Strontium ($^{87}Sr/^{86}Sr$) mapping: A critical review of methods and approaches

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Declaration of interests

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Abstract

The use of bioavailable strontium in different environments to provenance biological materials has become increasingly common since its first applications in ecology and archaeology almost four decades ago. Provenancing biological materials using strontium isotope ratios requires a map of bioavailable strontium, commonly known as an isoscape, to compare results with. Both producing the isoscape and using it to interpret results present methodological challenges that researchers must carefully consider. A review of current research indicates that, while many archives can be analysed to produce isoscapes, modern plant materials usually provide the best approximation of bioavailable strontium and can be used alone or combined with other archives if applying machine learning. Domain mapping currently produces the most accurate, most interpretable isoscapes for most research questions; however, machine learning approaches promise to provide more accurate and geographically wide-ranging isoscapes over time. Using strontium isotope analysis for provenancing is most successful when combined with other isotopes and/or trace elements as part of a likelihood approach. Strontium isoscapes that are both appropriate and sufficiently high resolution to answer specific research questions do not exist for most parts of the world. Researchers intending to incorporate strontium analysis into their research designs should expect to conduct primary sampling and analysis to create appropriate isoscapes or refine existing ones, which should themselves not be uncritically utilised. When sampling, it is essential to collect appropriate metadata; these metadata and the results of the analyses should be archived in one of several online databases to maximize their usefulness. With increasing amounts of primary data and the likely increased availability of machine learning approaches to mapping, strontium analysis will continue to improve as a method of provenancing.

Keywords

Isotopes, isoscapes, basemap, strontium, biosphere, provenance, machine learning

1. Introduction

The use of bioavailable strontium in different environments to provenance biological materials has become increasingly common since its first applications in ecology and archaeology almost four decades ago (Ericson 1985; Graustein and Armstrong 1983). Provenancing biological materials using strontium isotope ratios has applications for assessing human and animal mobility in archaeology (Hedman et al. 2018; Knipper 2009; Madgwick et al. 2019a; Schwartz et al. 2021), animal mobility in ecology, biology, conservation, and paleontology (Brennan et al. 2015; Copeland et al. 2010; Koch et al. 1995; Hamilton et al. 2021; Lugli et al 2017; Vogel et al. 1990), individual origins in forensics (Bartelink and Chesson 2019; Degryse et al. 2012), and product origins in food science (Di Paola-Naranjo et al. 2011; Durante et al. 2013; Voerkelius et al. 2010). Useful reviews of strontium isotope analysis have been undertaken before, especially as regards archaeological applications (Bentley 2006; Montgomery 2010), and many articles on the subject include good overviews of the relevant processes (Bartelink and Chesson 2019; Bataille et al. 2020; Britton 2020). However, in recent years there has been a marked expansion in biosphere mapping projects which employ wide-ranging methodologies. The resultant isoscapes, or maps of the distributions of strontium isotope ratios, underpin provenance interpretations and are commonly used, especially in archaeological interpretations. They require critical interrogation by researchers who draw on them.

Strontium has four naturally occurring isotopes: ^{88}Sr, ^{87}Sr, ^{86}Sr, and ^{84}Sr. ^{87}Sr is formed as the radiogenic daughter isotope of ^{87}Rb (rubidium); the decay of ^{87}Rb leads to different abundances of ^{87}Sr in rocks depending on their age and their original ^{87}Rb content (Dickin 1995). The ratio of the radiogenic ^{87}Sr to the naturally abundant ^{86}Sr is variable across lithologies of different ages and with different formation histories. Due to the 48.8 billion year half-life of ^{87}Rb (Faure and Mensing 2005, p. 77), the ratio of ^{87}Sr to ^{86}Sr does not change significantly over the time scales that are of interest to researchers in archaeology, biology, forensics, food science, and other disciplines that deal with the comparatively recent past. This relative stability of the $^{87}Sr/^{86}Sr$ ratio allows strontium isotopes to be used to provenance biological materials that have taken up strontium from their environments.

Using $^{87}Sr/^{86}Sr$ ratios to provenance biological materials requires a basemap of $^{87}Sr/^{86}Sr$ ratio distributions that sample results can be compared against. These basemaps are generally referred to as isoscapes (Bowen at al. 2009; Bowen et al. 2010; Bowen 2010) and have been developed in various ways. Some early studies used $^{87}Sr/^{86}Sr$ of underlying bedrock to estimate both $^{87}Sr/^{86}Sr$ values and distributions (Beard and Johnson 2000). However, it has since been recognized that $^{87}Sr/^{86}Sr$ values of underlying bedrocks provide a problematic analogue for the bioavailable strontium that is incorporated into the tissues of plants (see Vitousek et al. 1999; Warham 2011; Hamilton et al. 2019). Superficial deposits that may have non-local or partially non-local origins, precipitation and evapotranspiration patterns, aeolian dust, sea spray, the use of modern fertilizers, and other factors have been found to influence the bioavailable strontium, as is discussed in more detail below. Recent strontium isoscapes have tended to rely on sampling biological materials from known geographical locations, measuring their $^{87}Sr/^{86}Sr$ ratios, and mapping the results, either alone or in combination with geological data. Some studies have built isoscapes directly from geological materials or from biological materials with unknown geographic origins but considered likely to be local.

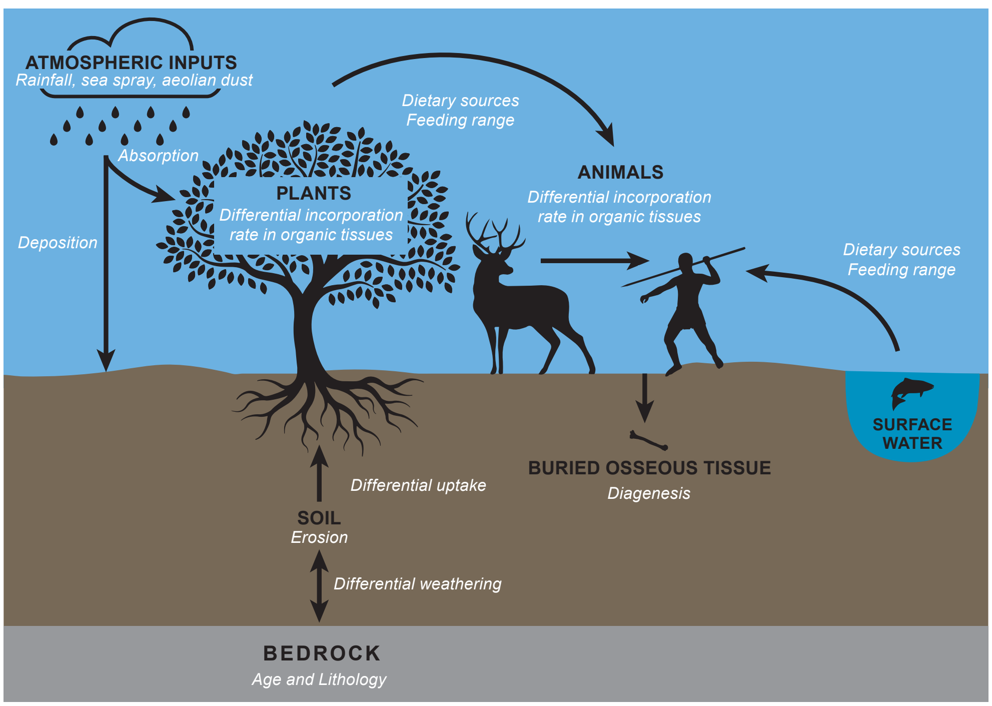
Despite the great utility of strontium isotope analysis in provenancing biological materials as well as increasing numbers of regional and global isoscapes (Adams et al. 2019; Bataille et al. 2020; Emery et al. 2018; Hedman et al. 2018; Kootker et al. 2016; Ladegaard-Pedersen et al. 2020; Pacheco-Fores et al. 2020; Scaffidi and Knudson 2020; Snoeck et al. 2020), challenges remain in using strontium isotopes to establish provenance (Ascough et al. 2018). This review seeks to critically discuss issues surrounding biosphere mapping as an aid to researchers drawing on strontium isotope research. It groups the issues into three categories: *sampling*: the use of particular strontium archives for building isoscapes; *mapping and modelling*: methods for transforming ratios into a usable isoscape, and *interpretation*: refining provenance by comparing new results against existing isoscapes.

