily applied to teeth and can be used to determine childhood origins and thus infer migration, the main difference lies in the sources of isotopic variation. Oxygen in biological or biogenic tissues (including human bones and teeth) is predominantly derived from consumed water (Daux *et al.* 2008; Longinelli 1984; Luz *et al.* 1984; Pederzani and Britton 2019; Podlesak *et al.* 2008). In most, but certainly not all, archaeological contexts, drinking water was obtained from local water sources, which are ultimately derived from precipitation (rain, snow). Oxygen isotopes ($\delta^{18}O$) in precipitation vary globally based on various geographic and climatic variables such as latitude, altitude, distance to coast, and temperature of precipitation (Dansgaard 1964; Gat 1996). Several decades of research have been devoted to understanding spatial and temporal variation in oxygen isotopes as these are one of the most reliable proxies for (paleo)-climate studies (Bowen and Wilkinson 2002). Because strontium and oxygen isotopes vary independently, they represent useful complementary isotope proxies, and when combined can potentially provide more interpretive power than either isotope proxy in isolation.

In paleomobility studies, strontium and/or oxygen isotope analysis is most commonly conducted on dental enamel. Many studies have primarily focused on using isotope data to distinguish between locals (locally born and raised individuals with a local isotope signal) and nonlocals (individuals with a nonlocal signal who were born and raised elsewhere and who subsequently migrated to the site/location where they were interred). One of the main interpretive limitations of this approach is that nonlocal individuals originating from isotopically similar regions are falsely identified as locals (false negatives). Another critical limitation is that isotope signals are never unique to a single location and many regions/locations are isotopically indistinguishable. This issue is known as equifinality (Price *et al.* 2007) and the implication of this is that it is very difficult, if not impossible, to pinpoint a specific origin based on isotope data alone.

Additionally, there has been substantial discussion, and little consensus, concerning how local isotope ranges should be estimated (*e.g.,* Valentine *et al.* 2015; Wright 2005). For strontium isotopes, many early studies (Ericson 1985; Nelson *et al.* 1986; Sealy *et al.* 1991) tended to focus on strontium isotope analysis of both enamel and bone with the assumption that owing to remodelling, bone $^{87}Sr/^{86}Sr$ should eventually equilibrate to the local isotope signal. Subsequent research has demonstrated that buried bone is highly susceptible to diagenetic alteration of biogenic $^{87}Sr/^{86}Sr$ (Budd *et al.* 2000; Hoppe *et al.* 2003; Trickett *et al.* 2003), and that the degree of post-mortem contamination of archaeological bone $^{87}Sr/^{86}Sr$ is highly variable and difficult to assess or remove (Price 1989). As such, strontium isotope analysis of bone as a sampling material has fallen out of favour, with the exception of cremated bone, which appears to retain the *in vivo* isotope signal (Snoeck *et al.* 2018). In the absence of independent data to assess local strontium isotope ranges, statistical assessments of human isotope data have also been used to generate local range estimates (Wright 2005), but these generally rely on dubious assumptions including that the majority of the sample population is of local origin, and that isotopic variation within the local population is random. One of the most commonly used methods to assess local strontium isotope ranges is to measure $^{87}Sr/^{86}Sr$ in locally derived materials, such as soils, water, animals and plants. In general, non-motile archaeological faunal remains are considered the best proxies for local range estimates (Price *et al.* 2002), but there is substantial variation in the targeting of sampling materials and species (Evans *et al.* 2010; Grimstead *et al.* 2017).

3 Isoscapes

Until fairly recently, the vast majority of isotope studies of human paleomobility represent site-based case studies limited to identifying local and nonlocal individuals. While these have been highly successful in this regard and for explorations of human paleomobility patterns more generally, one valid critique (Pestle *et al.* 2013) is that they provide little or no insight into the geographic origins of nonlocals (migrants). Subsequent studies have also made significant progress in our understanding of spatial isotopic variation at larger spatial scales. Much of this research has focused on developing larger regional databases of bioavailable strontium isotope ratios (BASR), based on a wide range of different sample proxies. These are essential for 'pushing the envelope' of isotope migration studies beyond the identification of nonlocals and towards a true provenance approach permitting scientific explorations of the geographic origins of people (or animals, materials, artefacts). It should be kept in mind that isotopic provenance studies are inherently exclusionary, meaning that the isotope data can only be used to exclude, and not pinpoint, possible places of origin. Nonetheless, a good understanding of spatial isotope variation can in many cases permit more nuanced interpretations of human isotope data and can sometimes be used to vastly reduce the possible areas of origin for nonlocal individuals, especially in combination with other lines of evidence.

Furthermore, advances in isotope method and theory in other fields of study have led to further development of these approaches in archaeology. Specifically, isotope studies of modern animal migrations in the field of wildlife ecology led to increased attention to mapping stable isotope variation at greater spatial scales (*e.g.,* Hobson and Wassenaar 2018; Hobson *et al.* 2010). Owing at least in part to the very substantial dedication of resources (time, labour, money) required to collect and analyse appropriate sampling materials over large areas, digital solutions to the problem of large-scale isotope mapping were investigated. This led to the development, approximately a decade ago, of the concept of isoscapes: spatially explicit predictive models of isotope variation (West *et al.* 2009). Regional and global isoscapes for various isotope systems have also been developed. For example, precipitation oxygen isoscapes are based on large datasets of oxygen isotope measurements of water collected at various locations across the globe as part of the Global Network of Isotope in Precipitation (GNIP) project of the IAEA (IAEA/WMO 2016). Using various spatial statistics to interpolate between measured data forms the basis for predictive models (maps) of isotopic variation across the landscape (isoscapes). In this sense, the development and use of large-scale isoscapes for light stable isotopes (*e.g.,* oxygen, hydrogen, carbon, and nitrogen) is further advanced than those for strontium or lead (Pb). In fact, for several years, online platforms such as the Online Isotopes in Precipitation Calculator, and ISOMAP (Isoscape Modelling Analysis and Prediction) have provided invaluable tools for using isotope data for a wide range of research applications across various fields ranging from climate change to ecology to forensics.

However, compared to isoscapes for light stable isotopes, strontium isoscapes have been less widely used despite their enormous potential for provenance and mobility studies in archaeology and beyond. Fortunately, in the last several years strontium isoscapes have also been developed for many regions of the world, and recently even a global bioavailable strontium isoscape was published (Bataille *et al.* 2020). One of the main limitations to the broader application of strontium isoscapes is the fact that most examples have simply been published as figures within articles lacking associated code and as such, they are generally not very user friendly. This trend is expected to change dramatically in the coming years with the increased focus on digital archaeology (quantitative and computational approaches in archaeological research), the publication of associated programming codes for user-friendly applications (*e.g.,* Bataille *et al.* 2018), the increasing availability of large digital repositories of isotope data (IsoArch, https://isoarch.eu/), and the development of online tools for inputting and interpreting archaeological strontium isotope data such as the British Isotope Domains website of the British Geological Survey. One persistent limitation concerning the use of isoscapes in general, and strontium isoscapes in particular, in paleomobility and provenance research, is that there has been a general lack of validation and testing of their accuracy and efficacy.

3.1 Circum-Caribbean isoscapes

One of the main aims of the current study is to further assess the reliability of isoscapes for the determination of individual geographic origins based on enamel strontium and oxygen isotope data. In this regard, this study represents a continuation of a previous validation study with similar aims using Circum-Caribbean strontium and oxygen isoscapes (figure 1). The original Circum-Caribbean strontium isoscape represents a three-source mixing model (which incorporates strontium inputs from bedrock, sea spray, and atmospheric dust) originally developed by Bataille *et al.* (2012) and was tested on several hundred baseline strontium isotope measurements from Mesoamerica (Hodell *et al.* 2004) and the insular Caribbean (Laffoon 2012; Laffoon *et al.* 2012). The multi-source Circum-Caribbean strontium isoscape performs better than a traditional single source model (bedrock only) in predicting the observed baseline $^{87}Sr/^{86}Sr$ ratios in both regional datasets: Mean Absolute Error (MAE) = 0.00040, Root Mean Square

