Strontium isotopes (^{87}Sr/^{86}Sr) in terrestrial ecological and palaeoecological research: empirical efforts and recent advances in continental-scale models

Brooke E. Crowley[1,2,†,*], Joshua H. Miller[1,3,†] and Clément P. Bataille[4]

[1]*Department of Geology, University of Cincinnati, 500 Geology and Physics Building, Cincinnati, OH 45221, U.S.A.*
[2]*Department of Anthropology, 481 Braunstein, University of Cincinnati, Cincinnati, OH 45221, U.S.A.*
[3]*Department of Paleobiology, National Museum of Natural History, Smithsonian Institution, PO Box 37012, Washington, DC 20013, U.S.A.*
[4]*Chevron Corporation, 1500 Louisiana Street, Houston, TX 77002, U.S.A.*

ABSTRACT

Strontium (Sr) isotope analysis can provide detailed biogeographical and ecological information about modern and ancient organisms. Because Sr isotope ratios (^{87}Sr/^{86}Sr) in biologically relevant materials such as water, soil, vegetation, and animal tissues predominantly reflect local geology, they can be used to distinguish geologically distinct regions as well as identify highly mobile individuals or populations. While the application of Sr isotope analysis to biological research has been steadily increasing, high analytical costs have prohibited more widespread use. Additionally, accessibility of this geochemical tool has been hampered due to limited understanding of (*i*) the degree to which biologically relevant materials differ in their spatial averaging of ^{87}Sr/^{86}Sr ratios, and (*ii*) how these differences may be affected by lithologic complexity. A recently developed continental-scale model that accounts for variability in bedrock weathering rates and predicts Sr isotope ratios of surface water could help resolve these questions. In addition, if this 'local water' model can accurately predict ^{87}Sr/^{86}Sr ratios for other biologically relevant materials, there would be reduced need for researchers to assess regional Sr isotope patterns empirically. Here, we compile ^{87}Sr/^{86}Sr data for surface water, soil, vegetation, and mammalian and fish skeletal tissues from the literature and compare the accuracy with which the local water model predicts Sr isotope data among these five materials across the contiguous USA. We find that measured Sr isotope ratios for all five materials are generally close to those predicted by the local water model, although not with uniform accuracy. Mammal skeletal tissues are most accurately predicted, particularly in regions with low variability in ^{87}Sr/^{86}Sr predicted by the local water model. Increasing regional geologic heterogeneity increases both the offset and variance between modelled and empirical Sr isotope ratios, but its effects are broadly similar across materials. The local water model thus provides a readily available source of background data for predicting ^{87}Sr/^{86}Sr for biologically relevant materials in places where empirical data are lacking. The availability of increasingly high-quality modelled Sr data will dramatically expand the accessibility of this geochemical tool to ecological applications.

Key words: water, soil, vegetation, skeletal tissues, fish, mammal, provenance.

CONTENTS

* Address for correspondence at address 1 (Tel: +1.513.556.7181; Fax: +1.513.556.6931; E-mail: brooke.crowley@uc.edu).

† Authors contributed equally.

I. INTRODUCTION

Strontium (Sr) isotope analysis is a powerful and increasingly utilized tool for studying spatial aspects of modern, historical, and fossil ecosystems. In addition to identifying habitat use and migration routes of individuals (e.g. Chamberlain *et al.*, 1997; Bacon *et al.*, 2004; Feranec, Hadly & Paytan, 2007; Hoppe & Koch, 2007; Gibson-Reinemer *et al.*, 2009; Sellick *et al.*, 2009; Baumann & Crowley, 2015), Sr isotopes can be used to detect the source of poached animals (e.g. Vogel, Eglington & Auret, 1990; Koch *et al.*, 1995), and identify the origin and range expansions of introduced species (e.g. Wolff *et al.*, 2012). Sr isotopes are useful for provenance studies because their spatial variability is primarily driven by underlying geology. Such spatially discrete isotopic patterning can be highly effective for documenting patterns of landscape use and mobility, and may be further enhanced when combined with other stable isotopes such as carbon (e.g. Hoppe, 2004; Britton, 2009; Widga, Walker & Stockii, 2010), nitrogen (e.g. Koch *et al.*, 1995), oxygen (e.g. Hoppe, 2004; Evans, Chenery & Fitzpatrick, 2006; Britton, 2009; Widga *et al.*, 2010; Baumann & Crowley, 2015), hydrogen (e.g. Matthews, 2007; Sellick *et al.*, 2009), or sulfur (e.g. Britton, 2009).

Early applications of Sr isotope analysis focused on quantifying weathering rates for minerals and rocks (e.g. Blum, Erel & Brown, 1993; Miller, Blum & Friedland, 1993; Horton *et al.*, 1999; Rose & Fullagar, 2005), and the sources and fates of Sr in rivers (e.g. Brass, 1976; Wadleigh, Veizer & Brooks, 1985; Goldstein & Jacobsen, 1987; Palmer & Edmond, 1989). However, the potential ecological insight offered by Sr isotope analysis is growing dramatically due to analytical advances (e.g. high performance laser ablation and multicollector inductively coupled plasma mass spectrometry), as well as recent regional- and continental-scale

models of Sr isotope distributions (e.g. Bataille & Bowen, 2012; Bataille, Lafoon & Bowen, 2012; Bataille *et al.*, 2014).

The relevance of Sr isotopes to ecological research has been recognized for over two decades. Ericson (1985) first demonstrated that Sr isotope ratios in human tooth enamel from archaeological contexts reflect local geology. Vogel *et al.* (1990) confirmed that Sr isotope ratios in elephant bone and ivory distinguish individuals from localities on different types of bedrock, and shortly thereafter, Sealy *et al.* (1991) demonstrated that biogenic Sr isotope ratios are preserved in fossil bone. Sr isotopes in fossil bone and dentine have the potential to be diagenetically altered, but fossil enamel is relatively resistant to post-burial alteration (Hoppe, Koch & Furutani, 2003). This increases the possibility of looking at changes in organismal mobility over time. Yet despite these advances, until recently, the number of studies utilizing Sr ratios remained relatively limited. Over the past decade, scientists have increasingly employed Sr isotope ratios to address questions about the mobility and origin of a variety of modern and ancient organisms including anadromous fish (e.g. Koch *et al.*, 1992; Kennedy *et al.*, 2000; Bacon *et al.*, 2004; Barnett-Johnson *et al.*, 2008; Walther & Thorrold, 2008; Miller *et al.*, 2011), migratory songbirds (e.g. Chamberlain *et al.*, 1997; Blum, Taliaferro & Holmes, 2001; Sellick *et al.*, 2009), large herbivores (e.g. Sealy *et al.*, 1991; Koch *et al.*, 1995; Hoppe *et al.*, 1999; Hoppe, 2004; Britton, 2009; Hedman *et al.*, 2009; Radloff *et al.*, 2010; Widga *et al.*, 2010; Baumann & Crowley, 2015), extinct hominins (Copeland *et al.*, 2012), and historic populations of people (e.g. Ericson, 1985; Ezzo, Johnson & Price, 1997; Price, Burton & Bentley, 2002; Bentley, 2006; Evans *et al.*, 2006; Juarez, 2008; Maurer *et al.*, 2012). Sr is also useful for sourcing agricultural products such as rice (Kawasaki, Oda & Hirata, 2002), corn (Benson, 2010), wine (Marchionni *et al.*, 2013), milk (Crittenden *et al.*, 2007), and illicit drugs (West *et al.*,

2009). Such provenance applications are possible because Sr isotope ratios in biological materials (e.g. soils, vegetation, and animal tissues) largely reflect those in local bedrock, which are dictated by rock age and mineralogy (Åberg, 1995).

Although there is great potential for using Sr isotope analysis to enhance the study of ecological systems, there has been little work synthesizing current knowledge of how different biologically relevant materials (e.g. surface water, ossified tissues from mammals or fish, vegetation, and soil) integrate local-to-regional Sr from weathered bedrock. Additionally, the accuracy of regional-scale models for predicting Sr isotope ratios in different materials is poorly constrained. Here, we compile a database of published Sr isotope data for surface water, soil, vegetation, and skeletal tissues from mammals and fish for the contiguous USA. Using this data set, we first evaluate the geographic coverage for each material. Next, we assess the accuracy at which a continental-scale local water model (Bataille & Bowen, 2012) can estimate Sr isotope ratios in biologically relevant materials. We also test how regional variability in lithology impacts the model's predictive power. This comprehensive survey summarizes the current state of knowledge on Sr isotope ratios in ossified tissues, vegetation, soil, and surface water across the USA. It also illustrates where additional empirical data would help resolve remaining uncertainties.

II. BACKGROUND ON STRONTIUM ISOTOPES

There are four naturally occurring isotopes of Sr. Three of these isotopes are stable (^{88}Sr, ^{86}Sr, and ^{84}Sr), while the fourth (^{87}Sr) is produced by the β-decay of rubidium (^{87}Rb) (reviewed in Banner, 2004; Bentley, 2006). Most applications of Sr isotopes compare the amount of radiogenic ^{87}Sr to the amount of stable ^{86}Sr in a given material. Sr isotope ratios vary considerably in rocks at the Earth's surface because they are affected by both chemical composition and age. For example, whereas mafic basalts display ^{87}Sr/^{86}Sr ratios between approximately 0.702 and 0.705 (which are reflective of the Earth's mantle), continental crustal rocks, such as granites, exhibit Sr isotope ratios around 0.716 (reviewed in Capo, Stewart & Chadwick, 1998; Banner, 2004). Carbonates form in equilibrium with sea water, which has had a relatively constant ^{87}Sr/^{86}Sr composition of 0.707–0.709 throughout the Phanerozoic (DePaolo & Ingram, 1985; McArthur, Howarth & Bailey, 2001). As a rock ages, its ^{87}Sr/^{86}Sr increases due to continued production of ^{87}Sr. Consequently, siliciclastic sediments typically have higher ^{87}Sr/^{86}Sr ratios than carbonates because they contain older detrital material (Bataille *et al.*, 2014). Very old (i.e. Archaean and Proterozoic) igneous rocks can exhibit Sr isotope ratios >0.8 (reviewed in Banner, 2004; Bataille & Bowen, 2012). Due to the extended half-life of ^{87}Rb (48.8×10^9 years), these patterns are stable over ecologically meaningful timescales.

The incorporation of Sr into biological systems begins with the weathering of rocks and subsequent release of Sr into soils and surface water. Mobilized Sr can then be incorporated into plants and animal tissues (e.g. Toots & Voorhies, 1965; Rosenthal, Cochran & Eves, 1972). The biological uptake of Sr (which readily substitutes for Ca^{2+} ions) occurs with negligible fractionation (reviewed in Åberg, 1995; Capo *et al.*, 1998; Banner, 2004), and the minimal fractionation that does occur can be accounted for during data normalization (Krabbenhoft *et al.*, 2010). Thus, for example, the Sr isotope composition of an animal's tissues reflects an integrated signal of Sr sources ingested by the organism during tissue formation. Therefore, ^{87}Sr/^{86}Sr can be used to track animal movement across landscapes or distinguish migrant and local individuals within a population (e.g. Chamberlain *et al.*, 1997; Hoppe & Koch, 2007; Sellick *et al.*, 2009; Baumann & Crowley, 2015).

The use of Sr isotopes for reconstructing an organism's geographical history or other ecological information relies on good knowledge of local to regional isotopic variability. These critical reference data can be empirically derived or modelled. Both approaches have advantages and disadvantages as discussed below.

(1) Empirical data

Measuring Sr isotope ratios for samples collected in the field is a primary means of quantifying variability in biologically available Sr. Empirical data are also essential for evaluating model predictions of bioavailable Sr. Although there are now automated analytical systems that allow for relatively high analytical throughput (e.g. Chesson *et al.*, 2012), the time and costs associated with collecting and analysing local samples for Sr isotope ratios remain substantial. Additionally, variability in the spatial and temporal span incorporated by different materials may pose challenges to comparing their Sr isotope ratios directly.