2. Sampling

2.1 Strontium archives

Because strontium is present in soils, groundwater, and surface waters and is taken up by plants, it is incorporated into every part of an ecosystem (Price et al. 2002, see fig. 1). This means that many potential strontium archives are available for sampling and building isoscapes. Archives frequently targeted for strontium isotope sampling are ground and surface waters (Adams et al. 2019; Evans et al. 2010; Frei and Frei 2011; Maurer et al. 2012; Voerkelius et al. 2010; Wang et al. 2018), soils and soil leachates (Adams et al. 2019; Hoogewerff et al. 2019; Maurer et al. 2012; Serna et al. 2020; Willmes et al. 2018), plants (Adams et al. 2019;

Britton et al. 2020; Evans et al. 2010; Maurer et al. 2012; Ryan et al. 2018; Snoeck et al. 2020; Willmes et al. 2018), invertebrates such as snail shells and insects (Britton et al. 2020; Evans et al. 2010; Hartman and Richards 2014; Maurer et al. 2012) and wild/domestic animal remains (Adams et al. 2019; Coutu et al. 2016; Maurer et al. 2012). Additional archives have been used in discipline-specific studies. Archaeologists have used archaeological materials recovered through excavation, including plant, animal, and human remains (Hedman et al. 2018; Kootker et al. 2019; Perry et al. 2008; Scaffidi and Knudson 2020). Food studies have tested regional products such as wine (Almeida and Vasconcelos 2001; Di Paola-Naranjo et al. 2011; Marchionni et al. 2013; Durante et al. 2013), cider (García-Ruiz et al. 2007), cheese (Pillonel et al. 2003), butter (Rossmann et al. 2000), olive oil (Medini et al. 2015), and vegetables (Swoboda et al. 2008).



[**Figure 1:** Strontium pathways (adapted from Bataille et al. 2020, fig. 1)]

The different strontium archives have strengths and weaknesses for building isoscapes. Groundwater, for example, is not likely to make up a significant part of strontium intake for terrestrial animals, including humans, unless well water is regularly consumed. Deep groundwater in particular may reflect underlying lithological formations rather than the more surficial deposits that tend to influence bioavailable strontium. The effect may be significant in areas where the underlying lithological formations are not the primary parent material of the surficial deposits or where deep groundwater is confined in aquifers. Surface sources such as river waters have been used to build strontium isoscapes (Frei and Frei 2011; Wang at al. 2018); however, some studies have indicated that surface waters may not reflect terrestrial bioavailable strontium as closely as other archives (Britton et al. 2020; Hamilton et al. 2019). Additionally, some studies have raised concerns that surface waters may experience contamination from anthropogenic soil treatments that affect $^{87}Sr/^{86}Sr$ ratios (Casado et al. 2019), though there is debate over whether this contamination is significant (Maurer et al. 2012). When sampling surface waters to construct strontium isoscapes, it is vital to avoid areas that could be affected by runoff or pollution. In addition, differences between strontium isotope ratios in surface waters and in terrestrial strontium archives must be considered in ecological studies involving aquatic or semi-aquatic animals. Semi-aquatic animals have been found to be a poor archive for identifying local terrestrial strontium ratios (Lambert 2019).

Soils and soil leachates are sometimes targeted when the effects of modern agriculture on topsoil strontium is either not a problem or desirable, as in provenancing agricultural products (Durante et al. 2013) and forensic case work (Hoogewerff et al. 2019), and have also sometimes been utilized in archaeological studies (e.g. Groves et al. 2013).

Modern plants are one of the most commonly targeted archives for building strontium isoscapes because they are easily collected, demonstrably local, ubiquitous and form the base of the food chain from which strontium enters the diet of animals and humans (Montgomery 2010, p. 328), whose tissues are often the materials to be provenanced. Using plants to map strontium ratios is potentially problematic, however, because of the possibility of point-bias: due to the idiosyncrasies of geological and environmental processes, a single plant may have a ratio that is not representative of the area where it grows. This problem can be addressed by analysing a large number of samples, but such an approach rapidly becomes prohibitively expensive. An alternative approach is the use of homogenized sampling, where multiple plants are collected from a radius around each collection point and combined during processing into a single analytical sample. Homogenized sampling has been shown to produce reliable results that are either not significantly different from each other or have small differences that can be attributed to expected natural variation (Johnson 2018).

Differences in strontium ratios have been found among plants with different root systems: grasses with shallow root systems tend to reflect the strontium ratios of the topsoil while shrubs and trees with deeper root systems tend to reflect the strontium ratios of deeper soils (Britton et al. 2020; Hartman and Richards 2014). Hartman and Richards (2014, p. 259-260) found this effect to be so pronounced in Northern Israel and the Golan that they suggest it may be possible to distinguish between grazing and browsing animals based on the difference in their strontium isotope ratios. However, Willmes et al. (2018, p. 79) found no significant difference between grasses, tree roots, and other plant samples in France, suggesting that the way shallow- and deep-rooted plants reflect bioavailable strontium may be dependent on local environmental factors such as soil types and precipitation regimes.

Homogenized sampling has also been used to address differences between deep- and shallow-rooted plants. Both types of plants are collected from each sampling area and blended, resulting in samples that represent an average of the available plant resources (Ventresca Miller et al. 2018). This approach has the benefit of being more likely to reflect the strontium in human consumption as a variety of plant and animal resources are certain to be consumed.

Snail shells are often considered a useful archive because they contain high concentrations of strontium, represent an average of the animals' grazing over several square meters over a few years, and are easy to dissolve for analysis. However, snail shells have been shown to more closely reflect the $^{87}Sr/^{86}Sr$ ratios of shallow-rooted, as opposed to deep-rooted plants (Hartman and Richards 2014), which makes them susceptible to the influences of modern soil treatments. Additionally, snail shells from a variety of substrates have been shown to produce strontium ratios strongly affected by rainwater (Evans et al. 2010), meaning they can provide a problematic archive. Insect carapaces are sometimes targeted for strontium analysis (Hartman and Richards 2014; Holder et al. 2014; Murphy et al. 2020); however, some insects travel over long distances and the species of insect should be taken into careful consideration before using their data to build isoscapes (cf. Bataille 2018, p. 4).

Wild animals, whether modern, historic, or ancient, are another archive with the advantage of generally being readily available. Recent specimens have come from historic collections (Coutu et al. 2016) and been collected as roadkill (Adams et al. 2019). Archaeologically, wild animal remains are collected along with geospatial data during excavation and stored for later study. Wild animal remains therefore provide a readily available source of strontium isotope data while avoiding additional field sampling. Large-bodied wild animals may also be targeted for strontium analysis because their greater home ranges are seen to average local strontium signatures and therefore be more likely to reflect human consumption patterns (cf. Crowley et al. 2017). Because wild animals are less likely to be as actively managed and controlled as domestic animals, their strontium isotope ratios may not be as affected by human behaviours (e.g. soil treatment), but human activity can affect their home range meaning they exploit more marginal areas (see Mulville et al. 2009). However, large-bodied wild animals can have very large feeding ranges, are sometimes managed by humans (see Madgwick et al. 2013; Sykes et al. 2016) and may have been hunted a long way from the site, factors which introduce substantial problems in using their remains to map local bioavailability. Additionally, the presence of large-bodied wild animal remains at specific sites depends on aspects of the palaeoeconomy, depositional practices, and preservation.

Small-bodied, non-migratory wild mammals are often preferred for sampling because their limited home ranges and short lifespans mean that their tissues preserve a local strontium isotope ratio specific to a particular point in time (Bentley 2006). This can be especially advantageous in archaeological applications if there are concerns that significant changes in erosion, climate, superficial deposits or other factors may have changed the bioavailable strontium in the soils over time meaning that modern plants would provide a poor analogue. Small-bodied wild animals also have the advantage of averaging the strontium ratios of the resources in their small home ranges. They have been used by a number of studies for establishing strontium isoscapes (e.g. Barberena et al. In Press; Hedman et al. 2018; Kootker et al. 2016; Serna et al. 2020). However, it should be considered that small-bodied mammals, especially commensal rodents, may be unintentionally transported outside their local ranges by human activities such as trade, as has been identified in one study of ancient Jordan (Perry et al. 2008). Additionally, the species of small-bodied animal may be significant in some environments. A study of archaeological materials in Utah, USA, used archaeological rodents and lagomorphs to characterize the strontium isotope ratios of nine archaeological sites (Lambert 2019). This study found that semi-aquatic muskrats did not provide reliable strontium ratios for characterizing sites and that terrestrial squirrels were preferable.

Domestic animals, both modern and ancient, also have the advantage of generally being readily available and have often been used to define bioavailable strontium (e.g. Bentley and Knipper 2005; Knudson et al. 2004; Sykes et al. 2006). However, herbivores such as cattle and caprines and omnivores such as pigs have different implications as strontium archives. Like large-bodied wild animals, domestic herbivores are likely to feed over large ranges, averaging the strontium ratios of their food sources. This behaviour can be considered more reflective of human consumption and therefore more useful for developing isoscapes to understand human provenance or mobility (Evans and Tatham 2004). However, the food sources of domestic herbivores tend to be more limited. Domestic pigs may feed over smaller ranges but are more likely to be fed diverse foods that more closely reflect the human diet (cf. Evans et al. 2009). Dogs provide another potentially useful analogue for human consumption (Guiry 2012). Regardless of species, the feeding regimes and patterns of mobility of domestic animals are often highly controlled (cf. Makarewicz and Sealy 2015). In the present, this may include the consumption of significant amounts of non-local feed, making domestic animals poorly reflective of their local environments. In the past, this could include control of pasturage as well as feeding in areas where soil treatments such as manuring had been applied, again changing their strontium isotope ratios. Management regimes such as transhumance can mean substantial seasonal movement of domesticates, potentially causing their biogenic ratios to reflect a mixture of lithological zones rather than a local value. Additionally, domestic animals have been exchanged over long distances since early in prehistory (Bentley and Knipper 2005; Grumbkow et al. 2013; Madgwick et al. 2019a; Shaw et al. 2009; Shaw et al. 2010; van der Jagt et al. 2012), meaning that they cannot be assumed *a priori* to be local even in archaeological contexts.