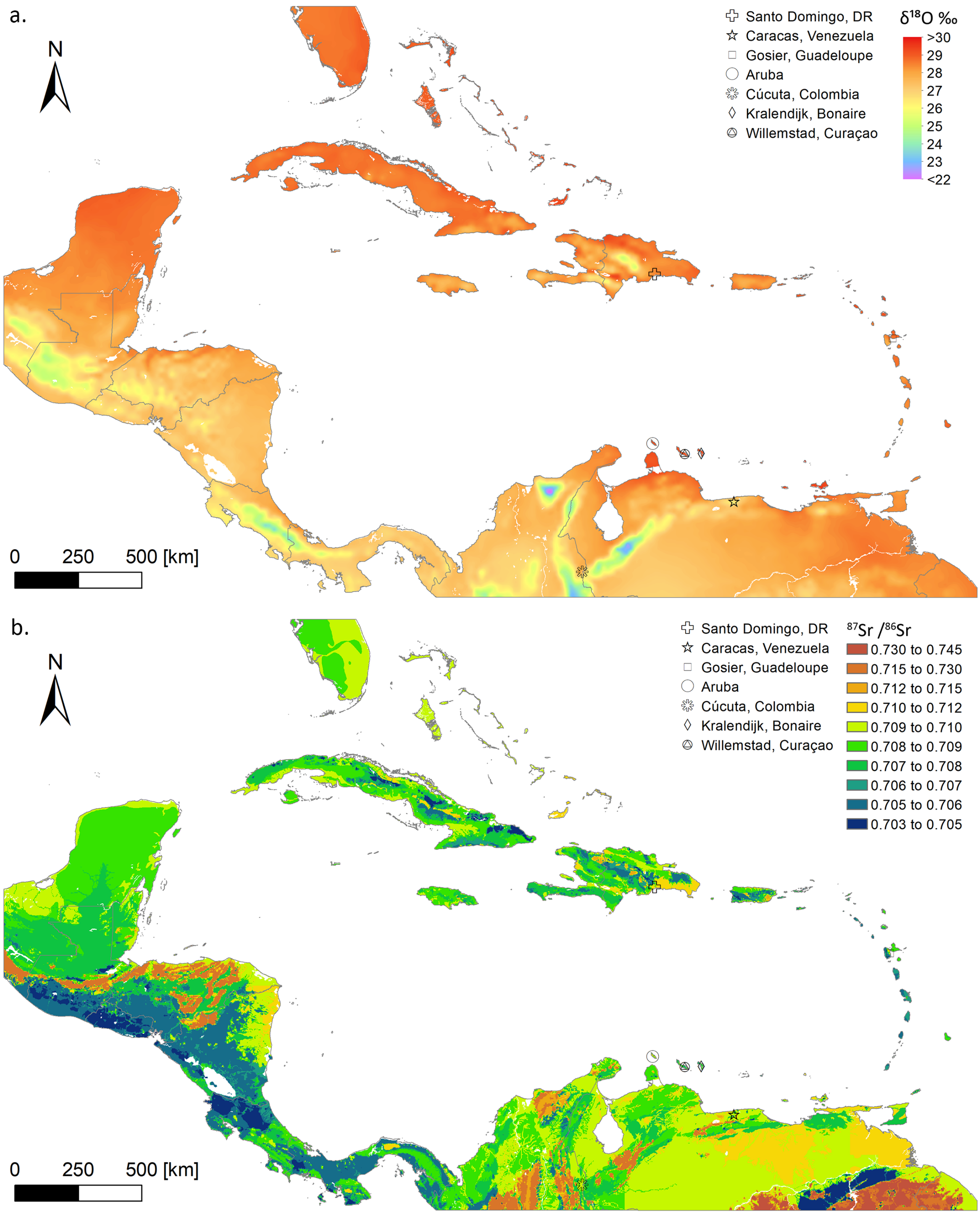

Figure 1: (a.) Enamel oxygen ($\delta^{18}O$) and (b.) bioavailable strontium ($^{87}Sr/^{86}Sr$) isoscapes of the Circum-Caribbean.

Error (RMSE) = 0.00087; and MAE = 0.00014, RMSE = 0.0010, respectively. Furthermore, the multi-source model is in good agreement with the range of (local) human $^{87}Sr/^{86}Sr$ ratios for dozens of sites from multiple islands and locations within the broader Caribbean (Laffoon 2012) as well as subsequent baseline measurements from previously unsampled or under-sampled regions. The enamel $^{87}Sr/^{86}Sr$ isoscape used in this paper is modified from the bioavailable multi-source mixing model of Bataille *et al.* (2012).

Within the insular Caribbean there is a spatial bias of representation in the original baseline bioavailable $^{87}Sr/^{86}Sr$ dataset (Laffoon *et al.* 2012) with much of the eastern and southern Caribbean well covered, while much of the northern (Bahamian archipelago) and western Caribbean (Haiti/Dominican Republic, Jamaica, Cuba) is represented by few data points relative to their land mass. A similar problem of spatial representation also plagues the bioavailable data from the mainland regions of the Circum-Caribbean with Mesoamerica very well represented, whereas much of Central America and Northern South America are represented by very few sampling locations or data points. Fortunately, further research has made substantial progress filling in some of the gaps in bioavailable $^{87}Sr/^{86}Sr$ data for many areas within the Circum-Caribbean: Puerto Rico (Pestle *et al.* 2013), Nevis and Carriacou (Giovas *et al.* 2016), Trinidad and Tobago (Ostapkowicz *et al.* 2017), Venezuela (Laffoon *et al.* 2018), the Bahamas and Turks and Caicos (Schulting *et al.* 2018), Barbados (Giovas *et al.* 2019), the Dominican Republic (Laffoon *et al.* 2019), Jamaica (Mickleburgh *et al.* 2019), Panama (Sharpe *et al.* 2021) and Cuba (Laffoon and Chinique de Armas 2022). Additionally, strontium isotope mapping on the Bahamian archipelago has demonstrated little or no influence of Sr inputs from atmospheric dust on bioavailable $^{87}Sr/^{86}Sr$ (Schulting *et al.* 2018). This insight suggests that a simpler two-source mixing model (including bedrock and sea spray) may adequately reflect the main sources of strontium to terrestrial Antillean ecosystems, although further research is required to assess if this is more broadly applicable across the entire Circum-Caribbean.

The Circum-Caribbean oxygen isoscape used herein is ultimately based on measurements of oxygen isotopes in precipitation from the GNIP (IAEA/WMO 2013) and the regionalized cluster-based water isotope prediction (RCWIP) model of Terzer *et al.* (2013). Details of the data conversion, reduction, and manipulation steps used to produce the oxygen isoscape are reported in Laffoon *et al.* (2017). The precipitation oxygen isoscape was converted to an enamel isoscape by converting the water $\delta^{18}O$ values to enamel $\delta^{18}O$ values based on the equation of Chenery *et al.* (2012). This enamel oxygen isoscape is generally consistent with measured human and animal enamel $\delta^{18}O$ values from a wide range of sites and islands throughout the Antilles (Laffoon *et al.* 2013). Isotope analyses of bioarchaeological samples in the insular Caribbean combined with strontium and/or oxygen isoscapes have already contributed to investigations of human paleomobility patterns, animal migrations and the provenance of artefacts made from faunal skeletal remains (Laffoon *et al.* 2014; 2017; 2018).

4 Materials and methods

The validation tests were applied to multiple modern individuals of known (childhood) origins from various locations within the Circum-Caribbean: Santo Domingo, Dominican Republic; Caracas, Venezuela; Gosier, Guadeloupe; unspecified location in Aruba; Cúcuta, Columbia; Kralendijk, Bonaire; Willemstad, Curaçao. In total, isotope data from seven individuals from seven different locations were assessed (table 1). Five of these derive from previous studies by Plomp *et al.* (2019; 2020), four of which include only strontium isotope data, and one with both strontium and oxygen Sr; one individual was previously reported (Laffoon *et al.* 2017) and is re-assessed here; and one individual is from a new analysis for the current study. Dental samples were donated (anonymously) from living individuals who self-reported their geographic location (as well as diet, habits) at the time of tooth formation and mineralization via a questionnaire, and permission for analyses was approved by the Medical Ethics Review Committee of the VU University Medical Center (Plomp *et al.* 2019). The sampled dental elements represent two deciduous first molars (dm1) and five permanent third molars (M3). Deciduous first molars primarily mineralize *in utero* (AlQahtani *et al.* 2010) and thus their enamel $\delta^{18}O$ should not be influenced by the breastfeeding effect, which can elevate $\delta^{18}O$ values (Lin *et al.* 2003). We furthermore sampled the enamel near the cusps, which tends to form earlier than enamel near the tooth cervix in order to avoid material formed after birth. Third molar enamel crowns form from late childhood through adolescence (circa 8-14 years) and thus are not impacted by breastfeeding effects (AlQahtani *et al.* 2010).

Sample processing and analyses were similar for all individuals and are reported in detail elsewhere

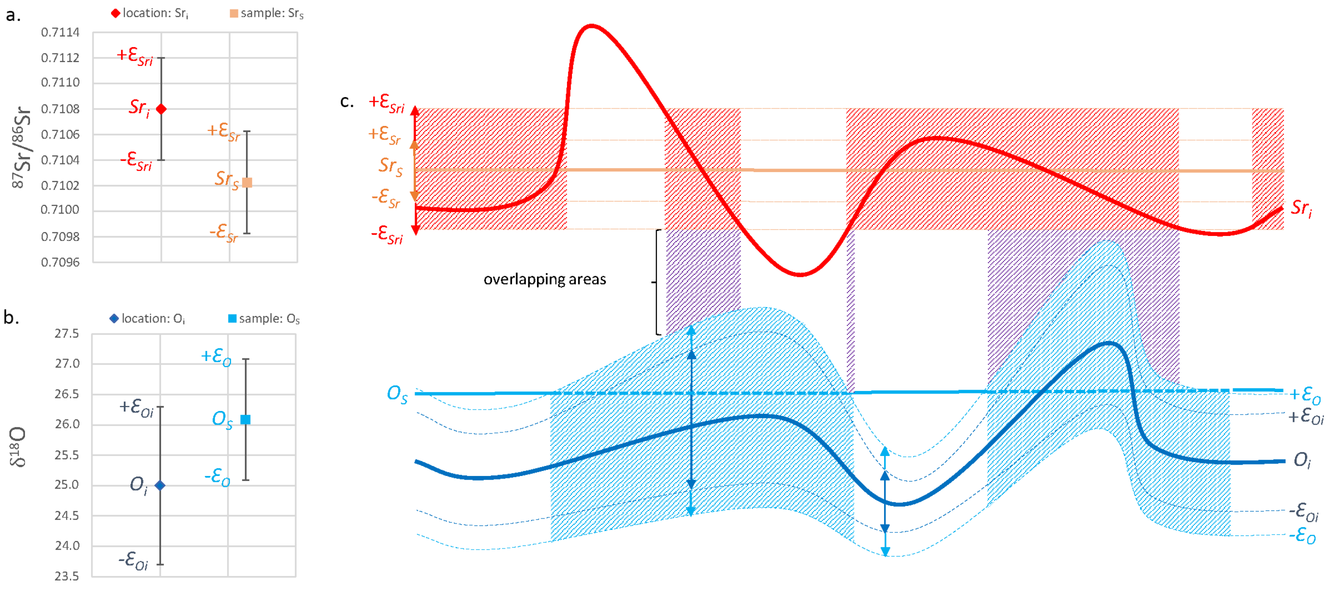

Figure 2: Sources of (a.) strontium and (b) oxygen uncertainties from measurements and raster map creation. Final areas from (c.) overlapping isotope uncertainty ranges.