Local Sr isotope ratios are typically evaluated using surface water (e.g. Chesson *et al.*, 2012), soils (Ingram & Weber, 1999; Barnett-Johnson *et al.*, 2008; Walther, Thorrold & Olney, 2008; Wolff *et al.*, 2012; Baumann & Crowley, 2015), vegetation (Keller, Blum & Kling, 2007; Reynolds, Quade & Betancourt, 2012), or ossified tissues from animals with limited home ranges (Hoppe *et al.*, 1999; Blum *et al.*, 2000; Porder, Payton & Hadly, 2003; Widga *et al.*, 2010; Christian, Banner & Mack, 2011; Reynolds *et al.*, 2012). While each of these materials offers meaningful data, they differ in how they integrate 'local' conditions. The spatial and temporal breadth across which materials accumulate Sr may also impact their isotopic comparability (e.g. Ezzo *et al.*, 1997; Hoppe *et al.*, 1999; Blum *et al.*, 2001; Porder *et al.*, 2003; Feranec *et al.*, 2007; Hedman *et al.*, 2009; Radloff *et al.*, 2010; Maurer *et al.*, 2012; Slater, Hedman & Emerson, 2014; Brennan *et al.*, 2015). For example, because root and soil depths vary, contributions from available Sr sources (e.g. weathering bedrock *versus* atmospheric or colluvial material) may differ among individual plant and soil samples (Hodell *et al.*, 2004; Poszwa *et al.*, 2004; Frei & Frie, 2011). Conversely, animal or surface water samples may incorporate Sr from multiple soil depths, or integrate Sr across a variety of geographic

scales (e.g. Graustein & Armstrong, 1983; Gosz & Moore, 1989; Miller *et al.*, 1993; Poszwa *et al.*, 2004; Bern, Townsend & Farmer, 2005; Chadwick *et al.*, 2009; Pett-Ridge, Derry & Kurtz, 2009; Reynolds *et al.*, 2012). Additionally, biologically relevant materials differ in the amount of time they take to form (and therefore in the amount of time they incorporate Sr). Whereas most mammalian teeth form over $1-3$ years, mammalian bone averages Sr input on annual to decadal scales (depending on lifespan). Otoliths form incrementally throughout a fish's life, foliage incorporates Sr on seasonal to annual scales, and soils may differentially integrate Sr across decades to millennia. Finally, the sources for a particular body of water may vary seasonally (due to shifting precipitation or run-off sources; e.g. Curtis & Stueber, 1973; Douglas, Chamberlain & Blum, 2002; Price *et al.*, 2002; Rose & Fullagar, 2005; Feranec *et al.*, 2007; Hamann & Kennedy, 2012; Brennan *et al.*, 2015), or on longer timescales (due to changes in bedrock weathering; e.g. Douglas *et al.*, 2013; Brennan *et al.*, 2015). Thus, the comparability of Sr data among different materials is a topic of debate (Semhi, Clauer & Probst, 2000; Walther & Thorrold, 2009).

Local to regional geologic complexity (i.e. lithological, structural, and temporal heterogeneity) also plays an important role in shaping variability in $^{87}Sr/^{86}Sr$ among sites. In geologically simple areas, such as ancient ocean basins, Sr isotope ratios should be relatively homogenous (e.g. Sillen *et al.*, 1998; Blum *et al.*, 2001; Keller *et al.*, 2007; Hedman *et al.*, 2009; Frei & Frie, 2011; Maurer *et al.*, 2012; Reynolds *et al.*, 2012). However, in lithologically, structurally, or temporally complex regions (e.g. mountainous zones, accretionary terranes), Sr isotope ratios are expected to be more variable (Hoppe *et al.*, 1999).

(2) Modelled data

To date, spatially explicit models of Sr isotope ratios, or 'Sr isoscapes', have only been developed for the USA and the circum-Caribbean (Xin, 1993; Sillen *et al.*, 1998; Blum *et al.*, 2000; Porder *et al.*, 2003; Maurer *et al.*, 2012; Reynolds *et al.*, 2012; Hegg, Kennedy & Fremier, 2013b). Modelled Sr ratios can provide local and regional reference data without the need to conduct laborious field and laboratory studies. For example, modelled Sr ratios can be used to assess quickly whether or not an individual is local to a specific site and identify potential geographic origins for immigrants (reviewed in Beard & Johnson, 2000; Bataille & Bowen, 2012; Bataille *et al.*, 2012, 2014). The ubiquitous challenges for Sr models are resolution limitations, spatial variability of model quality, and reliance on empirical data. Consequently, the accuracy of models varies with geographic location, bedrock complexity, and the presence of allochthonous surficial geology (e.g. alluvium, glacial drift).

The first strontium isoscape for North America was based solely on bedrock age (Hobson, Barnett-Johnson & Cerling, 2010). This model provided a starting point for testing the consistency of biologically available Sr isotope ratios for localities underlain by older and younger rock, and identifying the origins of non-local migratory animals.

Unfortunately, the match between modelled and empirical data was relatively poor, in part because the model did not account for differential weathering among lithologies. Although Sr isotope ratios in biological materials largely reflect bedrock, they do not necessarily reflect bulk bedrock. For example, because carbonates contain more Sr and weather faster than siliciclastics, bioavailable Sr from a region underlain by a mixture of clastic and chemical sedimentary rocks will be biased towards the $^{87}Sr/^{86}Sr$ of carbonate (Beard & Johnson, 2000). Similarly, different minerals within a single igneous or metamorphic rock can differ in their Sr isotope ratios, and those that weather more quickly (e.g. potassium feldspar or micas) will have a greater influence on bioavailable Sr (Stueber, Pushkar & Baldwin, 1972; Goldstein & Jacobsen, 1987; Horton *et al.*, 1999; Pretti & Stewart, 2002). Differential weathering may also cause glacial drift or alluvium to contribute more to the bioavailable Sr pool than local bedrock (e.g. Blum *et al.*, 1993; Rose & Fullagar, 2005). Such biases can result in poor matches between measured Sr isotope ratios in plants, animals, soils and water, and those estimated by bedrock models (Curtis & Stueber, 1973; Widga *et al.*, 2010; Hegg *et al.*, 2013b; Baumann & Crowley, 2015).

Recently, Bataille & Bowen (2012) developed a local water model that predicts $^{87}Sr/^{86}Sr$ in surface waters by coupling a bedrock model with a chemical weathering model that accounts for variable contributions of different lithologies to dissolved Sr in soil water. Establishing the degree to which this local water model predicts Sr isotope ratios for additional biologically relevant materials could significantly advance the accessibility of Sr isotopes to ecological research in the USA. However, a systematic test of its predictive power across multiple materials and across the model's spatial extent is required to assess fully the potential of this tool for provenance studies. Using our compiled database of empirical Sr data and the local water model, we test two hypotheses: (*i*) modelled $^{87}Sr/^{86}Sr$ more accurately predicts empirical Sr isotope ratios for water and animals (which are capable of averaging Sr across large regions) than soils and vegetation (which sample Sr from more localized areas); (*ii*) because areas that are less complex with respect to lithology and rock age can have more constant Sr isotope ratios at smaller spatial scales, we expect that modelled data from homogenous regions predict $^{87}Sr/^{86}Sr$ of biologically relevant materials better than modelled output for more heterogeneous regions. Specifically, we hypothesize that the variance of modelled *versus* empirical Sr isotope ratios is larger in more geologically complex regions.

III. METHODS

(1) Data acquisition

We compiled Sr isotope data for water, soil, and biological materials from the contiguous USA. Among the 69 works consulted (Hart & Tilton, 1966; Eastin & Faure, 1970; Jones & Faure, 1972; Stueber *et al.*, 1972; Curtis & Stueber,

1973; Brass, 1976; Fisher & Stueber, 1976; Graustein & Armstrong, 1983; Ericson, 1985; Wadleigh *et al.,* 1985; Stordal & Wasserburg, 1986; Goldstein & Jacobsen, 1987; Gosz & Moore, 1989; Palmer & Edmond, 1989; Koch *et al.,* 1992; Blum *et al.,* 1993, 2000, 2001; Miller *et al.,* 1993; Xin, 1993; Price *et al.,* 1994, 2002; Yang, Telmer & Veizer, 1996; Chamberlain *et al.,* 1997; Ezzo *et al.,* 1997; Hoppe *et al.,* 1999; Ingram & Weber, 1999; Beard & Johnson, 2000; Kennedy *et al.,* 2000; English *et al.,* 2001; Gross *et al.,* 2001; Douglas *et al.,* 2002; Pretti & Stewart, 2002; Van der Hoven & Quade, 2002; Cooke *et al.,* 2003; Porder *et al.,* 2003; Bacon *et al.,* 2004; Hoppe, 2004; Rose & Fullagar, 2005; Benson *et al.,* 2006; Singleton *et al.,* 2006; Hoppe & Koch, 2007; Price, Burton & Stoltman, 2007; Barnett-Johnson *et al.,* 2008; Quinn, Tucker & Krigbaum, 2008; Walther & Thorrold, 2008; Walther *et al.,* 2008; Benson, Stein & Taylor, 2009; Britton, 2009; Gibson-Reinemer *et al.,* 2009; Hedman *et al.,* 2009; Jorgenson *et al.,* 2009; Sellick *et al.,* 2009; West *et al.,* 2009; Benson, 2010; Widga *et al.,* 2010; Beehr, 2011; Christian *et al.,* 2011; Miller *et al.,* 2011; Chesson *et al.,* 2012; Hamann & Kennedy, 2012; Reynolds *et al.,* 2012; Wolff *et al.,* 2012; McCall, 2013; Hegg *et al.,* 2013*a*; Eerkens *et al.,* 2014; Slater *et al.,* 2014; Baumann & Crowley, 2015), data were available for modern precipitation, tap, surface and ground water, soil, dust, vegetation, feathers, shells, osseous material (dentine, bone and enamel) from a variety of modern and historic terrestrial and aquatic organisms, and enamel from Pleistocene fossil mammals (see Tables S1 and S2). Materials from archaeological sites were excluded because they are at risk of having been moved outside of their 'natural' range by humans. Although we endeavoured to include data for individual specimens, only summary data were available for some studies. When geographic coordinates of specimens were not included in publications, we used Google Earth to georeference reported geographic information (e.g. maps or locality names). When necessary, authors of publications were contacted to clarify locality information.

(2) Data analysis

(a) Predicting $^{87}Sr/^{86}Sr$

We focused our analyses on the $^{87}Sr/^{86}Sr$ ratios of five materials with good sample sizes and the widest geographic coverage: (*i*) surface water (lakes, rivers, streams and run-off), (*ii*) vegetation (wood, leaves, and roots from lichens, mosses, trees, shrubs, herbs and grasses), (*iii*) osseous tissues of mammals (bone, dentine and enamel), (*iv*) osseous tissues of fish (bone and otoliths), and (*v*) soil. Other materials (e.g. shells, feathers, dust and precipitation) were excluded from analysis due to small sample sizes. Prior to analysis, all specimens with multiple Sr data points (e.g. serially sampled teeth or water samples collected repeatedly from the same locality) were summarized to their median ratios.

Using the Bataille & Bowen (2012) local water model raster (http://wateriso.utah.edu/waterisotopes/pages/data_access/ArcGrids.html), we evaluated the modelled (expected) Sr isotope ratio for each specimen according to its georeferenced location. To account for spatial uncertainties in the local water model (see Appendix S1) as well as uncertainty in the georeferencing of empirical data points, expected (modelled) $^{87}Sr/^{86}Sr$ for each sample was calculated as the median Sr isotope ratio within a 5 km radius of the empirical point's geographic coordinates. Using the exact coordinate values to recover model estimates of empirical samples (or median values calculated across larger sampling radii) generates highly similar results. Specimens from the few areas where the local water model did not have estimated Sr isotope ratios were removed from the analysis.

We then calculated the difference between the observed (empirical) and modelled (expected) $^{87}Sr/^{86}Sr$ for each specimen. Given a perfect model, the expected ratio should exactly match the observed ratio of the specimen. Therefore, if the model performs well, observed *versus* expected ratios (i.e. 'Sr offset') should generally approach zero. To evaluate this expectation, we created histograms of Sr offset for specimens from each of the five materials and evaluated their frequency distributions using their kurtosis (peakedness) and skewness (severity of distribution tails). We further used distribution of Sr offsets to test for overall agreement between the empirical and modelled data by calculating a bootstrapped 95% confidence interval around the median ratio for each material. If the 95% confidence interval includes Sr offsets of 0.0, this is an indication that the local water model is a strong predictor of the Sr ratio for a particular material. We also evaluated overall model accuracy for each material by calculating the percent error for each specimen as the magnitude (absolute value) of Sr offset divided by the empirical (measured) Sr ratio, multiplied by 100. Finally, we plotted Sr offset for each specimen on maps of the contiguous USA to examine the spatial coverage for available empirical data for each material.

(b) Sr complexity, geographic standardization and spatial averaging

Spatial variability in Sr isotope ratios is directly related to geologic complexity, which may affect the predictive accuracy of the local water model. We used the regional isotopic variability predicted by the local water model to test the relationship between Sr offset and geologic (Sr) complexity. Specifically, we defined 'Sr complexity' as the standard deviation of modelled Sr ratios within a 50 km radius of each specimen's georeferenced location. Because empirically based studies are usually limited to one or a few localities, and because some studies produce more data than others, individual publications and the geologic settings in which their data were collected could dominate the analysis and bias the derived relationship between Sr offset and complexity. To mitigate these biases, we geographically standardized the data set by calculating the median Sr offset for all specimens of a particular material sampled from the same locality.