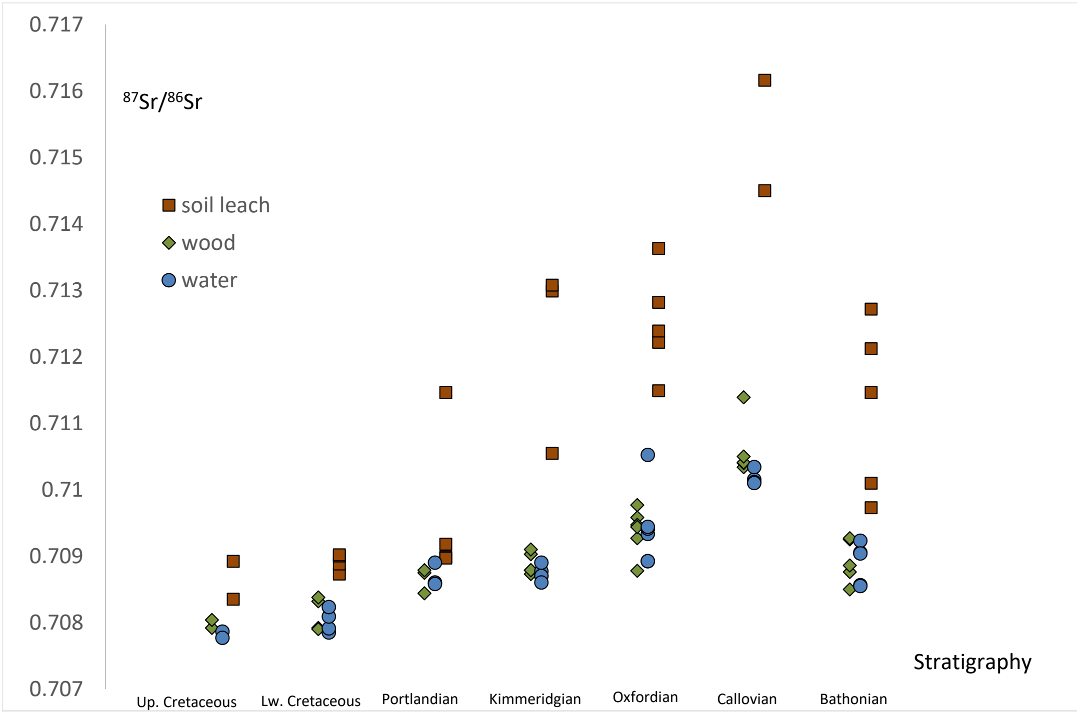
In archaeological applications, human remains are sometimes used to identify local strontium ratios. However, studies that use human tissues generally start from the assumption that most people living in an area would have been local, and therefore identify outliers as non-local (Scaffidi and Knudson 2020). This approach is potentially problematic because many human societies include highly mobile individuals. Economic practices and opportunities, enslavement, refugeeism, and mobility associated with matrilocal or patrilocal marriage customs are all reasons why many individuals in any given community may not have local origins. In the small sample sizes that are usually used when building isoscapes from archaeological humans remains, the effect of even a few migrants may be significant. Additionally, in some circumstances individuals may consume large proportions of non-local foods. If the imported foods are luxury products, probably consumed primarily by elite adults, their effects on strontium could potentially be avoided by focusing sampling on the second and third molar, which form during childhood and do not remodel. However, this approach may not work if the imported foods are staple foods, as is widely documented for ancient urban societies such as Classical Athens and Imperial Rome (Erdkamp 2005; Moreno 2007). In addition, essential food additives like imported sea salt may impact biogenic values, as has been suggested for ancient Maya Tikal (Wright 2005). Despite the issues, some scholars have argued that increasing datasets of strontium isotope analysis of human remains will ultimately allow for more accurate assessments of human origins than isoscapes built from proxy data are capable of providing (Burton and Hahn 2016; Evans et al. 2012).

When using animal or human remains to build strontium isoscapes, it is preferable to sample tooth enamel rather than other tissues. Tooth enamel does not remodel after it has mineralized and is more resistant to diagenetic change than other tissues (Budd et al. 2000; Bentley et al. 2004; Madgwick et al. 2012), although it still suffers some diagenetic effects, but not to a degree that would generally affect interpretation (Lewis 2015). Dental enamel can be sampled by mechanical extraction (i.e. cutting segments using a precision drill with diamond wheel) or through laser ablation. Laser ablation has the potential for higher temporal resolution, although time-averaging in the incorporation of biogenic strontium may mean this is often not of great interpretative value (Montgomery et al. 2010). Some studies have sought to use dentine and bone to establish local strontium ratios on the argument that diagenetic change means that these more porous tissues will reflect the strontium ratios of the sediment where they are buried (Stantis et al. 2019). However, there is some evidence that dentine and bone do not always fully re-equilibrate with diagenetic strontium from the burial environment (Madgwick et al. 2019b), meaning that how closely they reflect the local strontium ratio depends on post-depositional factors and the original biogenic composition. Because these factors cannot be determined *a priori*, it is advisable to use only enamel for strontium analysis. While analyzing enamel alone is fairly easy to accomplish for large-bodied animals, some inclusion of dentine may be unavoidable when processing the teeth of rodents, which are so small as to make the removal of dentine by mechanical means problematic. Some studies have sought to minimize this problem by targeting rodent incisors, which have a smaller proportion of dentine than other teeth (Serna et al. 2020).

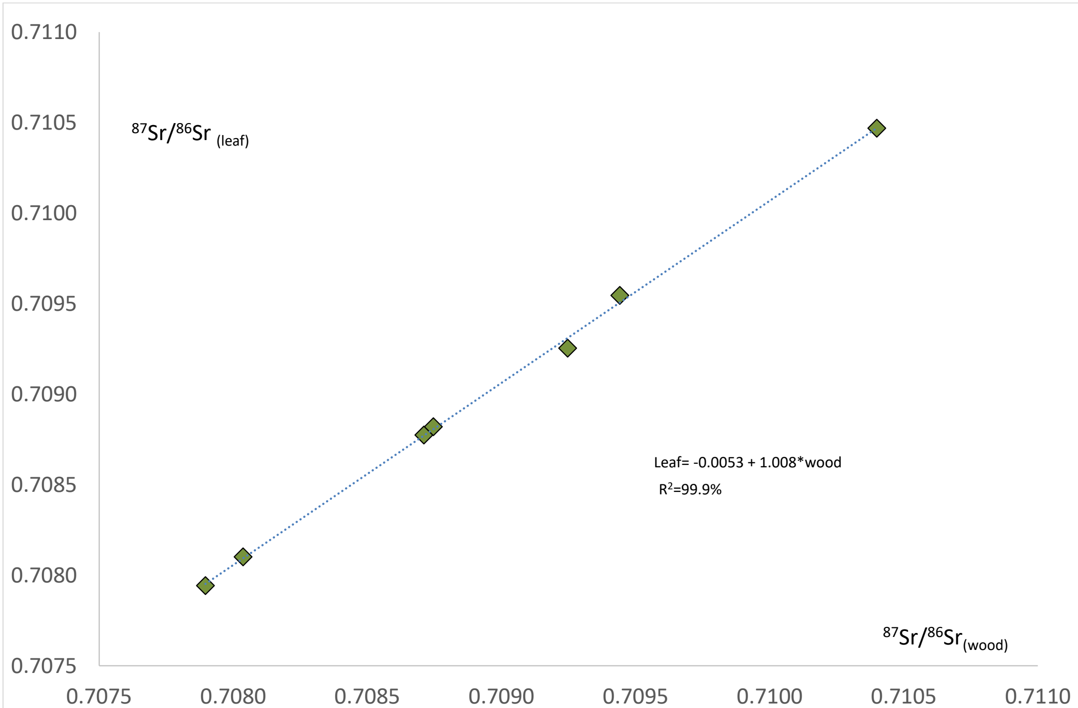
A final category of strontium archive used for mapping is modern food products. Establishing strontium isotope ratios for modern products of known geographical origin can be very useful when the goal is to provenance similar products of unknown origin. Large-scale projects have been undertaken in Europe to establish these kinds of comparative data (Asfaha et al. 2019; Voerkelius et al. 2010). Additionally, mapping projects in other disciplines have incorporated results from modern food studies (Emery et al. 2018).