(Laffoon 2012; Plomp *et al.* 2020). Briefly, dental samples were mechanically cleaned and then enamel was extracted using a hand-held dental drill. The samples were subsequently split in two for strontium and oxygen isotope analyses. For strontium isotope measurements, samples were dissolved in nitric acid, and then strontium was separated from the sample matrix using column chromatography with Sr-specific resin. Strontium isotope ratios were measured on a Thermo-Scientific TritonPlus Multi-Collector TIMS. Total procedural blanks were negligible, and instrument performance was monitored via the analyses of the international strontium standard (NBS-987) with a long-term mean $^{87}Sr/^{86}Sr$ of 0.710249 ±0.0001 (1σ). Oxygen isotopes were measured on a Thermo-Finnigan DeltaPlus IRMS coupled to a Gasbench II interface. Oxygen isotope results are normalized using an internal laboratory standard (VICS), with a long-term reproducibility of the international reference material (NBS-19) for $\delta^{18}O$ of <0.2‰. Enamel oxygen isotope results are reported in the δ notation, in parts per thousand (‰) relative to the international VPDB standard and converted to VSMOW using the equation of Coplen (1988).

In terms of assessing geographic origins, we use the Interval Approach previously outlined in Laffoon *et al.* (2017). The Interval Approach is a simple way of assigning a yes (possible origin) or no (not possible origin) to each raster cell (location) on a map (isoscape) by directly comparing the range of possible isotope values for each raster cell on the map with the measured isotope value(s), and associated individual error ranges/uncertainties, of the sample for strontium, oxygen, or both.

Both isotope databases had previously been uploaded into a GIS environment, where the globally estimated RCWIP oxygen data point values of approximately 18 km x 18 km distance were adjusted to match the strontium raster data set resolution of one km^2 using cubic spline interpolation. Both images were subsequently framed by a Caribbean boundary map (open source by http://naturalearthdata.org), creating two exactly overlapping maps (Laffoon *et al.* 2017), displaying only land coverage. These datasets form the base to calculate the interval approach results. The uncertainty ranges of each isotope value, as minimum and maximum extent, are compared with the isotope values by a logical operator. If the map values lie within the defined range, the logical operator assigns this pixel value one; if the value is outside the range, the pixel value is zero. The extent of this min-max range based on the combined uncertainties fills the interval of matching values, which defines the size of the remaining areas.

Regarding strontium, the combined errors of map variation (ε_{Sri}) and measurement uncertainty (ε_{Sr}) result

Sample ID	Location	Country	element	source	$^{87}Sr/^{86}Sr$	$^{87}Sr/^{86}Sr$	$\delta^{18}Oc$	$\delta^{18}Oc$	$\delta^{18}Oc$
							‰ VPDB	‰ VPDB	‰ VSMOW
					measured	*2SE*	*measured*	*1σ*	*Coplen 1988*
V464_V1	Caracas	Venezuela	dm	*1*	0.71013	0.00001	-4.9	0.1	25.8
DR_WL	Santo Domingo	Dom. Rep.	M3	*2*	0.70831	0.00001	-4.3	0.1	26.5
V463_G2	Le Gosier	Guadeloupe	dm	*3*	0.70820	0.00001	-4.5	0.1	26.2
W4-B16	Cúcuta	Columbia	M3	*2*	0.71175	0.00001			
W6-B13	?	A/B/C	M3	*2*	0.70950	0.00001			
W2-R8	Willemstad	Curaçao	M3	*2*	0.70945	0.00001			
W3-B4	Kralendijk	Bonaire	M3	*2*	0.70926	0.00001			

Table 1: Isotope values and relevant sampling information of known origin individuals from the Circum-Caribbean area. Data sources: 1) Laffoon *et al*. 2017; 2) Plomp *et al*. 2019; 3) this study.

in a total error of ±0.0008, which defines the range added or subtracted as a general mean error. Regarding oxygen, a novel path was chosen: firstly, the population variance is calculated by taking the square of the standard deviation of all measured values, with mean value at 0.687132‰, which was then rounded up to the significant digit ~ 0.7‰. To this value, a measurement uncertainty of 0.1 (reflecting the typical uncertainty of oxygen isotope measurements) was added, resulting in 0.8‰ measurement uncertainty (ε_O). An additional source of error was defined by the individual uncertainty for each data point from the RCWIP database (ε_{Oi}) which was transferred and interpolated to each individual pixel accordingly, ranging within the map between 0.9 and 1.4‰. To this floating error range, the measurement uncertainty of ±0.8‰ was added respectively. Based on the rules of propagating error, the square root of the sum of the squares of each source of uncertainty formed the total error for each pixel. This approach produces a greater uncertainty range for the oxygen results than previously calculated (Laffoon *et al.* 2017), producing a larger potential region of origin, but reducing the possibility of false positive results.

For samples where both strontium and oxygen values exist, these resulting maps once again were compared by a logical operator, leaving out all areas where either oxygen or strontium had turned out zero (figure 2). Figure 2 is a graphical representation of how the interval approach defines overlapping areas by cancelling out the rest, using a range of hypothetical values, and associated errors, for both strontium and oxygen (y-axes) and hypothetical variation of both over geographic location (x-axis).

5 Results

The strontium and oxygen isotope measurements and relevant sample information are listed in table 1. The results of the Interval Approach are presented in figure 3 (using both strontium and oxygen isoscapes) and figure 4 (using only strontium isoscapes). We assess the effectiveness of the Interval Approach in terms of both accuracy and precision. In this context, accuracy refers to whether or not the Interval Approach correctly identifies the known geographic origin of the individual as a potential place of origin, whereas precision refers to the extent that the Interval Approach constrains the number of potential places of origin within the overall map. In this section we present the results on a case-by-case basis starting with the three samples for which both strontium and oxygen isotopes were measured.

Individual V1 was previously analysed and discussed in the original proof of concept study (Laffoon *et al*. 2017). We reanalysed it herein using a slightly modified version of the Interval Approach, namely with an expanded range of uncertainty for the oxygen isotope measurements (see paragraph 6.4). Ultimately, the results of this re-analysis with Interval Approach are very consistent with those of the original study. More specifically, separately both the strontium and oxygen isoscapes greatly constrain the potential places of origin for this individual but when the two isoscapes are combined using the Interval Approach the potential places of origin are even more constrained (figure 3). In this case the method is both accurate in the sense that Caracas (the known origin for this individual) is correctly identified as a potential

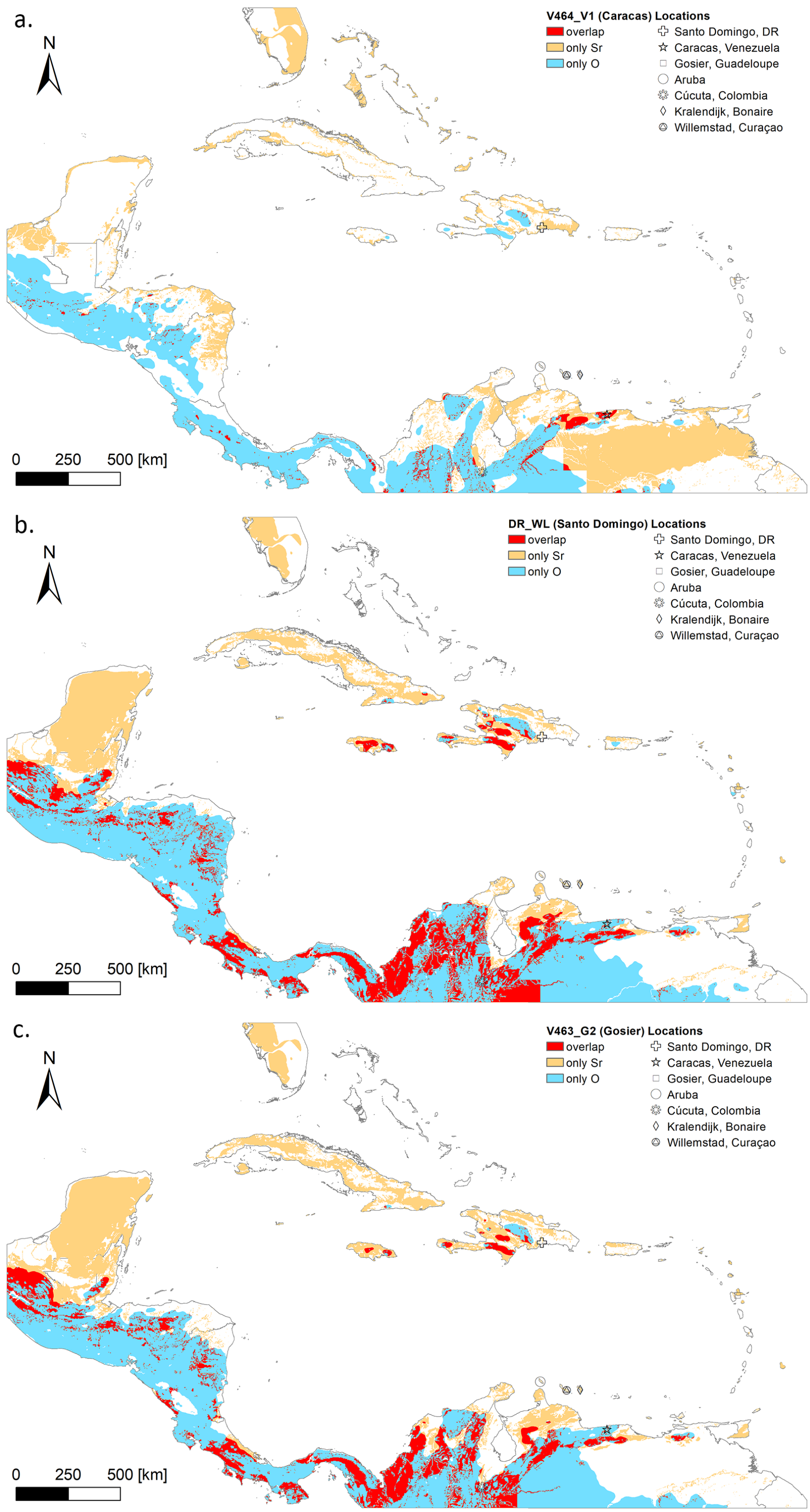

Figure 3: Results of the Interval Approach applied to strontium and oxygen isotope data and corresponding isoscapes.

place of origin, and precise in that the vast majority of the map area (>90% of the total raster cells) is excluded as a place of origin.