Using the geographically standardized data, we then applied quantile regression to quantify relationships between Sr complexity and the magnitude of Sr offset for each material. We separately analysed the 25th, 50th, and 75th quantiles for each material's distribution using the R library
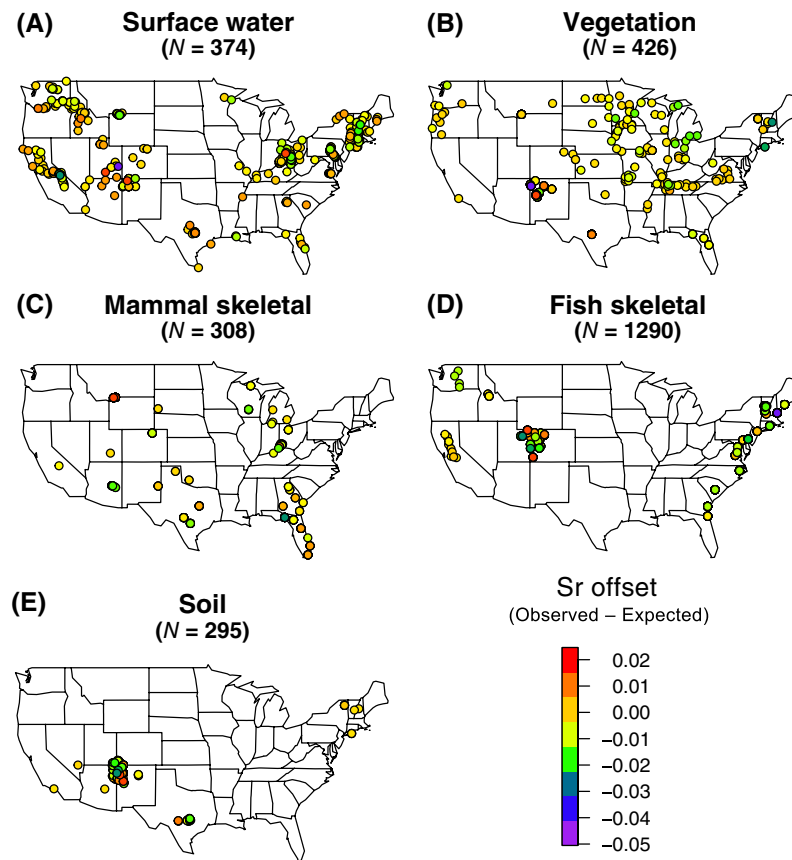
Fig. 1. Maps of compiled empirical Sr data for (A) surface water, (B) vegetation, (C) mammal skeletal tissues, (D) fish skeletal tissues, and (E) soil. Point colours illustrate the degree to which the Bataille & Bowen (2012) local water model overpredicts (negative values, cool colours), and underpredicts (positive values, warm colours) empirical data. Geographic distribution and sample sizes vary among materials. Sample numbers are raw and have not been geographically standardized.

'quantreg' (Hobson *et al.,* 2010; Bataille & Bowen, 2012; Chesson *et al.,* 2012) and bootstrap routine to estimate regression significance. Quantile regression is ideally suited for this analysis because relationships between Sr offset and Sr complexity are not expected to be simple. For example, it is intuitive that the local water model would accurately predict the Sr isotope ratios for mammals from a geologically homogenous region, and the variability in the quality of that prediction should be low. However, in a region with greater Sr complexity, $^{87}Sr/^{86}Sr$ for some individuals may be poorly predicted (due to the incorporation of more isotopically variable Sr or limitations in the resolution of the model). Thus, the variance in Sr offset is also expected to increase with increasing Sr complexity, producing a polygonal relationship. This is particularly likely for water bodies and animals, which can (but may not necessarily) integrate Sr across large geographic areas. To test our expectation that variance increases with Sr complexity (the standard deviation of modelled Sr ratios within 50 km from the sample location), we binned Sr offset into 0.01 units of standard deviation: low (<0.01), medium (0.100–0.199), and high (>0.200), and tested for stepwise changes in variance with increasing Sr complexity using Levene tests (Koenker, 2015).

Finally, to confirm the degree to which spatial averaging of a particular tissue might affect the predictive accuracy of the local water model, we also compared Sr offsets between small (<100 kg) and large (>100 kg) mammals (Table S1), anadromous and freshwater fish (Table S2), and fish and surface waters from small (<1000 km^2) *versus* large (>1000 km^2) watersheds using Wilcoxon rank sum tests (Sokal & Rohlf, 2012). We limited these comparisons to regions with low Sr complexity predicted by the local water model, which is where sample sizes and mean differences in $^{87}Sr/^{86}Sr$ among materials are the largest. All analyses were conducted in R version 3.1.1 (R Development Core Team, 2014), including the packages raster (Hijmans, 2015), fields (Nychka, Furrer & Sain, 2015), quantreg (Koenker, 2015), car (Fox & Weisberg, 2010) and moments (Komsta & Novomestky, 2015).

IV. RESULTS

(1) Geographic coverage is variable across materials

The geographic distribution of specimens for each material is variable (Fig. 1). Surface water ($N = 374$; 352 after
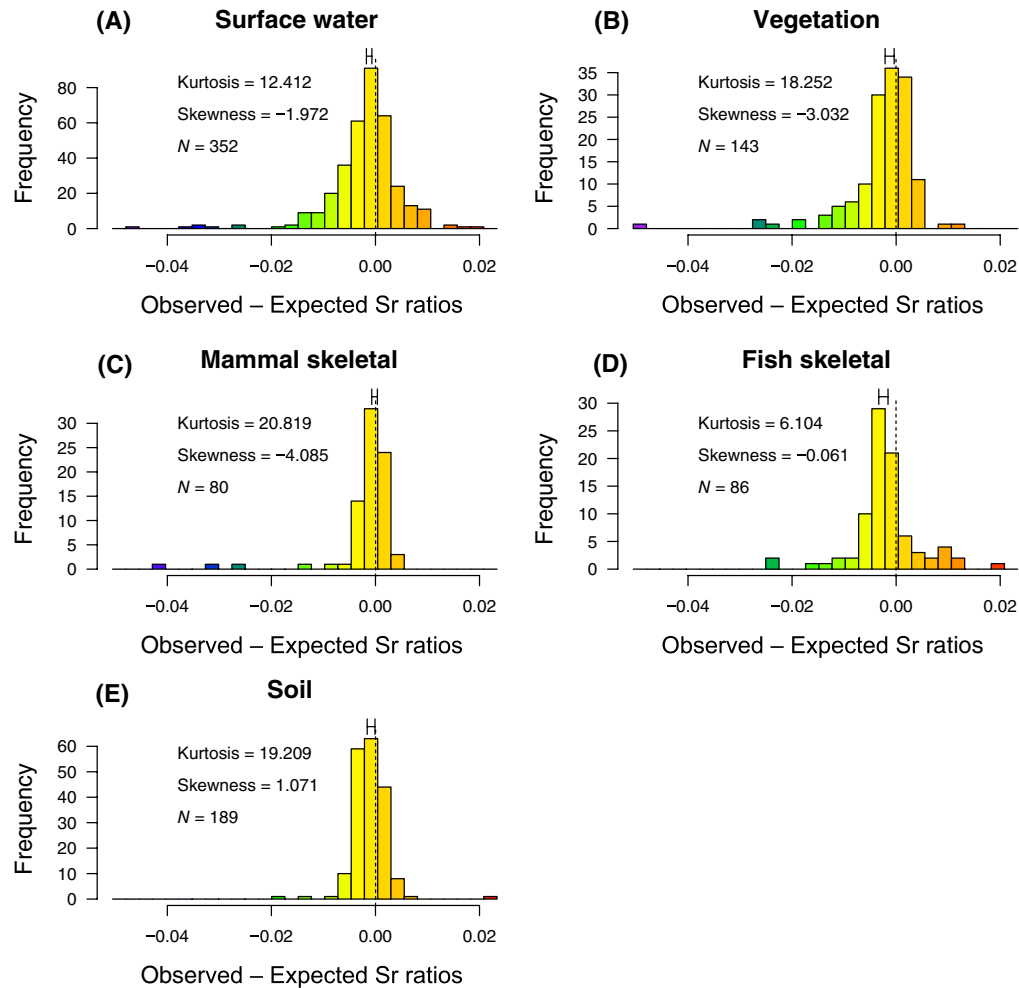
Fig. 2. Histograms of Sr offset between observed (empirical) and expected (modelled) data for (A) surface water, (B) vegetation, (C) mammal skeletal tissues, (D) fish skeletal tissues, and (E) soil. Data presented here have been geographically standardized to limit autocorrelation. Colours illustrate model overprediction and underprediction as in Fig. 1, dashed lines demark offsets of 0.0, and horizontal error bars are bootstrapped 95% confidence intervals around median estimates for each material. Kurtosis, skewness, and sample size (after geographic standardization) are also provided.

geographic standardization) and vegetation ($N = 426$; 143 after geographic standardization) have the broadest and densest coverage across the contiguous USA, with data from 32 and 28 states, respectively. Data from mammalian skeletal tissues ($N = 308$; 80 after geographic standardization) are available across 16 states, but are concentrated in the southeastern (Florida and Georgia) and midwestern USA (Kentucky, Ohio, Michigan, Indiana, and Wisconsin). The geographic coverage for soil ($N = 295$; 189 after geographic standardization) is the poorest. Soil data are only available from 10 states, which are split between restricted regions of the southwest (most data are from bordering regions of New Mexico, Arizona, Utah, and Colorado) and northeast. Data for fish skeletal tissues, which represent the largest number of individual specimens before geographic standardization ($N = 1290$; 86 after geographic standardization), exist for 18 states. These occur in pockets along the Atlantic coast, Colorado and Utah, and California and Washington.

(2) Measured Sr isotope ratios are generally well predicted by the local water model

After geographically standardizing the data, all frequency distributions show high kurtosis (Fig. 2), illustrating that Sr offset is generally focused around a single ratio (near 0.0 in all cases). However, only mammal skeletal tissues have bootstrapped median estimates that encompass 0.0 (Table 1). All other materials exhibit some tendency towards negative Sr offsets (i.e. model overprediction of Sr ratios). While the median offsets for surface waters, vegetation, fish skeletal tissues, and soils all approach 0.0 (Table 1), the magnitudes away from zero are all within the measurement limits of $^{87}Sr/^{86}Sr$ analyses, indicating they are securely non-zero. Skewness of Sr offset is negative for all materials with the exception of soil (Fig. 2). Skewness for soils and particularly fish osseous materials is low, highlighting a relatively even balance between overprediction and underprediction by the local water model. However, skewnesses for surface waters, mammal

Table 1. Calculated 95% bootstrapped confidence intervals for the median Sr offset of each material (after geographic standardization)

95% confidence interval	Surface water	Vegetation	Mammal skeletal	Fish skeletal	Soil
Upper	−0.0069	−0.00037	**0.00038**	−0.0017	−0.000092
Lower	−0.0017	−0.0021	**−0.00071**	−0.0033	−0.0016

Confidence intervals for mammals (in bold) surround 0.0, indicating very good agreement between modelled and empirical Sr data.

Table 2. Percent of data for each material predicted by the local water model within different degrees of model accuracy (modelled *versus* empirical Sr offset as a percent of empirical $^{87}Sr/^{86}Sr$ data)

Predictive accuracy	Surface water (%)	Vegetation (%)	Mammal skeletal (%)	Fish skeletal (%)	Soil (%)
Within 1%	80.7	84.6	93.8	81.4	97.9
Within 0.1%	17.3	16.1	35.0	10.5	29.6
Within 0.01%	2.3	0	7.5	0	5.8
Within 0.001%	0	0	2.5	0	0.5

Samples for which model predictions are within 0.001% mirror the precision of measurement error for empirical Sr ratio analysis.

skeletal tissues, and vegetation are higher, indicating a more extended negative tail (model overprediction) in these materials. Some materials are predicted more accurately than others (Table 2). 80.7–97.9% of the data (depending on material) are predicted within 1% of their empirical ratios, 10.5–35% of the data are predicted within 0.1%, 0–7.5% are predicted within 0.01%, and 0–2.5% are predicted within 0.001% (which is essentially equal to analytical measurement error). With the exception of soils, which have the highest percentage of data predicted within 1%, mammal skeletal tissues are the best predicted among the materials (Table 2).

(3) Increased Sr complexity leads to larger and more variable offset between modelled and measured Sr isotope ratios

Quantile regressions (Fig. 3, Table 3) show that there are generally significant positive relationships between the absolute value of Sr offset and regional Sr complexity. All quantiles reveal significant positive relationships for surface waters and vegetation. Significant positive relationships for fish skeletal tissues are only seen at the 75th quantile; while also positive, quantile regressions for soils and mammals are not significant. The median quantile for soils is negative and non-significant (Table 3).