2.2 *Comparing archives*

The ultimate goal of many strontium studies is to identify the geographical origins of individual humans or animals in the past or present, and many attempts have been made to identify the most suitable archives for building strontium isoscapes that can be used for this purpose (Britton et al. 2020; Ladegaard-Pedersen et al. 2020; Maurer et al. 2012; Ryan et al. 2018; Snoeck et al. 2020; Warham 2011). It is often assumed that strontium isoscapes that reflect local bioavailable strontium ratios will be the easiest to interpret. Plant samples have repeatedly been found to be the most suitable (Britton et al. 2020; Ryan et al. 2018) or one of the most suitable archives (Ladegaard-Pedersen et al. 2020; Maurer et al. 2012; Warham 2011) for establishing local ratios of bioavailable strontium. Modern surface waters have also been found to reflect bioavailable strontium ratios accurately (Ladegaard-Pedersen et al. 2020; Maurer et al. 2012; Warham 2011). Some studies have found little difference between soil leachates and plants (Ladegaard-Pedersen et al. 2020) while other studies have found significant differences (Warham 2011, see fig. 2), indicating that the two archives should not be assumed to be equivalent. A comparison of leaves and wood shows no impact caused by the accumulation of environmental dust on leaves, indicating that wood and leaves are equally accurate archives of bioavailable strontium (Warham 2011, see fig. 3). It is worth noting, however, that most studies that directly compare archives have taken place in Europe, and it has been argued that the conclusion that plants and water are the best archives for understanding bioavailable strontium is due to specific intensive agricultural practices in Europe and is not necessarily true globally (Adams et al. 2019, p. 234).



[**Figure 2:** An individual value plot of wood, water and soil leachate $^{87}Sr/^{86}Sr$ data for samples collected across the principal lithologies within the study area in Southern England. Drawn from data in Warham 2011.]



[**Figure 3:** A regression plot of the $^{87}Sr/^{86}Sr$ composition of unwashed leaf vs core wood, from plants collected from the Jurassic and Cretaceous outcrops of southern England. The high correlation between leaf, exposed to airborne deposits, and inner core composition of the plant demonstrates the lack of contaminating exotic dust on leaf surfaces. Drawn from data in Warham 2011.]

Differences have also been identified among plants of different root depths such as grasses, shrubs, and trees (Britton et al. 2020; Hartman and Richards 2014; Snoeck et al. 2020). Based on work in Ireland, Snoeck et al. (2020, p. 9) concluded that "sampling is a crucial part of assessing the variability in the isotope ratios of the biologically available strontium of a site or region". A study in Israel that gathered plants from protected areas not affected by recent agriculture found that ligneous plants tended to correlate more closely with underlying bedrocks while non-ligneous plants showed more of the effects of atmospheric strontium deposition through precipitation and aeolian dust (Hartman and Richards 2014). The same study found that invertebrates – snail shells and insects – correlated strongly with plants but more closely with non-ligneous plants. These differences indicate that ligneous plants should be preferentially sampled in areas where recent human activity may have affected soil conditions and also that caution should be exercised when using snail shells for the same reason – they are likely to be affected by the strontium ratios in the upper parts of the soil.

Although the utility of plants with root systems that penetrate below the topsoil layer for mapping bioavailable strontium has been demonstrated, this archive is not available or appropriate to all mapping studies. Instances where this archive may not be practicable include areas where significant erosion has occurred since the period under study (discussed below) and areas where deeper-rooted plants are not available. In these instances, archaeological materials (Scaffidi and Knudson 2020) or other archives may be employed to build strontium isoscapes.

2.3 Environmental influences on $^{87}Sr/^{86}Sr$ ratios

Several environmental factors outside of bedrock geology, age, and differential weathering have been identified as potentially influencing the isotope values of soils and plants in ways that may be problematic for some applications of strontium isotopes for provenancing. Even regions with relatively homogenous soil origins have been found to have diverse patterns of bioavailable strontium (Kootker et al. 2016).

Environmental influences on strontium isotope ratios include the presence of sediments such as shell-based sands (Evans et al. 2010), sediments transported outside their areas of origin by erosion, rivers and streams, and glaciers (Pacheco-Fores et al. 2020; Serna et al. 2020; Widga et al. 2017), non-local strontium ratios carried in river and stream waters (Sillen et al. 1998), and airborne and atmospheric inputs such as aeolian dust, volcanic ash, rainfall, and sea spray (Burton and Hahn 2016; Capo et al. 1998; Chadwick et al. 2009; Evans et al. 2010; Hartman and Richards 2014; Hoogewerff et al. 2019; Serna et al. 2020).

Whether researchers wish to avoid or incorporate environmental influences will depend on the goals of their particular study. Successfully avoiding or incorporating these influences requires researchers to carefully select the strontium archives they use to build an isoscape. For example, the contribution of non-local aeolian dust to the $^{87}Sr/^{86}Sr$ ratio can be significant (Capo et al. 1998), as is seen with continentally derived dust in the American southwest (Reynolds et al. 2012) and Saharan dust blown to Europe and the Mediterranean (Hartman and Richards 2014), though this effect is not seen in the more northerly maritime climate of the UK (Warham 2011). Marine aerosols are the dominant source of strontium in some wet environments in Hawaii (Vitousek et al. 1999). Mapping from plant or small animal archives will capture the effect of aeolian and atmospheric deposits, whereas mapping primarily from underlying bedrock strontium values will not. Archaeological applications, in particular, may find environmental influences problematic, since archaeology seeks to provenance materials in a past landscape that is likely to differ from the modern one. Erosion and fluvial transport may have changed the relationship between underlying bedrock and surficial deposits, and different weather patterns in the past may have resulted in differential contributions of rain, aeolian dust, and sea spray. Provenancing studies that focus on the present, however, such as food science and forensics, need to capture recent changes to soil chemistry.

Sea spray can contribute to local soil chemistry depending on proximity and winds patterns, and researchers may wish to avoid the effects of coastal sea spray if the affected areas are considered unlikely to have had important effects on the tissue to be analyzed. Some mapping studies that rely on plant archives have sought to minimize the effects of sea spray by targeting areas at least 50 m from coastlines (Snoeck et al. 2020). However, the assumption that the effects of sea spray are limited to a zone 50 m from the shoreline has been questioned. Alonzi et al. (2020) found that on the uninhabited island of Inishark in Ireland, the zone between 0 m and 50 m from the shoreline was the zone least affected by sea spray. They also found that, from 50 m to 200 m from the shoreline, the effects of sea spray were noticeable, though variable. Additionally, in some environments, large areas of territory have been found to be affected by sea spray (Evans et al. 2010) and the sea spray zone of Evans et al. 2018a was based on plant sulphur measurements plotted as distance from coast with a greater influence seen on the prevailing wind west coast than the east coast of Britain (Fig 3). Whenever possible, researchers should design their sampling strategies to identify and clarify sea spray effects rather than trying to avoid them based on a metrical assumption that may not be valid for their area.

2.4 Anthropogenic influences on $^{87}Sr/^{86}Sr$ ratios

In addition to environmental influences, there are many potential anthropogenic influences on strontium ratios. Fly-ash and other airborne pollutants can alter strontium in soils (Straughan et al. 1981). The issue of how agricultural liming may change the strontium composition of topsoils as well as surface waters due to runoff from fields into streams has recently come under scrutiny. Some studies have suggested that contamination of surface waters by agricultural products is significant (Böhlke and Horan 2000; Casado et al. 2019) while others have identified minimal effects for strontium contamination specifically (Frei and Frei 2011). However, the minimal effects identified by Frei and Frei (2011) for Jutland have been questioned. An alternate study aimed to differentiate the strontium ratios of surface waters near agricultural activities from those of surface waters too far away from agricultural activities to be affected by them (Thomsen and Andreasen 2019). This study found meaningful differences between contaminated and uncontaminated surface waters, with important implications for palaeomobility studies. The authors recommend sampling strontium archives from “pristine” landscapes, or landscapes that have never been used for agriculture (Thomsen and Andreasen 2019). In the literal sense, this is likely to be impossible for most areas of the globe given the long and widespread practice of agriculture. In the practical sense, it may be sufficient to avoid areas that have been used for cultivation in the past several hundred years, when the importing of non-local fertilizers became more common. It is also important to consider the effects on strontium in areas where seaweed is traditionally used for fertilizer (Blanz et al. 2019). Each project will have to assess the meaning of “pristine” landscapes for the purposes of their study. For Thomsen and Andreasen, this meant streams that originated within an area at least 300 m from land used as farmland in the present or documented past, and ponds that were at least 150 m from farmland.

Not all researchers share Thomsen and Andreasen's pessimism about the extent of contamination from agricultural liming, and further research has been undertaken to assess the validity of their criticism. Frei et al. (2020) found that although the strontium isotope composition was changed in topsoils due to agricultural liming, the effects were contained by the topsoil. The authors attribute these findings to the high organic content of the agricultural soils rather than to the specific characteristics of the type of soil studied, and therefore argue that their results are applicable to other agricultural areas. They were able to correlate observed changes in the strontium isotope composition of the surface waters with deeper, underlying geological features of the landscape, and argue that this is a sufficient explanation of the changes without attributing them to the effects of agricultural liming.

While the question of strontium contamination of surface waters through liming remains open, the potential for this and other soil treatments to affect the ratios of bioavailable strontium in topsoils has been demonstrated. There are several strategies for avoiding these distortions. First, selecting plants rather than surface waters as the archive to be sampled allows for targeted sampling that can avoid the effects of runoff. Next, researchers should avoid sampling plants from areas currently or recently used for agriculture or that are near rivers or streams (Maurer et al. 2012; Ryan et al. 2018). Finally, researchers should sample plants with root systems deep enough to penetrate below the layer of soil likely to be affected by the application of lime or fertilizer, i.e. trees and shrubs as opposed to grasses (cf. Frei et al. 2020, p. 11).