For Individual WL, the model results were both less accurate and less precise than for Individual V1. Santo Domingo, the known location of origin, was not identified as a potential place of origin. However, a few small areas, several kilometres to the west of the city were corresponding positively (figure 3). Similar to the case for Individual V1, the combination of the strontium and oxygen isoscapes reduced the number of potential places of origin substantially compared to the results of either one individually. In terms of precision, the number of potential places of origin was greatly constrained but less so than the case for Individual V1. More specifically, on the mainland, the model identified many scattered potential places of origin throughout Central America, and north-western South America. Within the insular Caribbean, most islands were excluded but potential places of origin were identified in much of Jamaica, large portions of southwestern Hispaniola (Haiti and the Dominican Republic), and few tiny pockets in south-eastern Cuba.

As expected, the overall model results for Individual G2 are similar to those of Individual WL since they possess very similar isotope values for both strontium and oxygen (figure 3). However, the model results for Individual G2 are far less accurate than for Individual WL. Interestingly, the eastern half of Guadeloupe (Grande-Terre) is identified as a potential place of origin based on strontium but not on oxygen isotopes. When combined, in the Interval Approach, not only is the known origin (in Le Gosier, Grande-Terre, Guadeloupe) excluded as a potential place of origin but so is the whole island of Guadeloupe, as well as the entire eastern Caribbean (Lesser Antilles archipelago). In this case, the strontium isoscape is much more accurate than the oxygen isoscape. For Individual G2 the precision is similar to Individual WL when both isoscapes are combined with large but dispersed potential places of origin throughout Central America, large portions of north-western and northern Southern America, and relatively few and small areas of Jamaica, south-western Hispaniola, and a tiny area of south-eastern Cuba identified.

For the remaining four individuals only strontium isotope data are available. For individual B16 with known origins in Cúcuta, Columbia, the model results did not accurately identify this location as a potential place of origin. However, numerous other locations in Columbia, including several relatively constrained pockets in the highlands, including areas in the east somewhat near Cúcuta were identified as potential places of origin (figure 4). The model results for this individual were the most precise in the sense that the greatest area within the map was excluded as a potential place of origin relative to the other samples in this study. Interestingly, the model results primarily only identify relatively small and isolated locations in inland and highland areas of the Circum-Caribbean as possible places of origin for this individual. This pattern probably reflects the fact that individual B16 has a fairly high $^{87}Sr/^{86}Sr$ ratio (0.71175) that is more characteristic of older, continental geological settings primarily underlain by metamorphic bedrock. In addition to possible locations in Columbia, the model results also identify potential origins in very restricted areas of Venezuela, Panama, Nicaragua, Honduras, Cuba and Hispaniola. The latter two seem unlikely on the basis of empirical baseline $^{87}Sr/^{86}Sr$ data for the insular Caribbean where such high ratios have only been reported for Trinidad and Tobago (Laffoon *et al.* 2012; Ostapkowicz *et al.* 2017).

The remaining three samples for which only strontium isotope data are available have known origins in the so-called ABC islands (Aruba, Bonaire, Curaçao) located in the southern Caribbean just north of the coast of central Venezuela. For individual B13 it is only known that the origin is from one of the three ABC islands, whereas individual R8 has known origins from Willemstad, Curaçao and individual B4 originates from Kralendijk, Bonaire. All three individuals have very similar $^{87}Sr/^{86}Sr$ ratios and hence the model outputs for all three are highly comparable (figure 4). For these three individuals the model outputs are neither particularly accurate nor precise. All three model outputs correctly identify at least two of the ABC islands as a potential place of origin. For R8 and B4 all islands are identified as potential locations, while for B13 only Aruba and Curaçao are within the range. Some neighbouring areas of northern coastal Venezuela are also identified as such. The model outputs are less precise than the other samples in this study with a fairly reduced area of the map excluded as potential places of origin, and by consequence, a fairly large area of the map identified as possible places of origin for all three individuals.

6 Discussion and conclusions

The relatively high degree of spatial variation in baseline isotope values in the Circum-Caribbean (figure 1), particularly for strontium, offers substantial

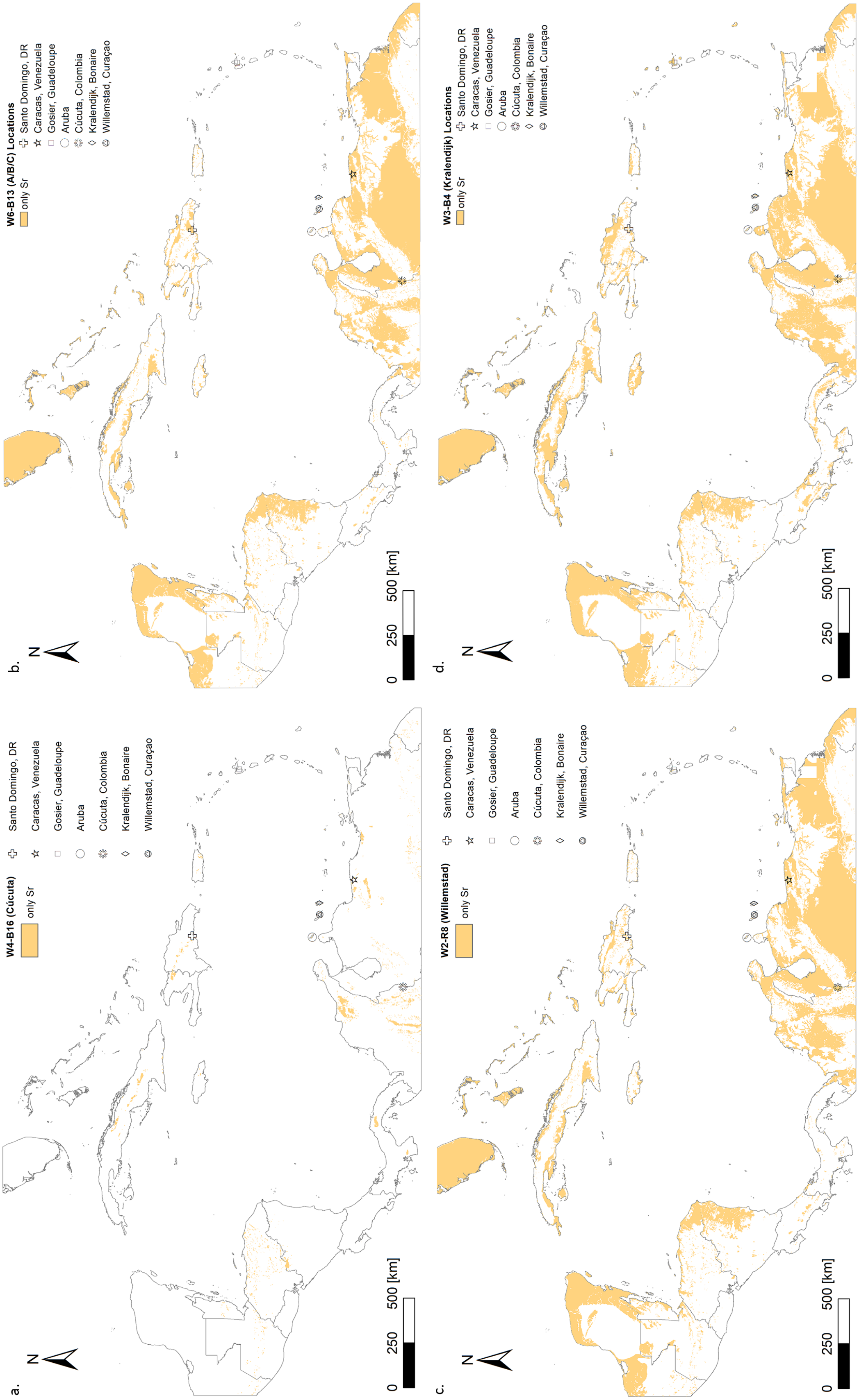

Figure 4: Results of the Interval Approach applied to strontium isotope data and corresponding isoscape.

potential for the application of isotopes and isoscapes to investigations of the geographic origins of a wide range of biogenic materials, including examinations of the childhood origins of individual humans from archaeological and forensic contexts. The application of the Interval Approach outlined herein to modern individuals with known geographic origins permits further assessment of the validity of this method. The model results of the Interval Approach for seven individuals detailed above raise numerous discussion points.