As predicted, variance in Sr offset increases with increasing Sr complexity (Fig. 4 and Table S3). Increased variance is highly significant between samples from regions with low and medium Sr complexity for all materials (Table S3). Surface water and vegetation both have data from regions with high Sr complexity, and we do observe an increase in variance between regions of medium and high Sr complexity for both materials. However, we did not quantitatively compare Sr offset between medium and high complexity regions due to small sample sizes and low power ($N = 3$ for each material at high complexity).

(4) Mammal skeletal tissues are most accurately predicted, particularly at low Sr complexities

We find significant pairwise differences in magnitude of Sr offset among materials at low Sr complexities (Fig. 5, Table 4). Sr offset for mammal skeletal tissues is significantly smaller than the offset for all other materials except soils (Table 4). Sr offset for soils is significantly smaller than that for fish skeletal tissues and surface water (Fig. 5, Table 4). All other comparisons are either marginally significant (i.e. not maintained after Bonferroni correction; fish skeletal–vegetation, fish skeletal–surface water, soil–vegetation) or non-significant (surface water–vegetation, mammal skeletal–soil). Among materials in regions with medium Sr complexities, mammal skeletal tissues tend to remain more accurately predicted. Sr offset for mammals is significantly smaller than that for vegetation, and the difference in Sr offset between mammal skeletal material and surface water is marginally significant (Fig. 5, Table 4). Results are highly consistent when modelled Sr ratios are estimated using the specimens' exact georeferenced coordinates (Fig. S1).

In regions with low Sr complexities, Sr isotope ratios are significantly better predicted for large-mammal (>100 kg) than small-mammal skeletal tissues (Wilcoxon Test: $W = 489$, $P = 0.018$; Fig. 6). This pattern is consistent when using a 10 kg cut-off between 'small' and 'large' mammals. Conversely, $^{87}Sr/^{86}Sr$ for surface water in small watersheds (<1000 km^2) is better predicted than in large ones ($W = 3864$, $P \ll 0.01$). There are no differences in Sr offset between fish skeletal tissues from small *versus* large watersheds ($W = 294$, $P = 0.39$; Fig. 6) or between anadromous and freshwater fish ($W = 336$, $P = 0.21$). All of these results are consistent when modelled Sr is the point estimate at the georeferenced locality, or the median modelled Sr ratio within either a 5 or 10 km radius (Fig. S2).
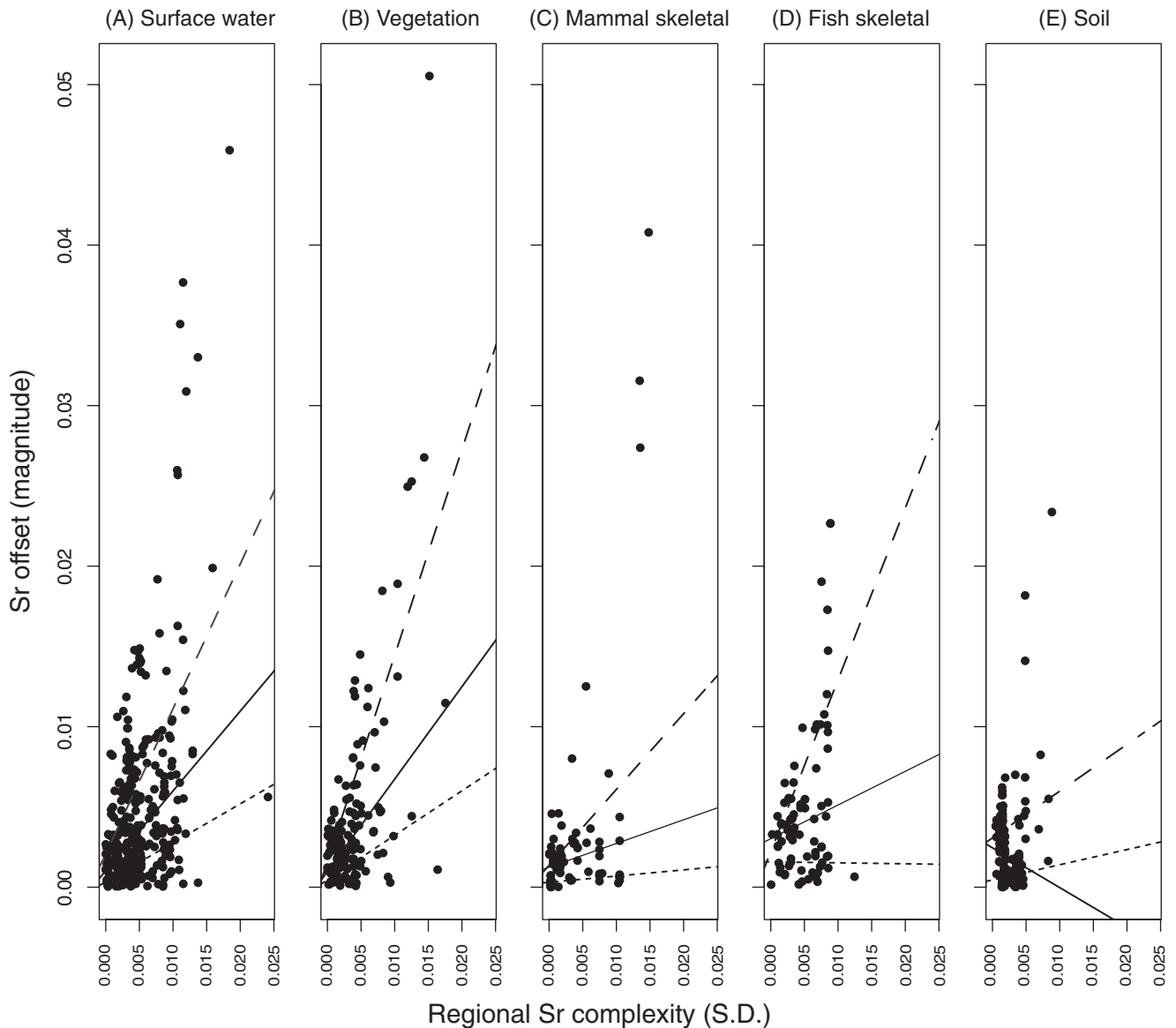
Fig. 3. 75th (dashed), 50th (solid) and 25th (dotted) quantile regressions for the magnitude (absolute value) of Sr offset against regional Sr complexity for (A) surface water, (B) vegetation, (C) mammal skeletal material, (D) fish skeletal material, and (E) soils. As predicted, the magnitude of Sr offset increases with regional Sr complexity in many cases. Strong separation between the 75th and 25th quantiles (particularly for surface waters, vegetation, and fish skeletal tissues) suggests that while offset between model estimates and empirical data can increase significantly with increasing complexity, the local water model can still predict Sr ratios with remarkable accuracy in complex regions. Regressions for mammal skeletal tissues and soils are non-significant. Quantile regression results for soil are less straightforward (e.g. crossed 25th and 50th quantile lines), and likely reflect the relatively poor spatial representation of soil data in the literature (limited or no empirical data were available from moderately or highly complex regions). Patterns for soil may also be complicated by temporal and spatial averaging of Sr across different depths and soil horizons.

V. DISCUSSION

(1) Model limitations

The predictive power of the Bataille & Bowen (2012) model is compromised by its limitations in formulation and capacity to incorporate all potential sources of Sr (see Appendix S1). A limitation particularly relevant to this study is the absence of an independent prediction of $^{87}Sr/^{86}Sr$ for

siliciclastic sediments. Bataille & Bowen (2012) applied the same average approach to predict the $^{87}Sr/^{86}Sr$ ratio in igneous rocks and siliciclastic sediments, effectively assuming that for siliciclastics, deposition age represents the actual age of the rock. Consequently, the Bataille & Bowen (2012) local water model does not capture the isotopic variability in siliciclastic sediments associated with their parent rock and recycling history. To predict the $^{87}Sr/^{86}Sr$ for local waters, Bataille & Bowen (2012) used a bedrock model

Table 3. Parameter estimates and bootstrapped significance values for 25th, 50th, and 75th quantile regressions for Sr offset *versus* regional Sr complexity for each material

Substrate	25th quantile Slope ± S.E.	50th quantile Slope ± S.E.	75th quantile Slope ± S.E.
Surface water	**0.24** ± 0.049**	**0.50** ± 0.099**	**0.90** ± 0.17**
Vegetation	**0.28* ± 0.13**	**0.58* ± 0.24**	**1.28** ± 0.37**
Soil	0.095 ± 0.12	−0.25 ± 0.22	0.29 ± 0.33
Fish skeletal	−0.0060 ± 0.12	0.21 ± 0.32	**1.07** ± 0.24**
Mammal skeletal	0.039 ± 0.028	0.15 ± 0.16	0.47 ± 0.76

Significant relationships are in bold. Single and double asterisks denote significance levels of $P < 0.05$ and $P < 0.01$, respectively.

that gives a relatively uniform $^{87}Sr/^{86}Sr$ prediction for siliciclastic sediments centered around 0.7115. This ratio will overpredict $^{87}Sr/^{86}Sr$ in regions where siliciclastic sediments have young and mafic parents (e.g. American Cordillera) and underpredict $^{87}Sr/^{86}Sr$ in regions where siliciclastic sediments have been recycled numerous times (e.g. Appalachian orogenic belt). This challenge leads to geographic trends in the prediction accuracy of the local water model. Combined with geographic biases among materials in our compiled data set, such model limitations may explain part of the difference in predictive accuracy observed among materials.

Bataille *et al.* (2014) partially resolved this model formulation issue by developing an independent siliciclastic sediment sub-model that leverages terrane age and lithology to account for the control of parent rock on the $^{87}Sr/^{86}Sr$ variability of siliciclastic sediments in Alaska, USA. Their modelling efforts also included a better representation of local variability in $^{87}Sr/^{86}Sr$ and better prediction uncertainty. Combined, such improvements enhanced the predictive power of the models, with 97 and 59% of empirical $^{87}Sr/^{86}Sr$ ratios of regional water samples predicted within 1 and 0.1% of predicted ratios, respectively (Bataille *et al.,* 2014). A similarly improved version of the model is in development for the contiguous USA.

## (2) Spatial sampling of empirical data is highly variable

Despite decades of research, the spatial coverage of available empirical data is remarkably poor and geographically biased; for most materials, samples have only been analysed from a handful of states, and geographic coverage differs among materials (Fig. 1). Consequently, there are large swathes of the contiguous USA with no empirical data to validate model predictions or make comparisons among materials. In particular there are relatively few samples taken from regions with high Sr complexity (i.e. regions with high Sr variability). Some materials have been geographically sampled more thoroughly than others, but limited overlap among materials prevents spatially standardized comparisons among them. This is a clear knowledge gap that future empirical studies should address.

(3) Variability in modelled *versus* measured Sr isotope ratios is larger in more complex regions

Our results support expectations that (*i*) the local water model predicts $^{87}Sr/^{86}Sr$ ratios better in more geologically homogenous areas, and (*ii*) variance in offset between observed (empirical) and expected (modelled) Sr isotope ratios is larger in more complex regions. Model accuracy is good at low Sr complexities, but variability in Sr offset (and offset itself in the case of surface waters and vegetation; Fig. 3) increases with Sr complexity. This finding is a meaningful caveat for those interested in studying geologically complex regions. Whereas many provenance research questions benefit from high Sr complexity, model accuracy in complex regions may not be sufficient to answer these questions and an empirical approach will likely be needed.

Interestingly, mammal skeletal tissues and soils show no significant linear relationships between Sr offset and geologic complexity, suggesting invariance in model fit with respect to underlying regional Sr variability. The soil sample is particularly depauperate in specimens from regions with greater Sr complexity (which may contribute to the lack of relationship). The mammal sample has better coverage (with subequal sampling of both low and medium complexity regions), yet still does not exhibit strong relationships between Sr offset and complexity.