2.5 Effect of groundcover (forested vs. unforested)

In addition to the issues of environmental and anthropogenic inputs, Johnson (2018) observed that land use appeared to affect the composition of strontium transmitted into the biosphere. Sherwood Forest and its abutting farmland are both founded on Triassic Sherwood Sandstone. The plant samples from the ancient woodland of the Sherwood Forest National Nature Reserve have higher average strontium ratios ($^{87}Sr/^{86}Sr$ = 0.71392 ±0.00402, 2SD, n=14), while those on arable farmland on the same substrate are lower ($^{87}Sr/^{86}Sr$ = 0.71036 ±0.00222, 2SD, n=13). Further studies are in progress to describe this in more detail, but the initial suggestion (Johnson 2018) is that the long-term existence of the forest has allowed the carbonate component of the soil to be leached out, leaving a silicate dominated soil with a more radiogenic signature. These findings are potentially very important but rely on a very limited sample set at present and broadly comparable mapping projects (e.g. Warham 2011) have not found the same pattern. If this process is, however, confirmed and applies to other mixed carbonate-silicate bedrock types, it would provide an important factor in mapping changes in the biosphere through periods of major deforestation. It also has major ramifications for analysing taxa that commonly source their food from forest environments.

2.6 Combining archives

Recent attempts to build broad strontium isoscapes, often employing machine learning, have combined multiple different archives (Bataille et al. 2018; Bataille et al. 2020; Emery et al. 2018; Willmes et al. 2018). The utility of combining the results from different strontium sources requires scrutiny. While some studies have shown strong correlation among different strontium archives (Adams et al. 2019), others have shown substantial offsets even when collected from the same or very close locations (Maurer et al. 2012). Diagenetically affected dentine, for example, has been shown to differ from plants from the same area due to the equilibration process (Madgwick et al. 2019b). In addition, a wide-ranging study by Bataille et al. (2018) found that the $^{87}Sr/^{86}Sr$ ratios of different archives from the same sites differ by a mean of 0.0025-0.005 on different lithologies, and Warham (2011) found differences of 0.0006-0.0047 on different lithologies in the lowland zone of England. The offsets observed in different strontium archives are easily of magnitude that could alter interpretation when defining provenance. This may introduce problems, particularly in cases where some areas of an isoscape are modeled on a different range of archives. This is a common problem as research traditions in different areas mean that the range of available data is diverse across regions (Bataille 2018, p. 9).

There is also the potential for artificially introducing noise to biosphere maps if archives that are less consistent with human and animal diet dominate models for some regions. Statistical procedures (e.g. random forest algorithms) can alleviate some modelling issues (Bataille et al. 2020), for example by ensuring map production is insensitive to outlying values. However, the nature of archives used in models still needs to be scrutinized. It remains important that models rely on archives that are consistent with human/faunal diet if reconstructing origins is the objective. Combining archives has an important role to play in building largescale isoscapes as this approach has the advantage of maximizing diverse and sparse data sources, but these isoscapes must be used with caution due to their associated uncertainties, not least in relation to how analogous certain archives are to diet. Targeted primary analysis of valid archives (e.g. plants) from pristine landscapes is advisable to characterize bioavailable strontium at a local (e.g. immediate site environs) level. These data can in turn be used for the refinement of large scale isoscapes. These points are expanded on in the machine learning section (3.3) below.

2.7 Sample sizes

Makarewicz and Sealy (2015, p. 150) point out that many strontium isotope “baselines” have used insufficient numbers of samples to capture the range of isotopic diversity often found in biomes. Rectifying this problem is challenging. Strontium isotope analysis is expensive to conduct, and its high cost is one of the prohibiting factors in gathering large datasets. Some guidelines about the likely data range of different biomes are emerging from current studies and data from Evans et al. (2018a) provides the means for exploration. On relatively isotopically homogenous carbonate formations such as chalk and limestone, the 1SD range uncertainty is in the 4th decimal place. Seven carbonate domains, with between 9 and 85 primary biosphere analyses per domain, give uncertainty values between +/- 0.0004 and 0.0008. By contrast, in old and radiogenic granitic terrains, the 1SD uncertainty is in the 3rd decimal place, and domains with between 2 and 14 primary biosphere analyses give values between 0.0012 and 0.0036. Sandstones, clays, and basic igneous rocks have reproducibility generally between these end member ranges. Converted to %, the limestones have c. +/- 0.05% and the radiogenic igneous rock 0.5% 1SD reproducibility.

Approaches to strontium biosphere mapping are diverse and, combined with differences in homogeneity across different lithologies, it is impossible to define a suitable sample size for characterizing an area. Poorly characterized areas, with little existing data of any archive inevitably require expanded samples, as do (generally radiogenic) areas that produce more heterogeneous ratios. One way of addressing this problem is through homogenized plant sampling (Johnson 2018, discussed above). By taking multiple plant samples from each sampling area and combining them into a single analytical sample, more environmental variability can be captured per sample. This can effectively improve coverage without adding undue expense.

2.8 Collecting metadata

Researchers being able to access detailed metadata relating to each sample in a strontium isotope dataset is essential for legacy benefits to be realised. Accurate and accessible metadata is required for researchers other than those who originally collected the samples to apply the resulting information to their research. There are calls in the literature to create metadata templates for strontium sampling so standardised metadata is always recorded (Bataille et al. 2020). While not all metadata may be relevant to the researchers initially collecting the samples and conducting the analyses, it is essential that future benefits are maximised across disciplines. Sourcing biosphere data, or indeed any isotope data, on a large scale remains a laborious task and the necessity of a centralized repository has come into sharp focus in recent years (Pauli et al. 2015).

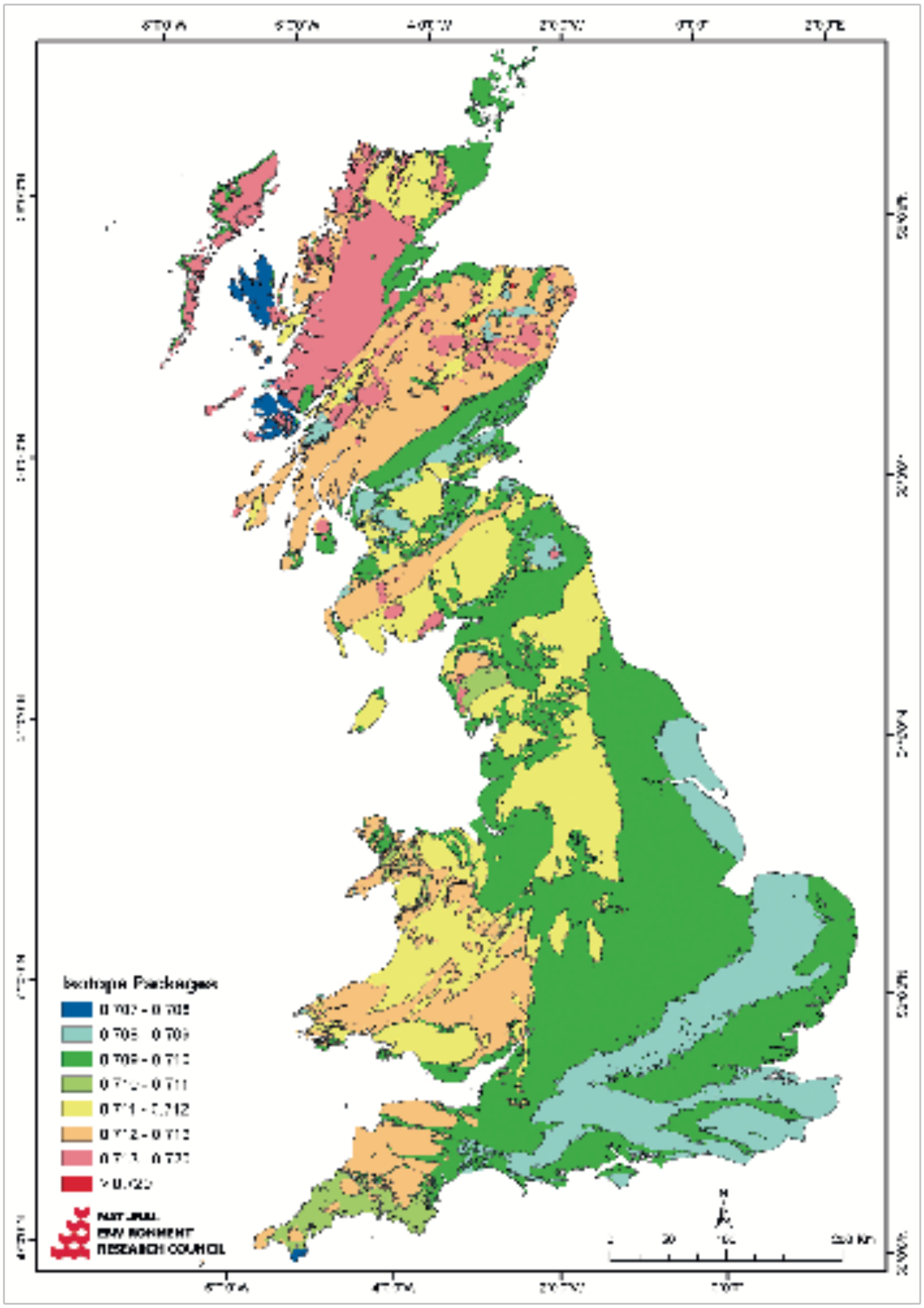
Several initiatives are being developed to ensure open access to large and diverse isotope datasets. The isotope data repository IsoBank (http://isobank.tacc.utexas.edu/en/, Pauli et al. 2017) is currently working to develop a set of templates that will guide metadata collection in different disciplines. Drafts of their recommendations are available on their website. The Faunal Isotopes Database within the global Neotoma Paleoecology Database (https://www.neotomadb.org/, Pilaar Birch and Graham 2015) provides the opportunity for archaeologists to contribute the published results of isotope studies on faunal remains, including strontium, and requests substantial metadata for each dataset and for individual specimens. This initiative benefits from wide-ranging integrated paleoecology databases, though is currently focused on North America and isotope datasets are sparse. The European, archaeology-specific isotope database IsoArch (https://isoarch.eu/, Salesse et al. 2018) provides locations and summaries of isotope studies in Europe, including bibliographic references, some metadata, and published isoscapes for some countries. IsoMemo (https://isomemo.com/) is another initiative currently under development. This big data project aims to combine isotope data from archaeology, ecology, and the environmental and life sciences by integrating multiple existing partner repositories. None of these initiatives have reached maturity and the best biosphere mapping datasets are currently those generated for the construction of primary isoscapes (e.g. Evans et al. 2018a). However, the open access storage of strontium isotope data from different archives, with a broad suite of standardised metadata, is certain to enhance the potential and resolution of isoscape construction in the future.