A previous study (Laffoon *et al*. 2017) using the same isoscapes and similar methods (*i.e.*, dual isoscapes and the Interval Approach) was applied to an individual from Caracas, Venezuela (V1, figure 3a). The results of this previous study (Laffoon *et al*. 2017) indicated a high degree of accuracy and precision when both strontium and oxygen isotopes were combined. This analysis was replicated herein with a slightly modified method but with identical results. More specifically, the origin identified by the model could be pinpointed to the area around Caracas and the nearby Venezuelan coastal range and thus was highly consistent with the known origin for this individual in Caracas. This example represents the best-case scenario for the overall method and approach. Unfortunately, the results for the other six individuals in this study were much more variable and less promising.

For the other two individuals for whom both strontium and oxygen isotope data were available (WL from Santo Domingo, Dominican Republic; and G2 from Gosier, Guadeloupe) the model results were somewhat less precise than for individual V1 but large areas of the Circum-Caribbean map (>50%) were excluded as potential places of origin. Since the isotope method for assessing origins is essentially exclusionary, the approach could be considered partially successful in the sense that many possible areas of origin have been excluded. However, much more concerning is the fact that the model outputs for both individuals were inaccurate (*i.e*., did not correctly identify the known origin as a potential place of origin). One difference between these two cases is that for individual WL, possible places of origin were identified fairly close (within ~10 km) to the actual location of origin in Santo Domingo. Whereas, for individual G2 the strontium isoscape correctly identified the eastern half of Guadeloupe as a potential place of origin but this was excluded on the basis of the oxygen isoscape. We consider these two examples as being partially successful in the sense that while both are moderately precise, the former is only slightly inaccurate while the latter is accurate based on one isotope system but not the other.

Highly variable results were also obtained from the four samples for which only strontium isotope data was available. For Individual B16 (from Cúcuta, Columbia), the results were very precise (approaching the precision obtained for Individual V1) with >90% of the map area excluded as a potential origin. The result was not very accurate given that the known origin was not identified as a potential place of origin, however, similar to the case for Individual WL where other, not too distant (<50 km) areas of eastern Columbia were identified as such. The results for the last three individuals were the least promising in terms of both accuracy and precision: while the model results correctly identified areas within the known islands of origin, extremely large areas of the overall map remained as possible places of origin.

In general, the model results obtained from the Interval Approach were highly variable and, in most cases, not particularly promising. We highlight several issues that need to be addressed to further develop these methods and hopefully improve the accuracy and precision of the results. First, one possible concern may be the errors associated with the isotope measurements themselves. This, however, seems unlikely to be a major concern given that the measurement uncertainties are much smaller (in some cases by an order of magnitude) than: the normal variation in isotopes within a single (local) individual (Plomp *et al*. 2020), the variance within most (archaeological) populations measured to date, and the uncertainties associated with the isoscape models themselves (Laffoon *et al*. 2017).

A second potentially confounding factor is the selection of the appropriate samples for isotope analysis. In this study, five teeth are third molars, which form during adolescence and thus are not impacted by breastfeeding effects. Two others are represented by deciduous first molars, which primarily form *in utero*. However, mineralization of deciduous first molars continues after birth, and it is possible that some of the sampled enamel from these teeth may reflect the breastfeeding period or a combination of enamel formed before and after birth, and therefore contain a mix of fetal and breastfeeding inputs. Other issues pertaining to the use of oxygen isotopes for provenance studies more generally include seasonal variation in oxygen isotope values and long-term climatic change (Pederzani and Britton 2019), the latter of course being more relevant for archaeological than for modern cases. There are also numerous processes that may cause significant differences between measured $\delta^{18}O$ values of local precipitation (upon which the oxygen isoscapes are based) and the $\delta^{18}O$ of consumed water (which is the primary source of $\delta^{18}O$ in biogenic tissues) such as fractionation from water storage, the consumption of imported (bottled) water, and cooking practices such as boiling and brewing (Brettell *et al.* 2012; Lee-Thorp 2008; Pederzani and Britton 2019), in addition to the previously discussed uncertainties associated with the conversions between water and enamel isotope values.

In terms of the reliability of the isoscapes, as previously mentioned for the Circum-Caribbean there is a distinct spatial bias in the representativeness of sampling for baseline strontium isotope ratios, with much of the northern and western insular Caribbean, and vast areas of Central America and northern South America grossly underrepresented relative to their size. A recent study that developed and tested the first global-scale strontium isoscape found that predictive models underperformed in areas that lacked spatially representative bioavailable $^{87}Sr/^{86}Sr$ data (Bataille *et al.* 2020). A related issue concerns the geospatial models that are used to predict bioavailable $^{87}Sr/^{86}Sr$ at large spatial scales. In the Caribbean, recent studies have demonstrated a notable lack of input from atmospheric sources (*e.g.*, Saharan dust) on bioavailable $^{87}Sr/^{86}Sr$ for certain islands and archipelagos (Schulting *et al.* 2018) that contrast with the process-based models used to generate the Circum-Caribbean strontium isoscape used in this study (Bataille *et al.* 2012). Furthermore, newer more robust geospatial models utilizing machine-learning approaches (Bataille *et al.* 2018) have been recently developed and offer potential for improving existing isoscapes at multiple scales, including for the Circum-Caribbean. Although not relevant for the current validation test, it should also be noted that the spatial limits of the Circum-Caribbean isoscape place *a priori* constraints on the Interval Approach. In other words, the approach will only identify possible places of origin within the geographical boundaries of the isoscape model.

Perhaps the most problematic issue limiting the utility of the isotope/isoscape method for provenance studies of modern individuals is the globalization of the food industry. For example, the three individuals in the present study from the ABC islands all have $^{87}Sr/^{86}Sr$ ratios that exceed the measured range of bioavailable $^{87}Sr/^{86}Sr$ for these islands (maximum = 0.70915), a value that is very close to the mean $^{87}Sr/^{86}Sr$ ratio of seawater (0.70918). It is possible that the limited sampling (n=20 samples in total) of biosphere $^{87}Sr/^{86}Sr$ has not been sufficient to reflect the overall variance of $^{87}Sr/^{86}Sr$ for these three islands. This seems like an unlikely explanation in this case, however, as most island and coastal settings can be characterized as having bioavailable $^{87}Sr/^{86}Sr$ reflecting a two end-member mixing model between bedrock and marine-derived strontium. In the case of the ABC islands, the bedrock values represent the low endmember of this mixing solution and marine strontium (from sea spray and/or precipitation) represents the high endmember. As such, terrestrial bioavailable $^{87}Sr/^{86}Sr$ is not expected to exceed that of seawater (~0.7092) for these islands, or most other islands of the Caribbean for that matter. In this context, it is more parsimonious that the high $^{87}Sr/^{86}Sr$ ratios of the three measured individuals from the ABC islands reflect the consumption of imported foods.

It is difficult to assess the impact of imported foods on $^{87}Sr/^{86}Sr$ ratios in human dental enamel and the effects may be highly variable both spatially and temporally. Previous studies using isotope analyses of strontium and oxygen of World War 2 casualties successfully identified the geographic origins of at least one individual, which was later confirmed by other lines of evidence (Font *et al.* 2015a). Another recent study was successful at precisely and accurately constraining the origin of historically documented individuals from early modern Brittany (Bataille *et al.* 2021; Colleter *et al.* 2021). There are also numerous forensic studies that have utilized various isotope measurements and geographic assignment models to investigate individual (modern) human origins with varying degrees of success (Bartelink *et al.* 2020; Chesson and Berg 2021; Chesson *et al.* 2018; Font *et al.* 2015b; Kamenov and Curtis 2017; Kramer *et al.* 2020; Lehn *et al.* 2015; Mehl *et al.* 2019; Meier-Augenstein and Fraser 2008). Other studies have attempted to assess the reliability of isoscapes, or baseline isotope datasets, against isotope data obtained from modern individuals but the results have been quite variable with some areas demonstrating congruencies and other areas major differences depending on location and the type of material sampled for isotope analyses (*e.g.*, Ammer *et al.* 2020; Kootker *et al.* 2020; Ueda and Bell 2021). These studies seem to suggest that the complications introduced by the globalized nature of the food economy are perhaps not insurmountable but may generally increase over time and with variable impacts depending on the extent that people consume imported foodstuffs. The degree that people consume nonlocal foods (with isotope ratios that do not reflect local sources) may be further exacerbated in small countries or islands where a large proportion of food is imported, as is the case in much of the insular Caribbean nowadays. By contrast, in regions where most if not all consumed food and water is of local origin the globalized food industry may have little effect on biogenic isotope values or the reliability of the isotopic approach to provenance studies.

In summary, the results of this study are highly variable in terms of both precision and accuracy. In this regard, they demonstrate a more complicated and nuanced situation than the results of the original validation study (Laffoon *et al*. 2017) of the Circum-Caribbean isoscapes which was based on a single sample and was, perhaps purely by coincidence, the most accurate and precise of the seven samples included in the present expanded study. The general lack of precision for many of the cases presented herein is probably best considered an inherent limitation of the method and not particularly problematic from an interpretive perspective. In many regions of the world, including the Circum-Caribbean, large and/or numerous areas have similar or even identical strontium and/or oxygen isotope values and some values or ranges of values are often much more common than others. If the measured isotope values in any given sample happen to be common in the study area, the isoscape provenance method will be limited in its capacity to constrain the potential places of origin. On the other hand, if a given sample happens to possess an isotope value, for one or more isotope systems, that is less common or particularly rare in a given study area, the degree that the origins can be constrained (the precision of the method) can be greatly improved. The lack of accuracy is a much more serious and confounding problem in this context. Perhaps counter-intuitively, the isotopic provenance method may actually work better for archaeological cases than for modern ones owing to the aforementioned complications of the globalized food industry, whereas attempts to validate the overall method and the isoscapes upon which they are based, generally require the use of modern individuals (with known origins).