Soils are a somewhat anomalous data set in several ways. First, sample sizes are very low for soils from regions with moderate or high Sr complexity. Second, there is variability in the age and depth of the soil samples that we included in our compiled data set. Most (but not all) authors reported sample depth. However, it is not clear how much time is integrated within particular samples. Soil formation processes and timing can vary considerably among regions with differing climate, topography, and vegetation cover. Therefore, spatial patterns for soil may be complicated by temporal averaging of Sr across different soil horizons. Third, soil samples can vary considerably in their clay, sand, and organic content. There does not appear to be a consensus in the literature as to which portion of soils should be analysed. Among the various sources we included in our analyses, authors reported analysing Sr eluted from various leachates including ammonium acetate (Bataille & Bowen, 2012; Bataille *et al.,* 2012, 2014), acetic acid (Miller *et al.,* 1993;
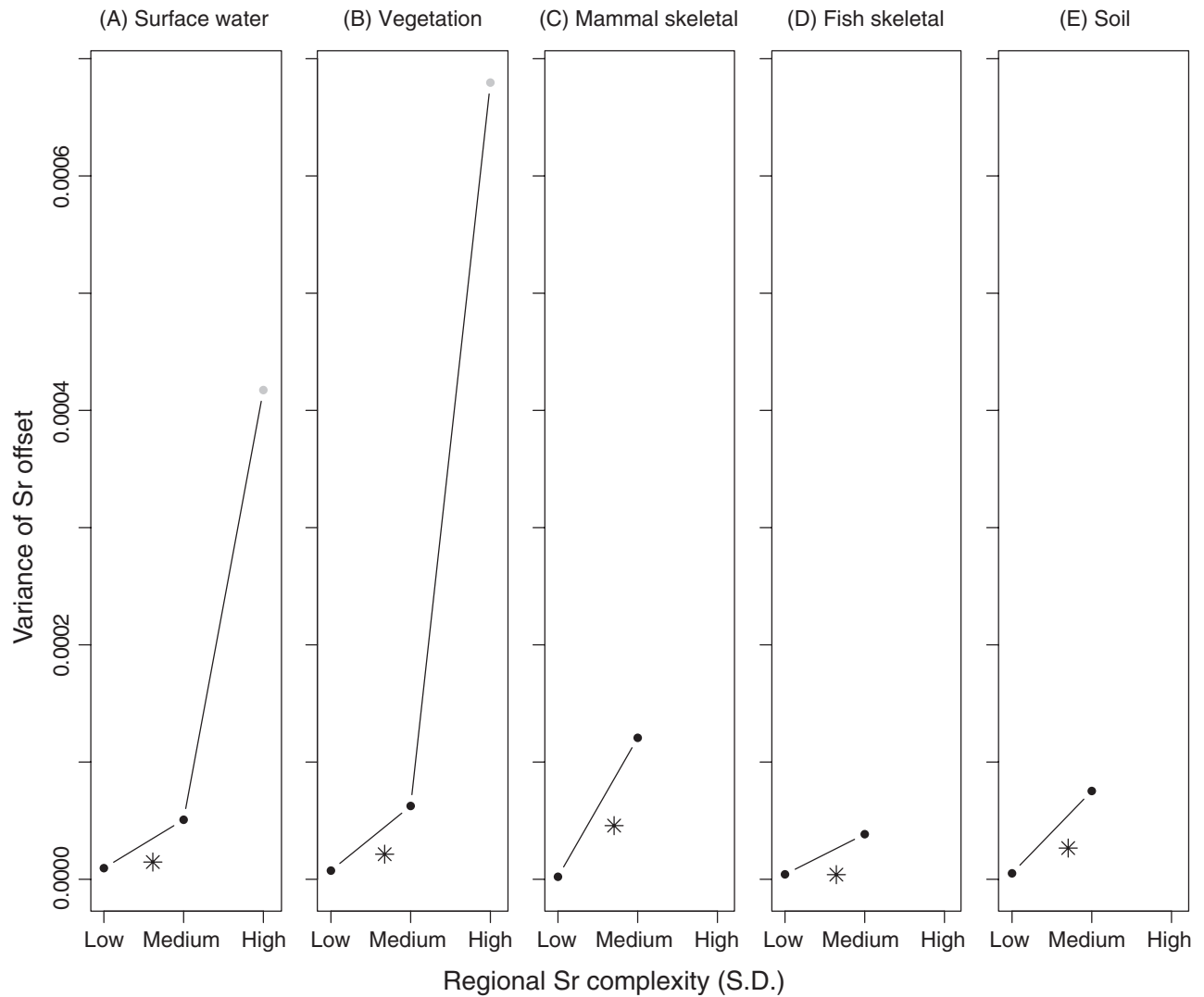
Fig. 4. Variance in the magnitude of Sr offsets among regions with low, medium, and high regional Sr complexity for (A) surface water, (B) vegetation, (C) mammal skeletal material, (D) fish skeletal material, and (E) soils. Grey dots represent groups from high-complexity regions with less than five specimens. Asterisks denote significant differences in variance between complexity bins. Variance increases significantly between low and medium Sr complexity for all five materials. There are qualitative increases in variance between medium and high Sr complexity regions for surface water and vegetation (sample sizes from complex regions are too small for statistical analysis).

Cooke *et al.,* 2003; Reynolds *et al.,* 2012), hydrochloric acid (Benson *et al.,* 2006, 2009; Benson, 2010), or hydrofluoric acid (Xin, 1993). Nevertheless, even with the added complications of the soil data set, including profound heterogeneity in composition and preparation techniques, the power of the Bataille & Bowen (2012) local water model to predict soils is not dramatically altered compared to other materials.

Predictive quality of the model for mammal skeletal tissues across our data set appears less influenced by differences in underlying geology than other materials (Fig. 3). While this may partially reflect geographic bias in the mammal data set (see Section V.4), together with lower overall Sr offset, this may also be an indication that mammal tissues and the model integrate Sr in similar ways (or at least the end result

is more equitable than for other materials), making mammal tissues particularly well suited for the model. However, depending on home range size, frequency of movement, and randomness of movements within a region, mobile mammals may not fully capture local Sr variability. Characterizing local ^{87}Sr/^{86}Sr ratios in more complex regions will likely require samples from less-mobile (or non-mobile) materials and larger sample sizes. Recognition of these trends will be particularly important when comparing data among regions.

(4) Modelled Sr isotope ratios are informative predictors of empirical data

Our results indicate that there are differences in how well Sr isotope ratios are predicted among materials
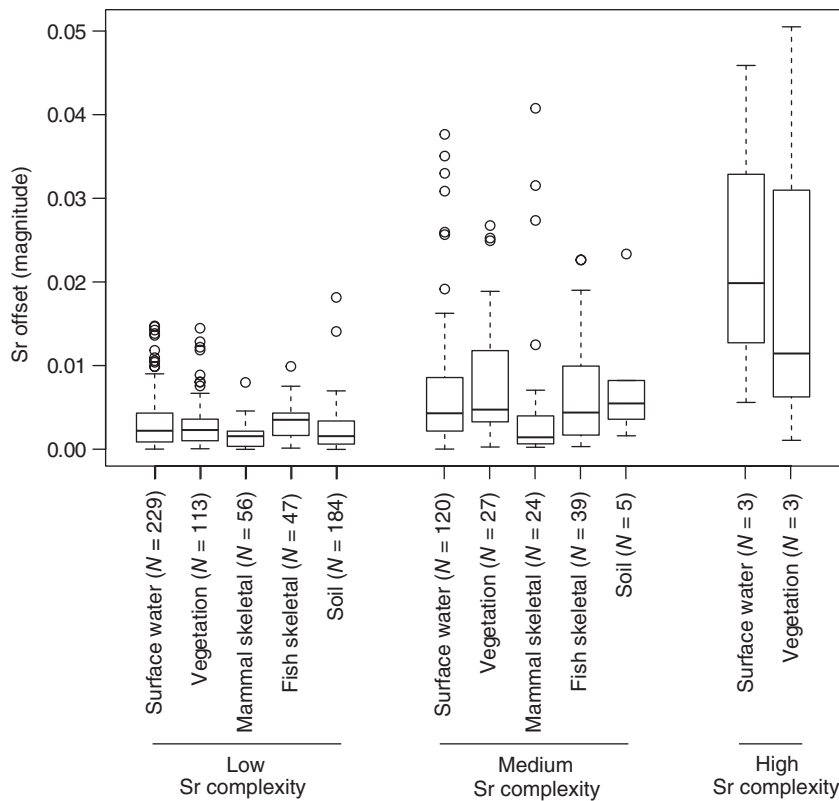
Fig. 5. Boxplots comparing the magnitude of Sr offsets among materials in regions of low, medium, and high Sr complexity across the contiguous USA. In regions of low Sr complexity, Sr offset for mammal skeletal tissues is significantly lower than all other materials except soil (Table 4). Soils have significantly lower offset than fish skeletal tissues. In regions of medium Sr complexity, the only comparisons with significant or marginally significant differences include mammal skeletal tissues, which continue to show relatively low Sr offset (e.g. they are particularly well-predicted; Table 4). In regions with high Sr complexity, there are only data for surface water and vegetation, and samples sizes are too small for statistical comparisons.

(Figs 2 and 5), providing partial support for our hypothesis that Sr ratios of animal tissues and water are better predicted than soils and vegetation. As expected, the offset between modelled and measured Sr ratios is slightly larger for vegetation than mammal skeletal tissues at low geologic complexities (Fig. 5, Table 4). However, somewhat unexpectedly, there is no difference between soils and mammal skeletal tissues, and mammal skeletal tissues are more accurately estimated than all other materials, including surface water and fish skeletal tissues.

As discussed above, soils are a rather anomalous data set. There are several possible reasons why the local water model predicts $^{87}Sr/^{86}Sr$ for mammalian skeletal tissues with higher confidence than other materials at low Sr complexities, including surface waters for which the model was built. First, whereas water samples come from a diversity of localities spanning the entire continent, the majority of the mammals included in our dataset come from Florida and portions of the central USA that are dominated by marine sediments (Fig. 1). Carbonate-dominated landscapes are less complicated for the local water model to predict. Additionally, $^{87}Sr/^{86}Sr$ for the siliciclastic sediments of the Mississippi basin are relatively similar to the average ratio that Bataille & Bowen (2012) used for this lithology. Consequently, the model might be expected

to perform especially well in these regions. However, we note that not all Sr ratios for mammals are well predicted in these regions (dark green point in Florida of Fig. 1). Moreover, mammals from other regions (e.g. southwest USA; Fig. 1) are also well predicted. Therefore, geographic bias cannot be solely responsible for the fit between modelled and measured Sr isotope data for mammals.

Second, temporal variability in the isotopic composition of surface waters may affect the match between modelled and empirical data. The isotopic composition of surface water can shift seasonally or from year to year due to variability in precipitation sources, weathering, and run-off (Graustein & Armstrong, 1983; Gosz & Moore, 1989), and is of particular concern in areas with glacial or permafrost cover (e.g. Semhi *et al.,* 2000; Keller, Blum & Kling, 2010; Hindshaw *et al.,* 2011). Although temporal variability in Sr isotope ratios is inconsistent and relatively small compared to spatial variability (reviewed in Brennan *et al.,* 2015), because the vast majority of water localities included in our data set were only sampled once, it is possible that measured Sr isotope ratios for these samples are seasonally biased and may vary slightly from the predicted average Sr ratio for their respective localities. While temporal variability in Sr can also affect fish otoliths or tooth enamel, this is less likely
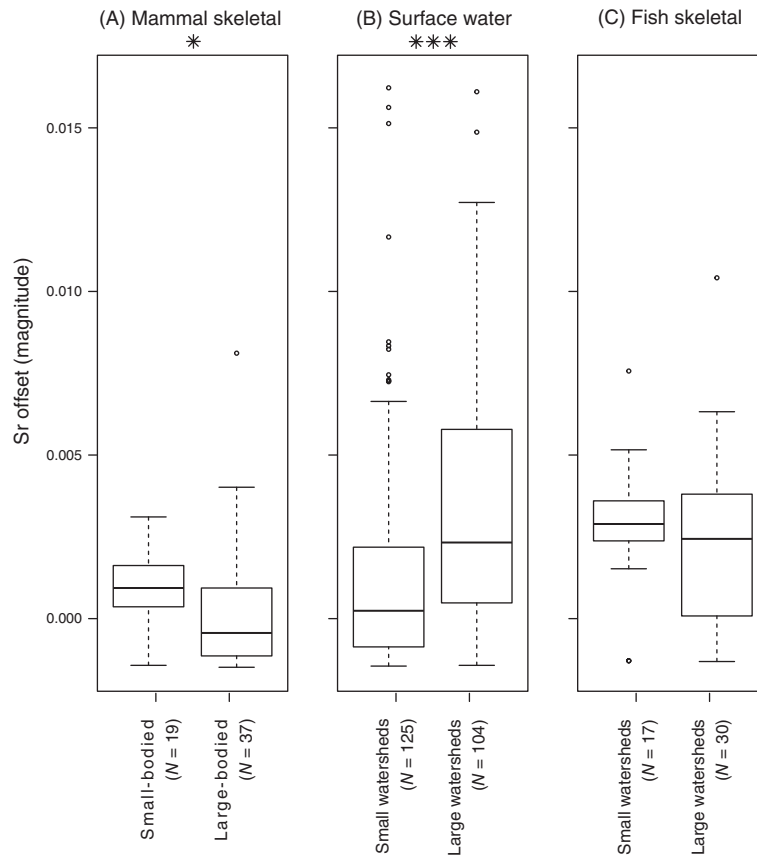
Fig. 6. Boxplots comparing magnitudes of Sr offsets for small- ($<100\,kg$) and large-mammal skeletal tissues, and surface water and fish skeletal tissues from small ($<1000\,km^2$) and large watersheds in regions with low Sr complexity. Modelled Sr isotope ratios are taken as the median within a $5\,km$ radius of each specimen's georeferenced locality using the Bataille & Bowen (2012) local water model. Single and triple asterisks denote significance levels of $P < 0.05$ and $P \ll 0.01$, respectively.