3. Mapping and modeling

Several methods of mapping have been employed in building strontium isoscapes from empirical data derived from analyzing different archives. These isoscapes are often the foundation of interpretation in establishing provenance and the nature of their construction requires careful scrutiny. In addition, associated uncertainties and guidelines for practical application need to be clearly outlined for end users. Principal methods comprise domain mapping, contour mapping using geostatistical methods, and machine learning approaches. Additionally, where empirical data is scarce, some researchers have sought to create predictive models of strontium ratio distributions over large regions based on data from bedrocks or surface waters.

3.1 Domain mapping

Domain mapping of strontium ratios, also referred to as the nominal approach (Bataille et al. 2018), relates directly analyzed ratios of targeted archives to specific areas (see fig. 4). Often, these areas are geological regions (Evans et al. 2010; Kootker et al. 2016; Madgwick et al. 2019b), but they may also be specific sites or site catchment areas in archaeological studies (Lambert 2019; Pacheco-Fores et al. 2020). Domain maps are the most straightforward way of creating an isoscape from the ratios of sampled archives, but also the most expensive and time consuming because the map can only be improved through increased sampling. Achieving good sample coverage, especially in areas with diverse lithologies, is challenging.



[**Figure 4:** An example of a domain map of the United Kingdom (Evans et al. 2010, fig. 1b)]

Domain maps take advantage of the frequent relationship between underlying lithologies and bioavailable strontium without assuming that a specific relationship exists in any given area.

Domain mapping is a useful approach in regions where superficial deposits derive from and mostly follow the outlines of underlying lithologies. To avoid the misleading effects of potential outliers, domain mapping requires averaging the strontium isotope ratios of multiple samples across each domain; however, homogenized sampling of plant archives (discussed above) can help address this problem without driving up the cost of the mapping project. The domain maps of Evans et al. (2010; 2018b) and Kootker et al. (2016) use median and interquartile range (IQR) and mean and 2SD of the trimmed dataset, respectively, to represent the domain range. This is done to make the mapping, based on plants/small animals, more applicable to studies of humans where, in the case of Evans et al. (2012), it has been shown that the IQR of plants is double that of human data in Britain.

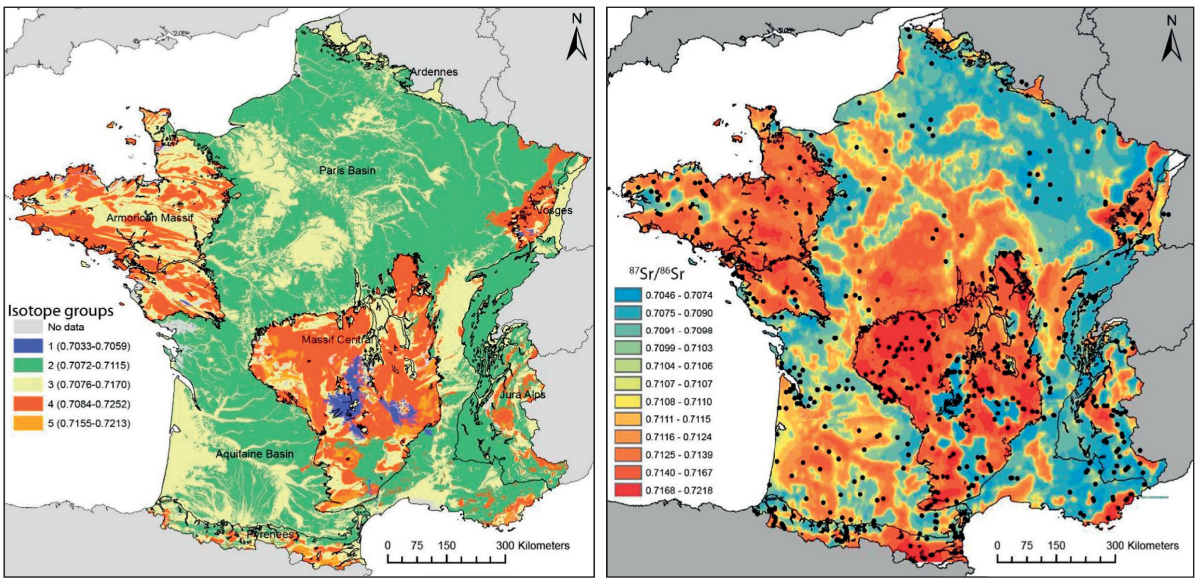
An approach comparable to domain mapping is used by Adams et al. (2019), who produce a Voronoi map as one method of creating an isoscape for Cape York, Australia, along with contour mapping methods. A Voronoi map does not assign sample values to geological regions, but rather produces a series of polygons around the locations of each of the sample points, with the value assigned to each polygon being the value of the point it contains. A Voronoi map more accurately represents sample heterogeneity than interpolated maps (discussed below) and does not require averaging sample values as a domain map does, but it does depend on representative sample distribution for accuracy.

The production of a Voronoi map can be a useful approach in regions where the bioavailable strontium is likely to have little relationship to the underlying lithologies generally used in domain mapping, or where this relationship cannot be assumed. This can be the case in places where superficial deposits do not derive from the underlying bedrock or where surface treatments are likely to have significantly altered the strontium isotope ratios of topsoils. Voronoi mapping of large areas may, however, become prohibitively expensive. Because no relationship between the strontium isotope ratio at a given location and the underlying geology is assumed, the only way to improve the specificity of the map is by adding more values.

An interactive, multi-isotope domain map for which strontium is a major focus is available for Great Britain through the British Geological Survey: https://www2.bgs.ac.uk/services/ngdc/citedData/catalogue/3b141dce-76fc-4c54-96fa-c232e98010ea.html (Evans et al. 2018a; Evans et al. 2018b). This useful resource allows researchers to enter isotope data on multiple proxies from analyzed specimens and then provides a map of their potential areas of origin. The ability to build and host interactive resources of this kind is an advantage of domain mapping, where the computing power needed to create customized maps is substantially lower than is required to create customized maps incorporating new data through machine learning (discussed below). The British Geological Survey's map is arguably the most detailed primary domain isoscape globally, dividing the British biosphere into 56 domains comprising 1km^2 hexagons. However, the whole of Great Britain is characterized by only c. 850 analyses, meaning one for approximately every 250 km^2 of landmass. This demonstrates the challenge of providing comprehensive sample coverage for large areas. Whilst this coverage sounds impossibly sparse, large areas of well characterized and relatively homogenous lithology (e.g. the chalklands of southern England), do not require dense sampling. Areas with greater heterogeneity, such as granites and other older lithologies, require much more sampling (cf. Evans et al. 2009), and it is certain that even this detailed isoscape will be subject to considerable iterative refinement as more data comes to light.