Several avenues for future research to address these concerns can be proposed. As the method and approach works similarly for humans as for other terrestrial animals, validating the approach using isotope values from modern fauna may represent an ideal sampling approach that avoids the complications of the consumption of imported food, if samples are taken from wild or captive animals with known dietary histories. Another possibility would be to focus on (archaeological) individuals of known origins who pre-date the recent rapid expansion of the global food economy, for example from pre-20th century or other historical contexts. In the context of the broader aims of this volume (promises and impasses in digital archaeology), we conclude that the combined isotope/isoscape method still holds much potential for provenance investigations more generally, and for constraining the natal origins of individuals in archaeological research more specifically. The current study, however, clearly indicates that caution is merited when applying isotopes and isoscapes to assess geographic origins in modern and forensic cases, at least within the study area of the Circum-Caribbean. The consumption of large quantities of non-local foods violates one of the main underlying principles of the isotope method, namely that isotope values of biogenic tissues will reflect the local environment. We suggest that more research is required, particularly systematic validation tests of multiple isotopes systems and multiple isoscapes, within the Caribbean and beyond, in order to assess the reliability of these methods for investigations of geographic origins before they can be more widely and effectively applied in archaeological and forensic research.

Acknowledgements

We would like to thank the editors, Karsten Lambers and Tuna Kalaycı, for inviting us to contribute to this volume. We are also grateful to all of the individuals who donated their dental samples, and to Suzan Verdegaal-Warmerdam of the Vrije Universiteit Amsterdam for measuring the oxygen isotope ratios. This manuscript was substantively improved by the constructive comments of the editors and two anonymous reviewers.

References

Adams, S., R. Grün, D. McGahan, J. Zhao, Y. Feng, A. Nguyen, M. Willmes, M. Quaresimin, B. Lobsey, M. Collard, and M. C. Westaway. 2019. "A strontium isoscape of north-east Australia for human provenance and repatriation." *Geoarchaeology* 34, 3: 231-251. DOI:10.1002/gea.21728.

AlQahtani, S.J., M.P. Hector, and H.M. Liversidge. 2010. "Brief communication: The London atlas of human tooth development and eruption." *American Journal of physical anthropology* 142, 3: 481-490. DOI:DOI:10.1002/ajpa.21258.

Ammer, S.T.M, L.M. Kootker, E.J. Bartelink, B.E. Anderson, E. Cunha, and G.R. Davies. 2020. "Comparison of strontium isotope ratios in Mexican human hair and tap water as provenance indicators." *Forensic Science International* 314: 110422. DOI:10.1016/j.forsciint.2020.110422.

Bartelink, E.J., L.A. Chesson, B.J. Tipple, S. Hall, and R. T. Kramer. 2020. "Multi-isotope approaches for region-of-origin predictions of undocumented border crossers from the US – Mexico border: Biocultural perspectives on diet and travel history." *Forensic Science and Humanitarian Action: Interacting with the Dead and the Living*: 369-384.

Bataille, C.P., and G.J. Bowen. 2012. "Mapping $^{87}Sr/^{86}Sr$ variations in bedrock and water for large scale provenance studies." *Chemical Geology* 304: 39-52. DOI:10.1016/j.chemgeo.2012.01.028.

Bataille, C.P., B.E. Crowley, M.J. Wooller, and G.J. Bowen. 2020. "Advances in global bioavailable strontium isoscapes." *Palaeogeography, Palaeoclimatology, Palaeoecology* 555: 109849. DOI:10.1016/j.palaeo.2020.109849.

Bataille, C.P., I.C.C von Holstein, J.E. Laffoon, M. Willmes, X. Liu, and G.R. Davies. 2018. "A bioavailable strontium isoscape for Western Europe: A machine learning approach." *PloS one* 13, 5: e0197386. DOI:10.1371/journal.pone.0197386.

Bataille, C.P., K. Jaouen, S. Milano, M. Trost, S. Steinbrenner, É. Crubézy, and R. Colleter. 2021. "Triple sulfur-oxygen-strontium isotopes probabilistic geographic assignment of archaeological remains using a novel sulfur isoscape of western Europe." *PLoS One* 16, 5: e0250383. DOI:10.1371/journal.pone.0250383.

Bataille, C.P., J.Laffoon, and G.J. Bowen. 2012. "Mapping multiple source effects on the strontium isotopic signatures of ecosystems from the circum-Caribbean region." *Ecosphere* 3, 12: 1-24. DOI:10.1890/ES12-00155.1.

Bentley, R.A. 2006. "Strontium isotopes from the earth to the archaeological skeleton: a review." *Journal of archaeological method and theory* 13, 2006: 135-187. DOI:10.1007/s10816-006-9009-x.

Bowen, G.J. 2010. "Isoscapes: spatial pattern in isotopic biogeochemistry." *Annual review of earth and planetary sciences* 38: 161-187. DOI:10.1146/annurev-earth-040809-152429.

Bowen, G.J., and B. Wilkinson. 2002. "Spatial distribution of $\delta^{18}O$ in meteoric precipitation." *Geology* 30, no. 4: 315-318. DOI:10.1130/0091-7613(2002)030%3C0315:SDOOIM%3E2.0.CO;2.

Brettell, R., J. Montgomery, and J. Evans. 2012. "Brewing and stewing: the effect of culturally mediated behaviour on the oxygen isotope composition of ingested fluids and the implications for human provenance studies." *Journal of Analytical Atomic Spectrometry* 27, no. 5: 778-785. DOI:10.1039/c2ja10335d.

Britton, K., M. Le Corre, M. Willmes, I. Moffat, R. Grün, M.A. Mannino, S. Woodward, and K. Jaouen. 2020. "Sampling plants and malacofauna in $^{87}Sr/^{86}Sr$ bioavailability studies: Implications for isoscape mapping and reconstructing of past mobility patterns." *Frontiers in Ecology and Evolution* 8: 579473. DOI:10.3389/fevo.2020.579473.

Budd, P., J. Montgomery, B. Barreiro, and R.G. Thomas. 2000. "Differential diagenesis of strontium in archaeological human dental tissues." *Applied geochemistry* 15, 5: 687-694. DOI:10.1016/S0883-2927(99)00069-4.

Chenery, C.A., V. Pashley, A.L. Lamb, H.J. Sloane, and J.A. Evans. 2012. "The oxygen isotope relationship between the phosphate and structural carbonate fractions of human bioapatite." *Rapid Communications in Mass Spectrometry* 26, 3: 309-319. DOI:10.1002/rcm.5331.

Chesson, L.A., and G.E. Berg. 2021. "The use of stable isotopes in postconflict forensic identification." *Wiley Interdisciplinary Reviews: Forensic Science* 4, 2: e1439. DOI:10.1002/wfs2.1439.

Chesson, L.A., B.J. Tipple, J.R. Ehleringer, T. Park, and E.J. Bartelink. 2018. "Forensic applications of isotope landscapes ("isoscapes") A tool for predicting region-of-origin in forensic anthropology cases." *Forensic anthropology: theoretical framework and scientific basis*: 127-148.

Colleter, R., C.P. Bataille, H. Dabernat, D. Pichot, P. Hamon, S. Duchesne, F. Labaune-Jean, S. Jean, G. Le Cloirec, S. Milano, M. Trost, S. Steinbrenner, M. Marchal, C. Guilbeau-Frugier, N. Telmon, É.

Crubézy, and K. Jaouen. 2021. "The last battle of Anne of Brittany: Solving mass grave through an interdisciplinary approach (paleopathology, biological anthropology, history, multiple isotopes and radiocarbon dating)." *PLoS One* 16, 5: e0248086. DOI: 10.1371/journal.pone.0248086.

Coplen, T.B. 1988. "Normalization of oxygen and hydrogen isotope data." *Chemical Geology: Isotope Geoscience Section* 72, no. 4: 293-297. DOI: 10.1016/0168-9622(88)90042-5.

Dansgaard, W. 1964. "Stable isotopes in precipitation." *Tellus* 16, 4: 436-468. DOI: 10.3402/tellusa.v16i4.8993.

Daux, V., C. Lécuyer, M. Héran, R. Amiot, L. Simon, F. Fourel, F. Martineau, N. Lynnerup, H. Reychler, and G. Escarguel. 2008. "Oxygen isotope fractionation between human phosphate and water revisited." *Journal of human evolution* 55, 6: 1138-1147. DOI: 10.1016/j.jhevol.2008.06.006.

Ehleringer, J.R., A.H. Thompson, D.W. Podlesak, G.J. Bowen, L.A. Chesson, T.E. Cerling, T. Park, P. Dostie, and H. Schwarcz. 2010. "A framework for the incorporation of isotopes and isoscapes in geospatial forensic investigations." *Isoscapes: Understanding movement, pattern, and process on earth through isotope mapping*: 357-387. DOI:10.1007/978-90-481-3354-3_17.

Ericson, J.E. 1985. "Strontium isotope characterization in the study of prehistoric human ecology." *Journal of human evolution* 14, 5: 503-514. DOI: 10.1016/S0047-2484(85)80029-4.

Evans, J.A., J. Montgomery, G. Wildman, and N. Boulton. 2010. "Spatial variations in biosphere 87Sr/86Sr in Britain." *Journal of the geological society* 167, 1: 1-4.