Table 4. Results from pairwise Wilcoxon tests comparing Sr offsets among materials for regions with (a) low Sr complexity, and (b) medium Sr complexity

Material	Vegetation	Mammal skeletal	Fish skeletal	Soil
(a) Low Sr complexities				
Surface water	12639 (0.73)	**4379 (0.00024)**	6364 (0.049)	**17461 (0.0028)**
Vegetation		**2106 (0.00041)**	3344 (0.010)	8814 (0.028)
Mammal skeletal			**2046 (0.0000014)**	4316.5 (0.066)
Fish skeletal				**6016 (0.000035)**
(b) Medium Sr complexities				
Surface water	1884 (0.19)	959 (0.010)	2263 (0.76)	344 (0.58)
Vegetation		**181 (0.0064)**	428 (0.20)	67 (1.0)
Mammal skeletal			600 (0.062)	31 (0.10)
Fish skeletal				77 (0.47)

Significance estimates (*P* values) are presented in parentheses. Comparisons that maintain significance after Bonferroni correction are in bold. Comparisons that are marginally significant (i.e. those that do not pass Bonferroni correction) are underlined.

to be a concern for our data set, which only includes bulk Sr isotope ratios representing multiple growing seasons for these materials.

Third, it is possible that the level of agreement between observed and expected Sr isotope ratios for mammals may reflect the manner in which mammals integrate spatial information. Perhaps unsurprisingly, we find that larger mammals, which can integrate Sr across broader spatial scales, have lower Sr offsets between modelled and empirical ratios than smaller mammals (Fig. 6). Model accuracy is also scale dependent for surface water (i.e. significant differences in Sr offsets for large *versus* small watersheds; Fig. 6), but not for fish. Better-predicted Sr isotope ratios for large mammals and small watersheds suggests that the manner

(or the ultimate underlying spatial resolution) in which these materials integrate Sr is most similar to the local water model. Larger Sr offsets for small mammals (and immobile materials such as plants and soils) likely reflect local heterogeneity that is not captured by the model, while larger Sr offsets for large watersheds may result from greater spatial averaging of these water bodies (e.g. the Mississippi River) compared to the model. It is notable that these results differ from those of Hegg *et al.* (2013*b*), who found that the power of their model to predict ^{87}Sr/^{86}Sr for river water using bedrock composition decreased for watersheds <1000 km^2 in the Pacific Northwest. Discrepancies could indicate particularities of the Pacific Northwest. Importantly, the smaller watersheds explored by Hegg *et al.* (2013*b*) also had lower geologic complexity than the larger watershed for which their model was constructed; thus, there may have been conflation of two important drivers of model quality. By expanding the geographic scope of our analyses, we are able to differentiate better the impacts of these two factors. We also suspect that the addition of differential weathering and bedrock ages increases the predictive accuracy of the Bataille & Bowen (2012) local water model for smaller watersheds.

While we recover differences in model predictive power among materials, it is possible that the local water model is simply inappropriate for estimating Sr isotope ratios of large water bodies or highly mobile fish. Bataille & Bowen (2012) produced an additional model called the 'catchment water model', which assimilates contributions of Sr from different rock lithologies and ages within a water catchment. Although it is a more complicated comparison, we can test if using the catchment water model to predict ^{87}Sr/^{86}Sr for surface water and fish (and using the local water model to predict vegetation, mammal skeletal tissues, and soil) (*i*) improves predictions for either surface waters or fish, and (*ii*) changes the relative predictive accuracy among the five materials in our database. Using this analytical design, surface waters are better predicted by the catchment water model than the local water model for regions with low Sr complexity (water: $W = 52550.5$, $P = 0.0040$), but fish skeletal tissues are not ($W = 3098$, $P = 0.066$). Sr offset for mammal skeletal tissues is still lower than surface water (although marginally so after Bonferroni correction; $W = 5861$, $P = 0.039$) and fish ($W = 2021$, $P = 0.0011$; Fig. S3; Table S4). Additionally, surface water from small rivers is still better predicted than large ones ($W = 4880$, $P \ll 0.01$), and Sr offset for fish skeletal tissues continues to be indistinguishable between small and large rivers ($W = 293$, $P = 0.81$). Thus, while the catchment water model improves predictions for surface waters, there are still limitations in the degree to which it can predict Sr isotope ratios.

There are several possible reasons why mammal skeletal tissues are consistently predicted more accurately than those of fish. First, whereas the primary Sr source for mammals (food) is typically locally derived, the primary Sr source for fish (water) may come from far upstream. Second, most of the sampled mammals likely incorporated Sr from smaller geographic areas than fish. The majority of fish that have been studied using Sr are highly mobile species such as salmon and shad (see Table S2). These fish are able to move upstream and among watersheds, which may result in a poor fit between modelled and predicted Sr for fish even in small watersheds. The consistent observation that there are no differences in Sr offset between anadromous and freshwater fish at low complexities using either the local water model ($W = 336$, $P = 0.21$) or the catchment water model ($W = 348$, $P = 0.97$), suggests that all of the fish taxa included in our analysis are generally mobile enough to decouple from model predictions (Brennan *et al.*, 2015).

Overall, our results suggest that the local water model developed by Bataille & Bowen (2012) provides informative estimates of local ^{87}Sr/^{86}Sr for all five materials (Fig. 2, Table 1). After geographic standardization, 80.7–97.9% and 10.5–35.0% of ^{87}Sr/^{86}Sr ratios were predicted within 1 and 0.1% of empirical ratios, respectively (Table 2). Particularly good agreement between predicted and measured Sr isotope ratios for large-bodied mammal skeletal tissues and small watersheds suggests that at least some research questions (e.g. exploration of migratory patterns in mammals) may be largely addressed without the need for additional exhaustive empirical sampling for local to regional background Sr ratios. However, Sr provenance work frequently relies on differences at the fourth decimal place to distinguish localities or identify mobile individuals. Thus, even 0.1% predictive accuracy may not be sufficient for some ecological applications.

VI. CONCLUSIONS

(1) Strontium isotope analysis can provide unique ecological and biogeographical information about modern and ancient organisms. Existing empirical Sr data for biologically relevant materials in the contiguous USA are spatially heterogeneous, and in most cases, geographically limited. Although we cannot evaluate the comparability of Sr isotope ratios among biologically relevant materials within a particular locality, overall our data synthesis indicates that the evaluated biologically relevant materials may differ slightly in the degree to which they record local Sr variability. Determination of which material is 'best' at approximating local Sr will depend on the question of interest as well as local to regional Sr variability.

(2) The Bataille & Bowen (2012) local water model appears to be exceptionally effective at predicting Sr isotope ratios for mammal skeletal tissues (particularly large-bodied mammals) and surface waters (particularly small watersheds).

(3) Modelled Sr data from surface water, vegetation, ossified animal tissues, and soils generally appear sufficient for establishing a comparative framework and evaluating the ecological significance of empirical samples.

(4) Although the local water model provides a readily available source of background data for predicting ^{87}Sr/^{86}Sr ratios in places where empirical data are lacking, its predictive accuracy may not be sufficient for some research questions.

VII. ACKNOWLEDGEMENTS

VIII. REFERENCES

References marked with asterisk have been cited within the supporting information.

ÅBERG, G. (1995). The use of natural strontium isotopes as tracers in environmental studies. *Water, Air, and Soil Pollution* **79**, 309–322.

BACON, C. R., WEBER, P. K., LARSEN, K. A., REISENBICHLER, R., FITZPATRICK, J. A. & WOODEN, J. L. (2004). Migration and rearing histories of chinook salmon (*Oncorhynchus tshawytscha*) determined by ion microprobe Sr isotope and Sr/Ca transects of otoliths. *Canadian Journal of Fisheries and Aquatic Sciences* **61**, 2425–2439.

BANNER, J. L. (2004). Radiogenic isotopes: systematics and applications to earth surface processes and chemical stratigraphy. *Earth-Science Reviews* **65**, 141–194.

BARNETT-JOHNSON, R., PEARSON, T. E., RAMOS, F. C., GRIMES, C. B. & MACFARLANE, R. B. (2008). Tracking natal origins of salmon using isotopes, otoliths, and landscape geology. *Limnology and Oceanography* **53**, 1633–1642.

BATAILLE, C. P. & BOWEN, G. J. (2012). Mapping $^{87}Sr/^{86}Sr$ variations in bedrock and water for large scale provenance studies. *Chemical Geology* **304-305**, 39–52.

BATAILLE, C. P., BRENNAN, S. R., HARTMANN, J., MOOSDORF, N., WOOLLER, M. J. & BOWEN, G. J. (2014). A geostatistical framework for predicting variability in strontium concentrations and isotope ratios in Alaskan rivers. *Chemical Geology* **389**, 1–15.

BATAILLE, C. P., LAFOON, J. & BOWEN, G. J. (2012). Mapping multiple source effects on the strontium isotopic signatures of ecosystems from the circum-Caribbean region. *Ecosphere* **3**, 1–24.

BAUMANN, E. & CROWLEY, B. E. (2015). Stable isotopes reveal ecological differences amongst now-extinct proboscideans from the Cincinnati region, USA. *Boreas* **44**, 240–254.

BEARD, B. L. & JOHNSON, C. M. (2000). Strontium isotope composition of skeletal material can determine the birth place and geographic mobility of humans and animals. *Journal of Forensic Sciences* **45**, 1049–1061.

BEEHR, D. E. (2011). *Investigation of middle woodland population movement in the midwestern United States using strontium isotopes*. Dissertation Thesis: University of Illinois at Urbana-Champaign, Urbana.

BENSON, L. V. (2010). Who provided maize to Chaco Canyon after the mid-12th-century drought? *Journal of Archaeological Science* **37**, 621–629.

BENSON, L. V., STEIN, J. R. & TAYLOR, H. E. (2009). Possible sources of archaeological maize found in Chaco Canyon and Aztec Ruin, New Mexico. *Journal of Archaeological Science* **36**, 387–407.

BENSON, L. V., STEIN, J., TAYLOR, H., FRIEDMAN, R. & WINDES, T. C. (2006). The agricultural productivity of Chaco Canyon and the source(s) of Pre-Hispanic maize found in Pueblo Bonito. In *Histories of Maize: Multidisciplinary Approaches to the Prehistory, Linguistics, Biogeography, Domestication, and Evolution of Maize* (eds J. STALLER, R. TYKOT and B. BENZ), pp. 289–314. Left Coast Press, Walnut Creek.

BENTLEY, R. A. (2006). Strontium isotopes from the earth to the archaeological skeleton: a review. *Journal of Archaeological Method and Theory* **13**, 135–187.

BERN, C. R., TOWNSEND, A. R. & FARMER, G. L. (2005). Unexpected dominance of parent-material strontium in a tropical forest on highly weathered soil. *Ecology* **86**, 626–632.

BLUM, J. D., EREL, Y. & BROWN, K. (1993). $^{87}Sr/^{86}Sr$ ratios of Sierra Nevada stream waters: implications for relative mineral weathering rates. *Geochimica et Cosmochimica Acta* **58**, 5019–5025.

BLUM, J. D., TALIAFERRO, E. H. & HOLMES, R. T. (2001). Determining the sources of calcium for migratory songbirds using stable strontium isotopes. *Oecologia* **126**, 569–574.

BLUM, J. D., TALIAFERRO, E. H., WEISSE, M. T. & HOLMES, R. T. (2000). Changes in Sr/Ca, Ba/Ca and $^{87}Sr/^{86}Sr$ ratios between trophic levels in two forest ecosystems in the northeastern USA. *Biogeochemistry* **49**, 87–101.

BRASS, G. W. (1976). The variation of the marine $^{87}Sr/^{86}Sr$ ratio during Phanerozoic time: interpretation using a flux model. *Geochimica et Cosmochimica Acta* **40**, 721–730.

BRENNAN, S. R., FERNANDEZ, D. P., ZIMMERMAN, C. E., CERLING, T. E., BROWN, R. J. & WOOLLER, M. J. (2015). Strontium isotopes in otoliths of a non-migratory fish (slimy sculpin): implications for provenance studies. *Geochimica et Cosmochimica Acta* **149**, 32–45.