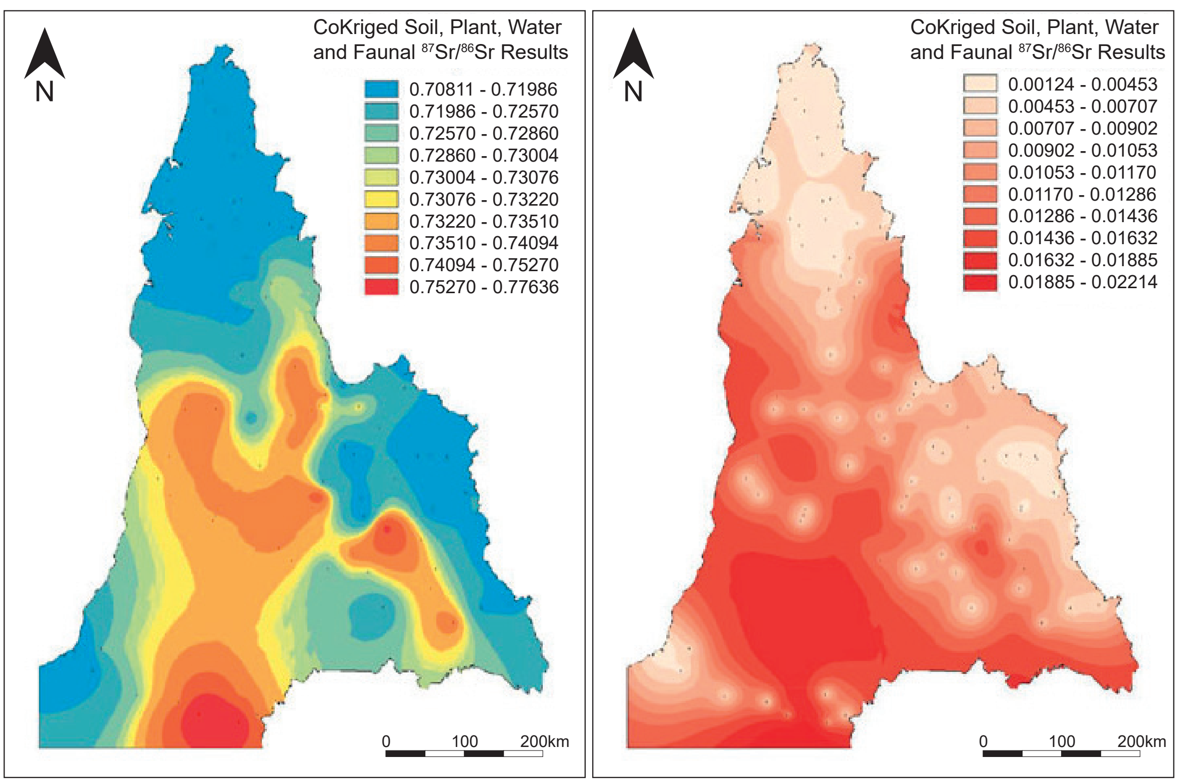
3.2 Contour mapping

Contour mapping, also referred to as a Bayesian continuous approach (as in Bataille et al. 2018), is another common approach to building strontium isoscapes. Contour mapping uses some form of geostatistics to extrapolate continuous distributions of isotope ratios across a landscape from the empirically known data points (see fig. 5). Some methods used include the Inverse Distance Weighting function in ArcGIS, simple/ordinary kriging, empirical Bayesian kriging, cokriging, and kriging with external drift (Adams et al. 2019; Emery et al. 2018; Wang et al. 2018; Willmes et al. 2018). Many studies test different methods against each other, using prediction errors to evaluate their goodness of fit. For example, Willmes et al. (2018) try both ordinary kriging and kriging with external drift to map from both soil leachates and plant samples. No significant difference was found between the maps made from soil leachate values and plant values, and for both sample types, kriging with external drift produced maps with lower prediction errors. This study also identified isotope groups through cluster analysis, and when these were used as covariate in the kriging with external drift, the process produced isoscapes closer to the expected distribution of variations.



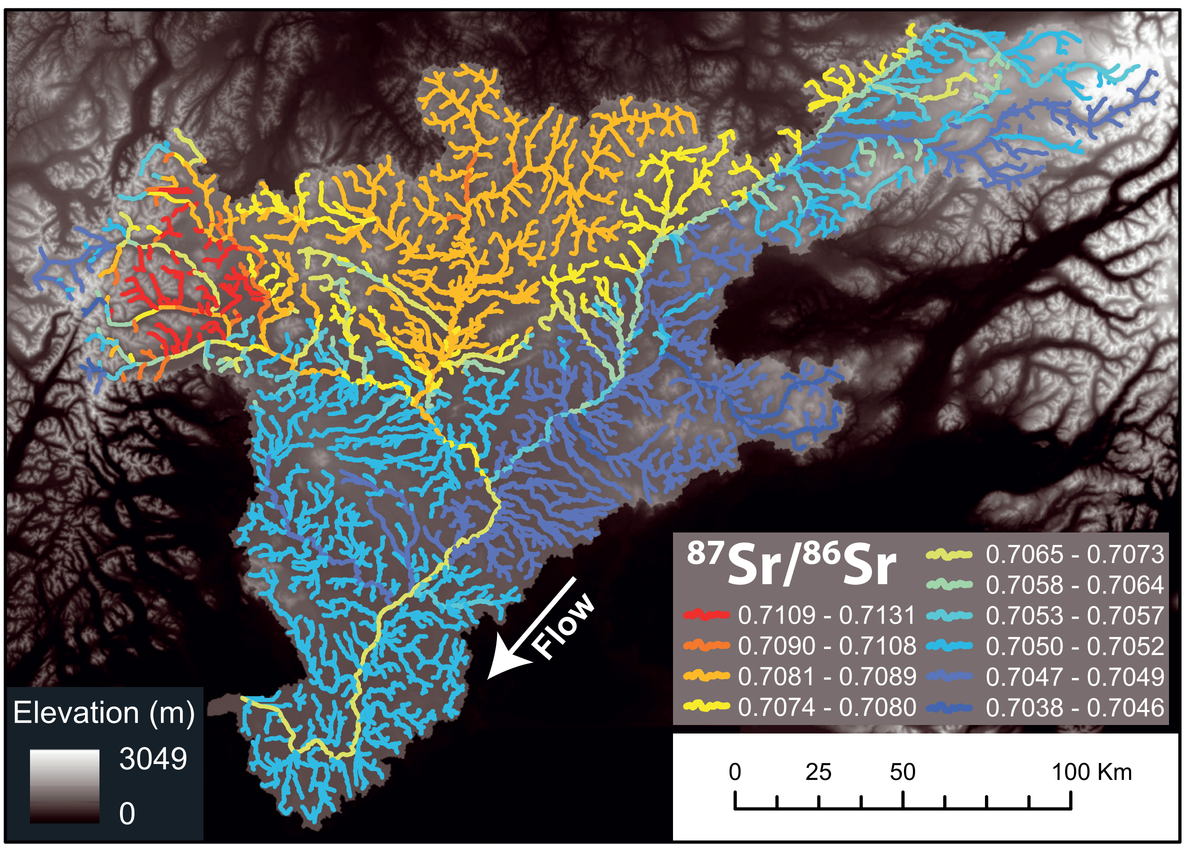
[**Figure 5:** A comparison of a domain map (left) and a contour map (right) made using the same strontium ($^{87}Sr/^{86}Sr$) isotope data for France. Adapted from Willmes et al. 2018, figs. 5 and 8.]

The usability of the isoscapes produced through contour mapping varies. Kriging has been criticized because it can extrapolate data beyond the data's original limits, adding uncertainty in provenance studies (Hoogewerff et al. 2019, p. 1040). Similarly, Adams et al. (2019, pp. 243-245) test their contour-mapped isoscapes against each other and against their Voronoi map, finding that although the simple kriging and empirical Bayesian kriging interpolated surfaces suggested lower error margins, the cokriged surface honored the heterogeneity of the data better and predicted values better (see fig. 6). They note, however, that interpolation requires spatial autocorrelation, a condition that often does not apply to lithological zones, which may be sharply delineated and abut zones with very different values. Interpolated models thus smooth the data, disguising the heterogeneity that may exist in the biosphere (Adams et al. 2019, p. 245). Data smoothing is a major issue of contour mapping, as the modelled interpolation creates gradual gradient boundaries between areas that produce different strontium isotope ratios. These progressive boundaries are not representative of the underlying lithologies that have a dominant role in dictating bioavailable strontium. Lithological boundaries are sharp rather than graduated and primary analysis has demonstrated that very different value can be produced in close proximity as a result (e.g. Evans et al. 2018a; Madgwick et al. 2019b). As a consequence of these issues Adams et al (2019, p. 247) ultimately conclude that their unmodeled isotope data rather than any of the geostatistical methods they tried would provide more reliable outcomes for provenancing human remains for their area of study.



[**Figure 6:** A cokriged contour map of Cape York, Australia (left) and the cokriged error (right). Note the relatively high error. Adams et al. 2019, p. 245, fig. 12, used with permission.]

Finally, contour mapping methods that rely on Euclidean distances may not work well when the target of the study is aquatic. For example, point A and point B in a stream may be close to each other when measured "as the crow flies" but point B may be substantially downstream from A and have additional inputs. Thus, if the goal of the study is to understand provenance in aquatic specimens, a Euclidean contour map is unlikely to produce a relevant model. For these cases, dendritic network models that reflect how strontium is carried in surface waters have been shown to be more effective (Brennan et al. 2016, see fig. 7).