Faure, G., and T.M. Mensing. 2005. *Isotopes: principles and applications*. Hoboken, N.J.: John Wiley & Sons, Inc.

Font, L., G. Jonker, P.A. van Aalderen, E.F. Schiltmans, and G.R. Davies. 2015. "Provenancing of unidentified World War II casualties: Application of strontium and oxygen isotope analysis in tooth enamel." *Science & Justice* 55, 1: 10-17. DOI: 10.1016/j.scijus.2014.02.005.

Font, L., G. Van Der Peijl, C. Van Leuwen, I. Van Wetten, and G.R. Davies. 2015. "Identification of the geographical place of origin of an unidentified individual by multi-isotope analysis." *Science & Justice* 55, 1: 34-42. DOI: 10.1016/j.scijus.2014.06.011.

Fricke, H.C., J.R. O'Neil, and N. Lynnerup. 1995. "Oxygen isotope composition of human tooth enamel from medieval Greenland: linking climate and society." *Geology* 23, 10: 869-872.

Funck, J., C. Bataille, J. Rasic, and M. Wooller. 2021. "A bio-available strontium isoscape for eastern Beringia: a tool for tracking landscape use of Pleistocene megafauna." *Journal of Quaternary Science* 36, 1: 76-90. DOI: 10.1002/jqs.3262.

Galimov, E.M., V.S. Sevastyanov, E.V. Kulbachevskaya, and A.A. Golyavin. 2005. "Isotope ratio mass spectrometry: δ^{13}C and δ^{15}N analysis for tracing the origin of illicit drugs." *Rapid Communications in Mass Spectrometry* 19, 10: 1213-1216. DOI: 10.1002/rcm.1896.

Gat, J.R. 1996. "Oxygen and hydrogen isotopes in the hydrologic cycle." *Annual Review of Earth and Planetary Sciences* 24, 1: 225-262. DOI: 10.1146/annurev.earth.24.1.225.

Giovas, C.M., G.D. Kamenov, S.M. Fitzpatrick, and J.Krigbaum. 2016. "Sr and Pb isotopic investigation of mammal introductions: Pre-Columbian zoogeographic records from the Lesser Antilles, West Indies." *Journal of Archaeological Science* 69: 39-53. DOI: 10.1016/j.jas.2016.03.006.

Giovas, C.M., G.D. Kamenov, and J. Krigbaum. 2019. "^{87}Sr/^{86}Sr and ^{14}C evidence for peccary (Tayassuidae) introduction challenges accepted historical interpretation of the 1657 Ligon map of Barbados." *PloS one* 14, 5: e0216458. DOI: 10.1371/journal.pone.0216458.

Grimstead, D.N., S. Nugent, and J. Whipple. 2017. "Why a standardization of strontium isotope baseline environmental data is needed and recommendations for methodology." *Advances in Archaeological Practice* 5, 2: 184-195. DOI:10.1017/aap.2017.6.

Hedman, K.M., P.A. Slater, M.A. Fort, T.E. Emerson, and J.M. Lambert. 2018. "Expanding the strontium isoscape for the American midcontinent: Identifying potential places of origin for Cahokian and Pre-Columbian migrants." *Journal of Archaeological Science: Reports* 22: 202-213. DOI: 10.1016/j.jasrep.2018.09.027.

Hillson, S. 1996. *Dental anthropology*. Cambridge: Cambridge University Press.

Hobson, K.A., R. Barnett-Johnson, and T. Cerling. 2010. "Using isoscapes to track animal migration." *Isoscapes: Understanding movement, pattern, and process on Earth through isotope mapping*: 273-298. DOI:10.1007/978-90-481-3354-3_13.

Hobson K.A. and L. Wassenaar. 2019. *Tracking Animal Migration with Stable Isotopes*. London United Kingdom: Academic Press/Elsevier.

Hobson, K.A., S.L. Van Wilgenburg, L.I. Wassenaar, R.L. Powell, C.J. Still, and J.M. Craine. 2012. "A multi-isotope ($\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$) feather isoscape to assign Afrotropical migrant birds to origins." *Ecosphere* 3, 5: 1-20. DOI: 10.1890/ES12-00018.1.

Hodell, D.A., R.L. Quinn, M. Brenner, and G. Kamenov. 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, 5: 585-601. DOI: 10.1016/j.jas.2003.10.009.

Hoppe, K.A., P.L. Koch, and T.T. Furutani. 2003. "Assessing the preservation of biogenic strontium in fossil bones and tooth enamel." *International Journal of Osteoarchaeology* 13, 1-2: 20-28. DOI: 10.1002/oa.663.

IAEA/WMO. 2016. "Global network of isotopes in precipitation." *The GNIP Database*. http://www.iaea.org/water.

International Atomic Energy Agency. 2013. "RCWIP (Regionalized Cluster-Based Water Isotope Prediction) Model-gridded precipitation $\delta^{18}O$ and $\delta^{2}H$ isoscape data." International Atomic Energy Agency, Vienna, Austria. http://www.iaea.org/water.

Kamenov, G.D., and J.H. Curtis. 2017. "Using carbon, oxygen, strontium, and lead isotopes in modern human teeth for forensic investigations: a critical overview based on data from Bulgaria." *Journal of forensic sciences* 62, 6: 1452-1459. DOI: 10.1111/1556-4029.13462.

Kootker, L.M., E. Plomp, S.T.M Ammer, V. Hoogland, and G.R. Davies. 2020. "Spatial patterns in $^{87}Sr/^{86}Sr$ ratios in modern human dental enamel and tap water from the Netherlands: implications for forensic provenancing." *Science of the Total Environment* 729: 138992. DOI: 10.1016/j.scitotenv.2020.138992.

Kramer, R.T., E.J. Bartelink, N.P. Herrmann, C.P. Bataille, and K. Spradley. 2020. "Application of stable isotopes and geostatistics to infer region of geographical origin for deceased undocumented Latin American migrants." *Forensic Science and Humanitarian Action: Interacting with the Dead and the Living*: 425-440.

Laffoon, J.E. 2012. "Patterns of paleomobility in the ancient Antilles: an isotopic approach.". PhD diss., Leiden University. https://hdl.handle.net/1887/20072.

Laffoon, J.E., Y. Chinique de Armas. 2022. "Investigating Variability in Indigenous Lifeways in the Pre-Colonial Greater Antilles: New multiple isotope results from Cuba". Proceedings of the 29th Congress of the International Association for Caribbean Archaeology.

Laffoon, J.E., G.R. Davies, M.L.P Hoogland, and C.L. Hofman. 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, 7: 2371-2384. DOI: 10.1016/j.jas.2012.02.002.

Laffoon, J.E., M.L.P Hoogland, G.R. Davies, and C.L. Hofman. 2019. "A multi-isotope investigation of human and dog mobility and diet in the pre-colonial Antilles." *Environmental Archaeology* 24, 2: 132-148. DOI: 10.1080/14614103.2017.1322831.

Laffoon, J.E., R. Rodríguez Ramos, L. Chanlatte Baik, Y. Narganes Storde, M. Rodríguez Lopez, G.R. Davies, and C.L. Hofman. 2014. "Long-distance exchange in the precolonial Circum-Caribbean: A multi-isotope study of animal tooth pendants from Puerto Rico." *Journal of Anthropological Archaeology* 35: 220-233. DOI: 10.1016/j.jaa.2014.06.004.

Laffoon, J.E., T.F. Sonnemann, M.M. Antczak, and A. Antczak. 2018. "Sourcing nonnative mammal remains from Dos Mosquises Island, Venezuela: new multiple isotope evidence." *Archaeological and Anthropological Sciences* 10: 1265-1281. DOI: 10.1007/s12520-016-0453-6.

Laffoon, J.E., T.F. Sonnemann, T. Shafie, C.L. Hofman, U. Brandes, and G.R. Davies. 2017. "Investigating human geographic origins using dual-isotope ($^{87}Sr/^{86}Sr$, $\delta^{18}O$) assignment approaches." *PloS one* 12, 2: e0172562. DOI: 10.1371/journal.pone.0172562.

Laffoon, J.E., R. Valcárcel Rojas, and C.L. Hofman. 2013. "Oxygen and carbon isotope analysis of human dental enamel from the Caribbean: implications for investigating individual origins." *Archaeometry* 55, 4: 742-765. DOI: 10.1111/j.1475-4754.2012.00698.x.

Lee-Thorp, J.A. 2008. "On isotopes and old bones." *Archaeometry* 50, 6: 925-950. DOI: 10.1111/j.1475-4754.2008.00441.x.

Lehn, C., A. Rossmann, and M. Graw. 2015. "Provenancing of unidentified corpses by stable isotope techniques – presentation of case studies." *Science & Justice* 55, 1: 72-88. DOI: 10.1016/j.scijus.2014.10.006.

Lin, G.P., Y.H. Rau, Y.F. Chen, C.C. Chou, and W.G. Fu. 2003. "Measurements of δD and $\delta^{18}O$ stable isotope ratios in milk." *Journal of Food Science* 68, 7: 2192-2195. DOI: 10.1111/j.1365-2621.2003.tb05745.x.

Longinelli, A. 1984. "Oxygen isotopes in mammal bone phosphate: a new tool for paleohydrological and paleoclimatological research?" *Geochimica et cosmochimica Acta* 48, 2: 385-390. DOI: 10.1016/0016-7037(84)90259-X.

Lugli, F., A. Cipriani, L. Bruno, F. Ronchetti, C. Cavazzuti, and S. Benazzi. 2022. "A strontium isoscape of Italy for provenance studies." *Chemical Geology* 587: 120624. DOI: 10.1016/j.chemgeo.2021.120624.