BRITTON, K. H. (2009). *Multi-isotope analysis and the reconstruction of prey species palaeomigrations and palaeoecology*. Dissertation Thesis: Durham University, Durham.

CAPO, R. C., STEWART, B. W. & CHADWICK, O. A. (1998). Strontium isotopes as tracers of ecosystem processes: theory and methods. *Geoderma* **82**, 197–225.

CHADWICK, O. A., DERRY, L.A., BERN, C.R., VITOUSEK, P.M.,. (2009). Changing sources of strontium to soils and ecosystems across the Hawaiian Islands. *Chemical Geology* **267**, 64-76.

CHAMBERLAIN, C. P., BLUM, J. D., HOLMES, R. T., FENG, X., SHERRY, T. W. & GRAVES, G. R. (1997). The use of isotope tracers for identifying populations of migratory birds. *Oecologia* **109**, 132–141.

CHESSON, L. A., TIPPLE, B. J., MACKEY, G. N., HYNEK, S. A., FERNANDEZ, D. P. & EHLERINGER, J. R. (2012). Strontium isotopes in tap water from the coterminous USA. *Ecosphere* **3**, 67.

CHRISTIAN, L. N., BANNER, J. L. & MACK, L. E. (2011). Sr isotopes as tracers of anthropogenic influences on stream water in the Austin, Texas, area. *Chemical Geology* **282**, 84–97.

COOKE, M. J., STERN, L. A., BANNER, J. L., MACK, L. E., STAFFORD, T. W. Jr. & TOOMEY, R. S. III (2003). Precise timing and rate of massive late Quaternary soil denudation. *Geology* **31**, 853–856.

COPELAND, S. R., SPONHEIMER, M., DE RUITER, D. J., LEE-THORP, J. A., CODRON, D., LE ROUX, P. J., GRIMES, V. & RICHARDS, M. P. (2012). Strontium isotope evidence for landscape use by early hominins. *Nature* **474**, 76–78.

CRITTENDEN, R. G., ANDREW, A. S., LEFOURNOUR, M., YOUNG, M. D., MIDDLETON, H. & STOCKMANN, R. (2007). Determining the geographic origin of milk in Australasia using multi-element stable isotope ratio analysis. *International Dairy Journal* **17**, 421–428.

CURTIS, J. B. & STUEBER, A. M. (1973). $^{87}Sr/^{86}Sr$ ratios and total strontium concentrations in surface waters of the Scioto River Drainage Basin, Ohio. *The Ohio Journal of Science* **73**, 166–175.

DEPAOLO, D. J. & INGRAM, B. L. (1985). High-resolution stratigraphy with strontium isotopes. *Science* **227**, 938–941.

DOUGLAS, T. A., BLUM, J. D., GUO, L. D., KELLER, K. & GLEASON, J. D. (2013). Hydrogeochemistry of seasonal flow regimes in the Chena River, a subarctic watershed draining discontinuous permafrost in interior Alaska (USA). *Chemical Geology* **335**, 48–62.

DOUGLAS, T. A., CHAMBERLAIN, C. P. & BLUM, J. D. (2002). Land use and geologic controls on the major elemental and isotopic ($\delta^{15}N$ and $^{87}Sr/^{86}Sr$) geochemistry of the Connecticut River watershed, USA. *Chemical Geology* **189**, 19–34.

EASTIN, R. & FAURE, G. (1970). Seasonal variation of the solute content and the $^{87}Sr/^{86}Sr$ ratio of the olentangy and Scioto rivers at Columbus, Ohio. *The Ohio Journal of Science* **70**, 170–179.

EERKENS, J. W., BARFOD, G. H., JORGENSON, G. A. & PESKE, C. (2014). Tracing the mobility of individuals using stable isotope signatures in biological tissues: "locals" and "non-locals" in an ancient case of violent death from Central California. *Journal of Archaeological Science* **41**, 474–481.

ENGLISH, N. B., BETANCOURT, J. L., DEAN, J. S. & QUADE, J. (2001). Strontium isotopes reveal distant sources of architectural timber in Chaco Canyon, New Mexico. *Proceedings of the National Academy of Sciences* **98**, 11891–11896.

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

EVANS, J. A., CHENERY, C. A. & FITZPATRICK, A. P. (2006). Bronze age childhood migration of individuals near Stonehenge, revealed by strontium and oxygen isotope tooth enamel analysis. *Archaeometry* **48**, 309–321.

EZZO, J. A., JOHNSON, C. M. & PRICE, T. D. (1997). Analytical perspectives on prehistoric migration: a case study from east-central Arizona. *Journal of Archaeological Science* **24**, 447–466.

FERANEC, R. S., HADLY, E. A. & PAYTAN, A. (2007). Determining landscape use of Holocene mammals using strontium isotopes. *Oecologia* **153**, 943–950.

FISHER, R. S. & STUEBER, A. M. (1976). Strontium isotopes in selected streams within the Susquehanna River Basin. *Water Resources Research* **12**, 1061–1068.

FOX, J. & WEISBERG, S. (2010). *An R Companion to Applied Regression*. Second Edition. Sage, Thousand Oaks.

FREI, K. M. & FREI, R. (2011). The geographic distribution of strontium isotopes in Danish surface waters – A base for provenance studies in archaeology, hydrology and agriculture. *Applied Geochemistry* **26**, 326–340.

GIBSON-REINEMER, D. K., JOHNSON, B. M., MARTINEZ, P. J., WINKELMAN, D. L., KOENIG, A. E. & WOODHEAD, J. D. (2009). Elemental signatures in otoliths of hatchery rainbow trout (*Oncorhynchus mykiss*): distinctiveness and utility for detecting origins and movement. *Canadian Journal of Fisheries and Aquatic Sciences* **66**, 513–524.

GOLDSTEIN, S. J. & JACOBSEN, S. B. (1987). The Nd and Sr isotopic systematics of river-water dissolved material: implications for the sources of Nd and Sr in seawater. *Chemical Geology* **66**, 245–272.

GOSZ, J. R. & MOORE, D. I. (1989). Strontium isotope studies of atmospheric inputs to forestsed watersheds in New Mexico. *Biogeochemistry* **8**, 115–134.

GRAUSTEIN, W. C. & ARMSTRONG, R. L. (1983). The use of strontium-87/strontium-86 ratios to measure atmospheric transport into forested watersheds. *Science* **219**, 289–292.

GROSS, E. L., PATCHETT, J., DALLEGGE, T. A. & SPENCER, J. E. (2001). The Colorado River system and Neogene sedimentary formations along its course: apparent Sr isotopic connections. *The Journal of Geology* **109**, 449–461.

HAMANN, E. J. & KENNEDY, B. P. (2012). Juvenile dispersal affects straying behaviors of adults in a migratory population. *Ecology* **93**, 733–740.

HART, S. R. & TILTON, G. R. (1966). The isotope geochemistry of strontium and lead in Lake Superior sediments and water. In *The Earth Beneath the Continents: A Volume of Geophysical Studies in Honor of Merle A. Tuve* (eds J. S. STEINHART and T. J. SMITH), pp. 127–137. American Geophysical Union, Washington.

HEDMAN, K. M., CURRY, B. B., JOHNSON, T. M., FULLAGAR, P. D. & EMERSON, T. E. (2009). Variation in strontium isotope ratios of archaeological fauna in the Midwestern United States: a preliminary study. *Journal of Archaeological Science* **36**, 64–73.

HEGG, J. C., KENNEDY, B. P., CHITTARO, P. M. & ZABEL, R. W. (2013a). Spatial structuring of an evolving life-history strategy under altered environmental conditions. *Oecologia* **172**, 1017–1029.

HEGG, J. C., KENNEDY, B. P. & FREMIER, A. K. (2013b). Predicting strontium isotope variation and fish location with bedrock geology: understanding the effects of geologic heterogeneity. *Chemical Geology* **360-361**, 89–98.

HIJMANS, R. J. (2015). raster: geographic data analysis and modeling. R package, version 2.3-24. Available at http://CRAN.R-project.org/package=raster Accessed 10.08.2015.

HINDSHAW, R. S., TIPPER, E. T., REYNOLDS, B. C., LEMARCHAND, E., WIEDERHOLD, J. G., MAGNUSSON, J., BERNASCONI, S. M., KRETZSCHMAR, R. & BOURDON, B. (2011). Hydrological control of stream water chemistry in a glacial catchment (Damma Glacier, Switzerland). *Chemical Geology* **285**, 215–230.

HOBSON, K. A., BARNETT-JOHNSON, R. & CERLING, T. (2010). Using isoscapes to track animal migration. In *Isoscapes: Understanding Movement, Pattern, and Process on Earth through Isotope Mapping* (eds J. B. WEST, G. J. BOWEN, T. E. DAWSON and K. P. TU), pp. 273–298. Springer Science + Business Media B.V., New York.

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

HOPPE, K. A. (2004). Late Pleistocene mammoth herd structure, migration patterns, and Clovis hunting strategies inferred from isotopic analyses of multiple death assemblages. *Paleobiology* **30**, 129–145.

HOPPE, K. A. & KOCH, P. L. (2007). Reconstructing the migration patterns of late Pleistocene mammals from northern Florida, USA. *Quaternary Research* **68**, 347–352.

HOPPE, K. A., KOCH, P. L., CARLSON, R. W. & WEBB, S. D. (1999). Tracking mammoths and mastodons: reconstruction of migratory behavior using strontium isotope ratios. *Geology* **27**, 439–442.

HOPPE, K. A., KOCH, P. L. & FURUTANI, T. T. (2003). Assessing the preservation of biogenic strontium in fossil bones and tooth enamel. *International Journal of Osteoarchaeology* **13**, 20–28.

HORTON, T. W., CHAMBERLAIN, C. P., FANTLE, M. & BLUM, J. D. (1999). Chemical weathering and lithologic controls of water chemistry in a high-elevation river system: Clark's Fork of the Yellowstone River, Wyoming and Montana. *Water Resources Research* **35**, 1643–1655.

INGRAM, B. L. & WEBER, P. K. (1999). Salmon origin in California's Sacramento – San Joaquin river system as determined by otolith strontium isotopic composition. *Geology* **27**, 851–854.

JONES, L. M. & FAURE, G. (1972). Strontium isotope geochemistry of Great Salt Lake, Utah. *Geological Society of America Bulletin* **83**, 1875–1880.

JORGENSON, G. A., EERKENS, J. W., BARFORD, G. H. & BARTELINK, E. J. (2009). Migration patterns in the prehistoric California Delta: analysis of strontium isotopes. *Proceedings of the Society for California Archaeology* **23**, 1–7.

JUAREZ, C. A. (2008). Strontium and geolocation, the pathway to identification for deceased undocumented Mexican border-crossers: a preliminary report. *Journal of Forensic Sciences* **53**, 46–49.

KAWASAKI, A., ODA, H. & HIRATA, T. (2002). Determination of strontium isotope ratio of brown rice for estimating its provenance. *Soil Science and Plant Nutrition* **48**, 635–640.

KELLER, K., BLUM, J. D. & KLING, G. W. (2007). Geochemistry of soils and streams on surfaces of varying ages in Arctic Alaska. *Arctic, Antarctic, and Alpine Research* **39**, 84–98.

KELLER, K., BLUM, J. D. & KLING, G. W. (2010). Stream geochemistry as an indicator of increasing permafrost thaw depth in an Arctic watershed. *Chemical Geology* **273**, 76–81.

KENNEDY, P. G., BLUM, J. D., FOLT, C. L. & NISLOW, K. H. (2000). Using natural strontium isotopic signatures as fish markers: methodology and application. *Canadian Journal of Fisheries and Aquatic Sciences* **57**, 2280–2292.

KOCH, P. L., HALLIDAY, A. N., WALTER, L. M., STEARLEY, R. F., HUSTON, T. J. & SMITH, G. R. (1992). Sr isotopic composition of hydroxyapatite from recent and fossil salmon: the record of lifetime migration and diagenesis. *Earth and Planetary Science Letters* **108**, 277–287.

KOCH, P. L., HEISINGER, J., MOSS, C., CARLSON, R. W., FOGEL, M. L. & BEHRENSMEYER, A. K. (1995). Isotopic tracking of change in diet and habitat use in African Elephants. *Science* **267**, 1340–1343.

KOENKER, R. (2015). quantreg: quantile regression. R package, version 5.11. Available at http://CRAN.R-project.org/package=quantreg Accessed 10.08.2015.

KOMSTA, L. & NOVOMESTKY, F. (2015). moments: moments, cumulants, skewness, kurtosis and related tests. R package, version 0.14. Available at http://CRAN.R-project.org/package=moments Accessed 10.08.2015.

KRABBENHOFT, A., EISENHAUER, A., BOHM, F., VOLLSTAEDT, H., FIETZKE, J., LIEBETRAU, V., AUGUSTIN, N., PEUCKER-EHRENBRINK, B., MULLER, M. N., HORN, C., HANSEN, B. T., NOLTE, N. & WALLMANN, K. (2010). Constraining the marine strontium budget with natural strontium isotope fractionations (^{87}Sr/^{86}Sr*, $\delta^{88/86}$Sr) of carbonates, hydrothermal solutions and river waters. *Geochimica et Cosmochimica Acta* **74**, 4097–4109.

MARCHIONNI, S., BRASCHI, E., TOMMASINI, S., BOLLATI, A., CIFELLI, F., MULINACCI, N., MATTEI, M. & CONTICELLI, S. (2013). High-precision ^{87}Sr/^{86}Sr analyses in wines and their use as a geological fingerprint for tracing geographic provenance. *Journal of Agricultural and Food Chemistry* **61**, 6822–6831.

MATTHEWS, J. H. (2007). *Research in Motion: Patterns of Large-Scale Migration in Dragonflies and Birds*. University of Texas at Austin, Austin.

MAURER, A.-F., GALER, S. J. G., KNIPPER, C., BEIERLEIN, L., NUNN, E. V., PETERS, D., TÜTKEN, T., ALT, K. W. & SCHÖNE, B. R. (2012). Bioavailable ^{87}Sr/^{86}Sr in different environmental samples — Effects of anthropogenic contamination and implications for isoscapes in past migration studies. *Science of the Total Environment* **433**, 216–229.

MCARTHUR, J. M., HOWARTH, R. J. & BAILEY, T. R. (2001). Strontium isotope stratigraphy: LOWESS Version 3: best fit to the Marine Sr – Isotope Curve for 0–509 Ma and accompanying look – up table for deriving numerical age. *The Journal of Geology* **109**, 155–170.

MCCALL, A. E. (2013). *The relationship of stable isotopes to Late Woodland and Fort ancient agriculture, mobility, and paleopathologies at the Turpin Site*. University of Cincinnati, Cincinnati.

MILLER, E. K., BLUM, J. D. & FRIEDLAND, A. J. (1993). Determination of soil exchangeable-cation loss and weathering rates using Sr isotopes. *Nature* **362**, 438–441.

MILLER, J. A., BUTLER, V. L., SIMENSTAD, C. A., BACKHUS, D. H. & KENT, A. J. R. (2011). Life history variation in upper Columbia River Chinook salmon (*Oncorhynchus tshawytscha*): a comparison using modern and ~500-year-old archaeological otoliths. *Canadian Journal of Fisheries and Aquatic Sciences* **68**, 603–617.

NYCHKA, D., FURRER, R. & SAIN, S. (2015). fields: tools for spatial data. R package, version 8.2-1. Available at http://CRAN.R-project.org/package=fields Accessed 10.08.2015.

PALMER, M. R. & EDMOND, J. M. (1989). The strontium isotope budget of the modern ocean. *Earth and Planetary Science Letters* **92**, 11–26.

PETT-RIDGE, J. C., DERRY, L. A. & KURTZ, A. C. (2009). Sr isotopes as a tracer of weathering processes and dust inputs in a tropical granitoid watershed, Luquillo Mountains, Puerto Rico. *Geochimica et Cosmochimica Acta* **73**, 25–43.

PORDER, S., PAYTON, A. & HADLY, E. A. (2003). Mapping the origin of faunal assemblages using strontium isotopes. *Paleobiology* **29**, 197–204.

POSZWA, A., FERRY, B., DAMBRINE, E., POLLIER, B., WICKMAN, T., LOUBET, M. & BISHOP, K. (2004). Variations of bioavailable Sr concentration and ^{87}Sr/^{86}Sr ratio in boreal forest ecosystems. Role of biocycling, mineral weathering and depth of root uptake. *Biogeochemistry* **67**, 1–20.

PRETTI, V. A. & STEWART, B. W. (2002). Solute sources and chemical weathering in the Owens Lake watershed, eastern California. *Water Resources Research* **38**, 1127.

PRICE, T. D., BURTON, J. H. & BENTLEY, R. A. (2002). The characterization of biologically available strontium isotope ratios for the study of prehistoric migration. *Archaeometry* **44**, 117–135.

PRICE, T. D., BURTON, J. H. & STOLTMAN, J. B. (2007). Place of origin of prehistoric inhabitants of Aztalan, Jefferson Co., Wisconsin. *American Antiquity*, **72**, 524–538.

PRICE, T. D., JOHNSON, C. M., EZZO, J. A., BURTON, J. H. & ERICSON, J. A. (1994). Residential mobility in the prehistoric Southwest United States: a preliminary study using strontium isotope analysis. *Journal of Archaeological Science* **21**, 315–330.

QUINN, R. L., TUCKER, B. D. & KRIGBAUM, J. (2008). Diet and mobility in Middle Archaic Florida: stable isotopic and faunal evidence from the Harris Creek archaeological site (8Vo24), Tick Island. *Journal of Archaeological Science* **35**, 2346–2356.

RADLOFF, F. G. T., MUCINA, L., BOND, W. J. & LE ROUX, P. J. (2010). Strontium isotope analyses of large herbivore habitat use in the Cape Fynbos region of South Africa. *Oecologia* **164**, 567–578.

R Development Core Team (2014). *R: A Language and Environment for Statistical Computing*. R Foundation for Statistical Computing, Vienna.

REYNOLDS, A. C., QUADE, J. & BETANCOURT, J. L. (2012). Strontium isotopes and nutrient sourcing in a semi-arid woodland. *Geoderma* **189**, 574–584.

ROSE, S. & FULLAGAR, P. D. (2005). Strontium isotope systematics of base flow in Piedmont Province watersheds, Georgia (USA). *Applied Geochemistry* **20**, 1571–1586.

ROSENTHAL, H. L., COCHRAN, O. A. & EVES, M. M. (1972). Strontium content of mammalian bone, diet and excreta. *Environmental Research* **5**, 182–191.

SEALY, J. C., VAN DER MERWE, N. J., SILLEN, A., KRUGER, F. J. & KRUEGER, H. W. (1991). ^{87}Sr/^{86}Sr as a dietary indicator in modern and archaeological bone. *Journal of Archaeological Science* **18**, 399–416.

SELLICK, M. J., KYSER, T. K., WUNDER, M. B., CHIPLEY, D. & NORRIS, D. R. (2009). Geographic variation of strontium and hydrogen isotopes in avian tissue: implications for tracking migration and dispersal. *PLoS ONE* **4**, e4735.

SEMHI, K., CLAUER, N. & PROBST, J. L. (2000). Strontium isotope compositions of river waters as records of lithology-dependent mass transfers: the Garonne River and its tributaries (SW France). *Chemical Geology* **168**, 173–193.

SILLEN, A., HALL, G., RICHARDSON, S. & ARMSTRONG, R. (1998). ^{87}Sr/^{86}Sr ratios in modern and fossil food-webs of the Sterkfontein Valley: implications for early hominid habitat preference. *Geochimica et Cosmochimica Acta* **62**, 2463–2473.

SINGLETON, M. J., MAHER, K., DEPAOLO, D. J., CONRAD, M. E. & DRESEL, P. E. (2006). Dissolution rates and vadose zone drainage from strontium isotope measurements of groundwater in the Pasco Basin WA unconfined aquifer. *Journal of Hydrology* **32**, 39–58.

SLATER, P. A., HEDMAN, K. M. & EMERSON, T. E. (2014). Immigrants at the Mississippian polity of Cahokia: strontium isotope evidence for population movement. *Journal of Archaeological Science* **44**, 117–127.

*SMITH, F. A., LYONS, S. K., ERNEST, S. K. M., JONES, K. E., KAUFMAN, D. M., DAYAN, T., MARQUET, P. A., BROWN, J. H. & HASKELL, J. P. (2003). Body mass of late Quaternary mammals. *Ecology* **84**, 3402 (updated version obtained from senior author).

SOKAL, R. R. & ROHLF, F. J. (2012). *Biometry*. Fourth Edition. W.H. Freeman, New York.

STORDAL, M. C. & WASSERBURG, G. J. (1986). Neodymium isotopic study of Baffin Bay water: sources of REE from very old terranes. *Earth and Planetary Science Letters* **77**, 259–272.

STUEBER, A. M., PUSHKAR, P. & BALDWIN, A. D. (1972). Survey of ^{87}Sr/^{86}Sr ratios and total strontium concentrations in Ohio stream and ground waters. *The Ohio Journal of Science* **72**, 97–104.

TOOTS, H. & VOORHIES, M. R. (1965). Strontium in fossil bones and the reconstruction of food chains. *Science* **149**, 854–855.

VAN DER HOVEN, S. J. & QUADE, J. (2002). Tracing spatial and temporal variations in the sources of calcium in pedogenic carbonates in a semiarid environment. *Geoderma* **108**, 259–276.

VOGEL, J. C., EGLINGTON, B. & AURET, J. M. (1990). Isotope fingerprints in elephant bone and ivory. *Nature* **346**, 747–749.

WADLEIGH, M. A., VEIZER, J. & BROOKS, C. (1985). Strontium and its isotopes in Canadian rivers: fluxes and global implications. *Geochimica et Cosmochimica Acta* **49**, 1727–1736.

WALTHER, B. D. & THORROLD, S. R. (2008). Continental-scale variation in otolith geochemistry of juvenile American shad (*Alosa sapidissima*). *Canadian Journal of Fisheries and Aquatic Sciences* **65**, 2623–2635.

WALTHER, B. D. & THORROLD, S. R. (2009). Inter-annual variability in isotope and elemental ratios recorded in otoliths of an anadromous fish. *Journal of Geochemical Exploration* **102**, 181–186.

WALTHER, B. D., THORROLD, S. R. & OLNEY, J. E. (2008). Geochemical signatures in otoliths record natal origins of American shad. *Transactions of the American Fisheries Society* **137**, 57–69.

WEST, J. B., HURLEY, J. M., DUDÁS, F. Ö. & EHLERINGER, J. R. (2009). The stable isotope ratios of marijuana. II. Strontium isotopes relate to geographic origin. *Journal of Forensic Sciences* **54**, 1261–1269.

WIDGA, C., WALKER, J. D. & STOCKLI, L. D. (2010). Middle Holocene Bison diet and mobility in the eastern Great Plains (USA) based on δ^{13}C, δ^{18}O, and ^{87}Sr/^{86}Sr analyses of tooth enamel carbonate. *Quaternary Research* **73**, 449–463.

WOLFF, B. A., JOHNSON, B. M., BRETON, A. R., MARTINEZ, P. J. & WINKELMAN, D. L. (2012). Origins of invasive piscivores determined from the strontium isotope ratio (^{87}Sr/^{86}Sr) of otoliths. *Canadian Journal of Fisheries and Aquatic Sciences* **69**, 724–739.

XIN, G. (1993). *Strontium Isotope Study of the Peconic River Watershed, Long Island*. SUNY Stony Brook, New York.

YANG, C., TELMER, K. & VEIZER, J. (1996). Chemical dynamics of the "St. Lawrence" riverine system: δD_{H2O}, $\delta^{18}O_{H2O}$, $\delta^{13}C_{DIC}$, $\delta^{34}S_{sulfate}$, and dissolved ^{87}Sr/^{86}Sr. *Geochimica et Cosmochimica Acta* **60**, 851–866.

IX. SUPPORTING INFORMATION

Additional supporting information may be found in the online version of this article.

Appendix S1. Discussion of spatial uncertainties in the local water model developed by Bataille & Bowen (2012).

Table S1. Mammalian taxa included in analyses, their geologic age ranges, and body masses.

Table S2. Fish taxa included in analyses.

Table S3. Results from stepwise Levene tests comparing the homogeneity of variance in Sr offsets between low and medium Sr complexity sites for each material.

Table S4. Results from pairwise Wilcoxon tests comparing Sr offsets among materials for regions with (a) low Sr complexity, and (b) medium Sr complexity.

Fig. S1. Boxplots comparing the magnitude of Sr offsets for materials from regions with low, medium, and high Sr complexity across the contiguous USA.

Fig. S2. Boxplots comparing magnitudes of Sr offsets for small- (<100 kg) and large-mammal (>100 kg cut-off) skeletal tissues, and surface water and fish skeletal tissues from small and large watersheds using the georeferenced locality ('0 km buffer'), the median modelled Sr ratio within a 5 km radius ('5 km buffer'), and the median modelled ratio within a 10 km radius ('10 km buffer') for regions with low Sr complexity.

Fig. S3. Boxplots comparing magnitudes of Sr offsets among materials from low, medium, and high Sr complexity settings across the contiguous USA.