Luz, B., Y. Kolodny, and M. Horowitz. 1984. "Fractionation of oxygen isotopes between mammalian bone-phosphate and environmental drinking water." *Geochimica et Cosmochimica Acta* 48, 8: 1689-1693. DOI: 10.1016/0016-7037(84)90338-7.

Makarewicz, C.A., and J. Sealy. 2015. "Dietary reconstruction, mobility, and the analysis of ancient skeletal tissues: expanding the prospects of stable isotope research in archaeology." *Journal of archaeological science* 56: 146-158. DOI: 10.1016/j.jas.2015.02.035.

Mehl, A.C.S.B., L. Yumi Ishida, and R. Alexandre de Faria. 2019. "Analysis of Carbon, Oxygen, Strontium and Lead Isotopes in Human Teeth: Inferences for Forensic Investigation." In *XXVI Brazilian Congress on Biomedical Engineering: CBEB 2018, Armação de Buzios, RJ, Brazil, 21-25 October 2018 (Vol. 1)*: 71-77. Singapore: Springer. DOI: 10.1007/978-981-13-2119-1_11.

Meier-Augenstein, W., and I. Fraser. 2008. "Forensic isotope analysis leads to identification of a mutilated murder victim." *Science & Justice* 48, 3: 153-159. DOI: 10.1016/j.scijus.2007.10.010.

Mickleburgh, H.L., J.E. Laffoon, J.R. Pagán Jiménez, A.A.A Mol, S. Walters, Z.J.M Beier, and C.L. Hofman. 2019. "Precolonial/early colonial human burials from the site of White Marl, Jamaica: New findings from recent rescue excavations." *International Journal of Osteoarchaeology* 29, 1: 155-161. DOI: 10.1002/oa.2707.

Nelson, B.K., M.J. DeNiro, M.J. Schoeninger, D.J. De Paolo, and P.E. Hare. 1986. "Effects of diagenesis on strontium, carbon, nitrogen and oxygen concentration and isotopic composition of bone." *Geochimica et Cosmochimica Acta* 50, 9: 1941-1949. DOI: 10.1016/0016-7037(86)90250-4.

Ostapkowicz, J., F. Brock, A.C. Wiedenhoeft, C. Snoeck, J. Pouncett, Y. Baksh-Comeau, R. Schulting, P. Claeys, N. Mattielli, M. Richards, and A. Boomert. 2017. "Black pitch, carved histories: radiocarbon dating, wood species identification and strontium isotope analysis of prehistoric wood carvings from Trinidad's Pitch Lake." *Journal of Archaeological Science: Reports* 16: 341-358. DOI: 10.1016/j.jasrep.2017.08.018.

Pederzani, S., and K. Britton. 2019. "Oxygen isotopes in bioarchaeology: Principles and applications, challenges and opportunities." *Earth-Science Reviews* 188: 77-107. DOI: 10.1016/j.earscirev.2018.11.005.

Pestle, W.J., A. Simonetti, and L.A. Curet. 2013. "$^{87}Sr/^{86}Sr$ variability in Puerto Rico: geological complexity and the study of paleomobility." *Journal of Archaeological Science* 40, 5: 2561-2569. DOI: 10.1016/j.jas.2013.01.020.

Plomp, E., I.C.C. von Holstein, J.M. Koornneef, R.J. Smeets, J.A. Baart, T. Forouzanfar, and G.R. Davies. 2019. "Evaluation of neodymium isotope analysis of human dental enamel as a provenance indicator using 10^{13} Ω amplifiers (TIMS)." *Science & Justice* 59, 3: 322-331. DOI: 10.1002/ajpa.24059.

Plomp, E., I.C.C von Holstein, L.M. Kootker, S.J.A Verdegaal-Warmerdam, T. Forouzanfar, and G.R. Davies. 2020. "Strontium, oxygen, and carbon isotope variation in modern human dental enamel." *American Journal of Physical Anthropology* 172, 4: 586-604. DOI: 10.1016/j.scijus.2019.02.001.

Podlesak, D.W., A. Torregrossa, J.R. Ehleringer, M.D. Dearing, B.H. Passey, and T.E. Cerling. 2008. "Turnover of oxygen and hydrogen isotopes in the body water, CO_2, hair, and enamel of a small mammal." *Geochimica et Cosmochimica Acta* 72, 1: 19-35. DOI: 10.1016/j.gca.2007.10.003

Price, T.D. 1989. "Multi-element studies of diagenesis in prehistoric bone." *The chemistry of prehistoric human bone*: 126-154.

Price, T.D., J.H. Burton, and R. A. Bentley. 2002. "The characterization of biologically available strontium isotope ratios for the study of prehistoric migration." *Archaeometry* 44, 1: 117-135. DOI: 10.1111/1475-4754.00047.

Price, T.D., J.H. Burton, and J.B. Stoltman. 2007. "Place of origin of prehistoric inhabitants of Aztalan, Jefferson Co., Wisconsin." *American Antiquity* 72, 3: 524-538. DOI: 10.2307/40035859.

Scaffidi, B.K., and K.J. Knudson. 2020. "An archaeological strontium isoscape for the prehistoric Andes: Understanding population mobility through a geostatistical meta-analysis of archaeological $^{87}Sr/^{86}Sr$ values from humans, animals, and artifacts." *Journal of Archaeological Science* 117: 105121. DOI: 10.1016/j.jas.2020.105121.

Schellenberg, A., S. Chmielus, C. Schlicht, F. Camin, M. Perini, L. Bontempo, K. Heinrich, S.D. Kelly, A. Rossmann, F. Thomas, E. Jamin, and M. Horacek. 2010. "Multielement stable isotope ratios (H, C, N, S) of honey from different European regions." *Food chemistry* 121, 3: 770-777. DOI: 10.1016/j.foodchem.2009.12.082.

Schulting, R., M. Richards, J. Pouncett, B. Naqqi Manco, E. Freid, and J. Ostapkowicz. 2018. "Absence of Saharan dust influence on the strontium isotope ratios on modern trees from the Bahamas and

Turks and Caicos Islands." *Quaternary Research* 89, 2: 394-412. DOI:10.1017/qua.2018.8.

Schwarcz, H.P., L. Gibbs, and M. Knyf. 1991. "Oxygen isotope analysis as an indicator of place of origin." In *Snake Hill: An Investigation of a Military Cemetery from the War of 1812,* edited by S. Pfeiffer and R. Williamson: 263-268. Toronto: Dundurn.

Sealy, J.C., N.J. van der Merwe, A. Sillen, F.J. Kruger, and H.W. Krueger. 1991. "$^{87}Sr/^{86}Sr$ as a dietary indicator in modern and archaeological bone." *Journal of Archaeological Science* 18, 3: 399-416. DOI:10.1016/0305-4403(91)90074-Y.

Sharpe, A.E., N. Smith-Guzmán, J. Curtis, I. Isaza-Aizpurúa, G.D. Kamenov, T.A. Wake, and R.G. Cooke. 2021. "A preliminary multi-isotope assessment of human mobility and diet in pre-Columbian Panama." *Journal of Archaeological Science: Reports* 36: 102876. DOI:10.1016/j.jasrep.2021.102876.

Snoeck, C., J. Pouncett, P. Claeys, S. Goderis, N. Mattielli, M. Parker Pearson, C. Willis, A. Zazzo, J.A. Lee-Thorp, and R.J. Schulting. 2018. "Strontium isotope analysis on cremated human remains from Stonehenge support links with west Wales." *Scientific reports* 8, 1: 10790. DOI:10.1038/s41598-018-28969-8.

Terzer, S., L.I. Wassenaar, L.J. Araguás-Araguás, and P.K. Aggarwal. 2013. "Global isoscapes for $\delta^{18}O$ and $\delta^{2}H$ in precipitation: improved prediction using regionalized climatic regression models." *Hydrology and Earth System Sciences* 17, 11: 4713-4728. DOI:10.5194/hess-17-4713-2013.

Trickett, M.A., P. Budd, J. Montgomery, and J. Evans. 2003. "An assessment of solubility profiling as a decontamination procedure for the $^{87}Sr/^{86}Sr$ analysis of archaeological human skeletal tissue." *Applied Geochemistry* 18, 5: 653-658. DOI:10.1016/S0883-2927(02)00181-6.

Ueda, M., and L.S. Bell. 2021. "Assessing the predictability of existing water-to-enamel geolocation models against known human teeth." *Scientific Reports* 11, 1: 15645. DOI:10.1038/s41598-021-95153-w.

Valentine, B., G.D. Kamenov, J.M. Kenoyer, V. Shinde, V. Mushrif-Tripathy, E. Otarola-Castillo, and J. Krigbaum. 2015. "Evidence for patterns of selective urban migration in the Greater Indus Valley (2600-1900 BC): a lead and strontium isotope mortuary analysis." *PLoS One* 10, 4: e0123103. DOI:10.1371/journal.pone.0123103.

Van der Merwe, N.J., J.A. Lee-Thorp, J.F. Thackeray, A. Hall-Martin, F.J. Kruger, H. Coetzee, R.H.V. Bell, and M. Lindeque. 1990. "Source-area determination of elephant ivory by isotopic analysis." *Nature* 346, 6286: 744-746. DOI:10.1038/346744a0.

West, J.B., G.J. Bowen, T.E. Dawson, and K.P. Tu. 2010. *Isoscapes: understanding movement, pattern, and process on Earth through isotope mapping*. Dordrecht: Springer Science & Business Media. DOI:10.1007/978-90-481-3354-3.

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, 4: 555-566. DOI:10.1016/j.jas.2004.11.011.