[**Figure 7:** A dendritic network model showing the non-Euclidean relationships among $^{87}Sr/^{86}Sr$ values across the hydrological system. Adapted from Brennan et al. 2016, fig. 3, used with permission.]

3.3 Machine learning

Machine learning is increasingly being used to combine different sources of strontium isotope data to provide isoscapes for more of the globe than has been addressed through domain and contour mapping (Bataille et al. 2018; Bataille et al. 2020). Machine learning appears to produce more accurate results than contour mapping methods. For example, Bataille et al. (2018) tested random forest regression against other machine learning, linear, and non-linear regression models including generalized regression boosting tree, cubist regressions, and ordinary kriging. They found that random forest regression was the most accurate and that machine learning algorithms in general perform significantly better than linear and non-linear regressions at predicting $^{87}Sr/^{86}Sr$ variations over Western Europe. They hypothesize that this is due to their ability to use both categorical and continuous predictor variables and from their insensitivity to non-normally distributed data and outliers in the training dataset. Machine learning does not always outperform methods like linear regression, however. In a recent study from the GEMAS project, random forest models were found to be better overall, but not for some bedrock types, which "serves as a reminder that RF does not always outperform classical regression" (Hoogewerff et al. 2019, p. 1040). Similarly, marked variation was noted between the modeled and measured maps produced for certain areas in this study, such as southern Norway and the Baltic (Hoogewerff et al. 2019, p. 1043). This highlights the importance of primary measured data and warns against modelling on the basis of datasets with sparse coverage.

An advantage of machine learning is that it allows for further refinement of isoscapes by modeling the data and then combining the measured and modeled data (Hoogewerff et al. 2019; Bataille et al. 2020). This approach can result in more diverse values in some areas

(Hoogewerff et al. 2019), which is problematic for using the models for provenancing. A machine learning approach can also guide and improve future sampling, however, and areas where the measured and modelled results have noticeable discrepancies can be prioritized for additional samples (Hoogewerff et al. 2019, p. 1040-1041).

One potential drawback to machine learning approaches is that the isoscapes generated may not be suitable for all applications. As discussed above, different strontium archives reflect bioavailable strontium in different ways, and not all archives are suitable for all research goals. For example, the isoscape generated by Bataille et al. (2020) incorporates many samples taken by the GEMAS project that targeted modern agricultural and grazing lands. While useful for modern food science studies, these are areas that archaeologists try to avoid when biosphere sampling to minimize the effects of recent anthropogenic changes to soil composition. It is possible that, as machine learning approaches develop, platforms may allow researchers to select the archives they wish to include in isoscape generation.

Another drawback to machine learning approaches such as random forest regression is that they can be computationally intensive. For large datasets with numerous covariates, computational requirements may begin to exceed the capabilities of desktop computers (Bataille et al. 2020), making these isoscapes difficult to produce for large areas. Methods of production are also inaccessible to most researchers.

In general, machine learning provides greater opportunities for generating isoscapes across large areas, for mapping areas that currently have few empirical samples, and for usefully combining archives. However, isoscapes produced by machine learning still have to be trained with accurate local data to be useable to answer specific questions. General basemaps produced by machine learning can be calibrated for more specific studies using new isotope data, resulting in more accurate maps than could be achieved with either the general basemap or a basemap constructed from the newly collected data alone (Bataille et al. 2018; Bataille et al. 2020).

Despite the great promise of mapping using machine learning, it is essential to remember that the maps produced can only be as good as the data used to produce them. While it can be encouraging to see strontium ratios mapped across large areas of the globe, these maps are likely to be less accurate for areas where there are few empirical samples. Like all isoscapes, those produced by machine learning should not be applied uncritically. Researchers should query whether an isoscape was modeled from sufficient, appropriate empirical data to be relevant to their specific question. It is also essential to consider the possibility that additional sampling may be needed to refine the isoscape to meet their needs.

3.4 Incorporating geological data

Expected strontium ratios have sometimes been modeled directly from bedrock ages and lithologies (Beard and Johnson 2000; Duxfield et al. 2020); however, many recent studies prefer using archives that more directly reflect bioavailable strontium. Although strontium isotope ratios in soils ultimately derive from parent bedrocks, those parent bedrocks may not underlie the soils. For example, a strontium isotope study in the Basin of Mexico (Pacheco-Fores et al. 2020) found that local differences in strontium ratios could be detected in the Basin of Mexico despite the underlying geology being mostly similar, probably due to different erosional processes bringing different components from areas outside the Basin and depositing them in particular places.

In some areas, strontium ratios between underlying bedrock and superficial deposits may not be significantly different (Snoeck et al. 2020). In these areas, mechanistic models that predict the $^{87}Sr/^{86}Sr$ ratio in soils by applying a radiogenic equation to geochemical knowledge of bedrocks can be applied (Bataille et al. 2020). Mechanistic models have the advantage that bedrock geology is fairly well known globally, so this method does not require additional expensive sampling to build an isoscape. Such mechanistic modeling has been increasingly improved in successive studies through machine learning and the addition of empirical data (Bataille et al. 2014; Bataille et al. 2018; Bataille et al. 2020).

Other issues with incorporating geological data into isoscapes include the fact that older rocks have been found to have increasingly diverse $^{87}Sr/^{86}Sr$ ratios (Bataille et al. 2020), making it difficult to predict their $^{87}Sr/^{86}Sr$ ratios in the absence of testing. Additionally, differential weathering means the soils overlying rocks of the same age may have different $^{87}Sr/^{86}Sr$ ratios. This can also be a problem for incorporating geological data into machine learning models because these models have so far relied on age without addressing weathering (cf. discussion in Bataille et al. 2018). It may therefore be necessary to sample heavily weathered bedrocks at a higher density (Bataille et al. 2018, p. 19) and incorporate this empirical data into models. Willmes et al. (2018) take another approach to incorporating geological data, using cluster analysis to identify groups of isotope ranges, with lithology rather than age proving to be the best predictive variable. They then use this data as covariate in developing an isoscape using kriging with external drift, which improves the accuracy of the maps made without the geological isotope groups.

3.5 Research design: samples, mapping method, and research questions

Given that relatively small differences in strontium isotope ratios can have considerable interpretive potential and that bioavailable strontium can vary widely at a local scale, project-specific isoscapes are most desirable. It may be possible to work toward isoscapes that can be applied in studies across disciplines through the combination of machine learning and local sampling. The modeled isoscapes produced by machine learning have been shown to improve the accuracy of new modeled isoscapes when they are used as one of the sources of strontium data in the new model (Bataille et al. 2020). The best solution is probably to continue modeling more refined “base” isoscapes through machine learning while recognizing that these need to be refined through local sampling targeted at the particular question researchers are seeking to answer. This iterative process of refinement will reduce ambiguity and increase resolution over time. Modeled isoscapes play a key role in characterizing variability on a scale that is challenging to achieve through primary sampling.

4. Interpreting strontium data against isoscapes

Interpreting the results of strontium analyses against an isoscape to determine provenance is never straightforward. Similar strontium ratios are found in wide-ranging locations and equifinality is a constant barrier to interpretation. Almost no strontium ratio is unique and strontium isotope ratios are therefore better at eliminating possible areas of origin than identifying them (Montgomery 2010, p. 336-337; van der Jagt et al. 2012). In short, the approach should be considered as exclusive and is invariably most effective if it can be combined with other proxies (Madgwick et al. 2019a).

The two main methods of interpreting strontium results, the calculation of residuals (or basic comparison of biosphere and sample values) and the use of Bayesian statistics, are outlined and illustrated in Pouncett (2020). Either method is generally more straightforward when provenancing the source that was used to build the isoscape, such as when wines of various origins are compared against wines of known origins (Durante et al. 2015). Complexity increases when there is a disconnect between the material to be provenanced and the source of the isoscape, as when human remains are provenanced against isoscapes produced by sampling plants. Source material (e.g. humans and large mammals in archaeology) is often not suited to the production of isoscapes and therefore this disconnect is a common problem.

The isoscapes produced by domain mapping, contour mapping, and machine learning all include uncertainty and error, and these errors are expressed in different ways. In domain mapping, a domain is defined geographically, and sample data are grouped and statistically assessed for each domain. The domains will therefore have differing uncertainties. In the interactive biosphere map of Evans et al. (2018b), the IQR of sample $^{87}Sr/^{86}Sr$ is allocated to each domain such that when the map is interrogated, it will give a positive indication if the input value falls within the IQR. The data held for each domain can be accessed by clicking on cells within the domain. It is important to note that this method focuses, to a varying extent depending on the statistical parameters used, on the central tendency of the datasets rather than reflecting the full range of data via the extreme data points. This is because the data range of the sampled material (predominantly plants) is greater than ranges for the material they are used to interpret (i.e. predominantly tooth enamel). Snoeck et al. (2020) use median absolute deviation (MAD) to estimate the variation. This is the median of the difference between the value of each sample and the median for the lithological zone from which it derives. They produce maps for $^{87}Sr/^{86}Sr$ compositions and MAD that should be used in parallel when exploring origins.

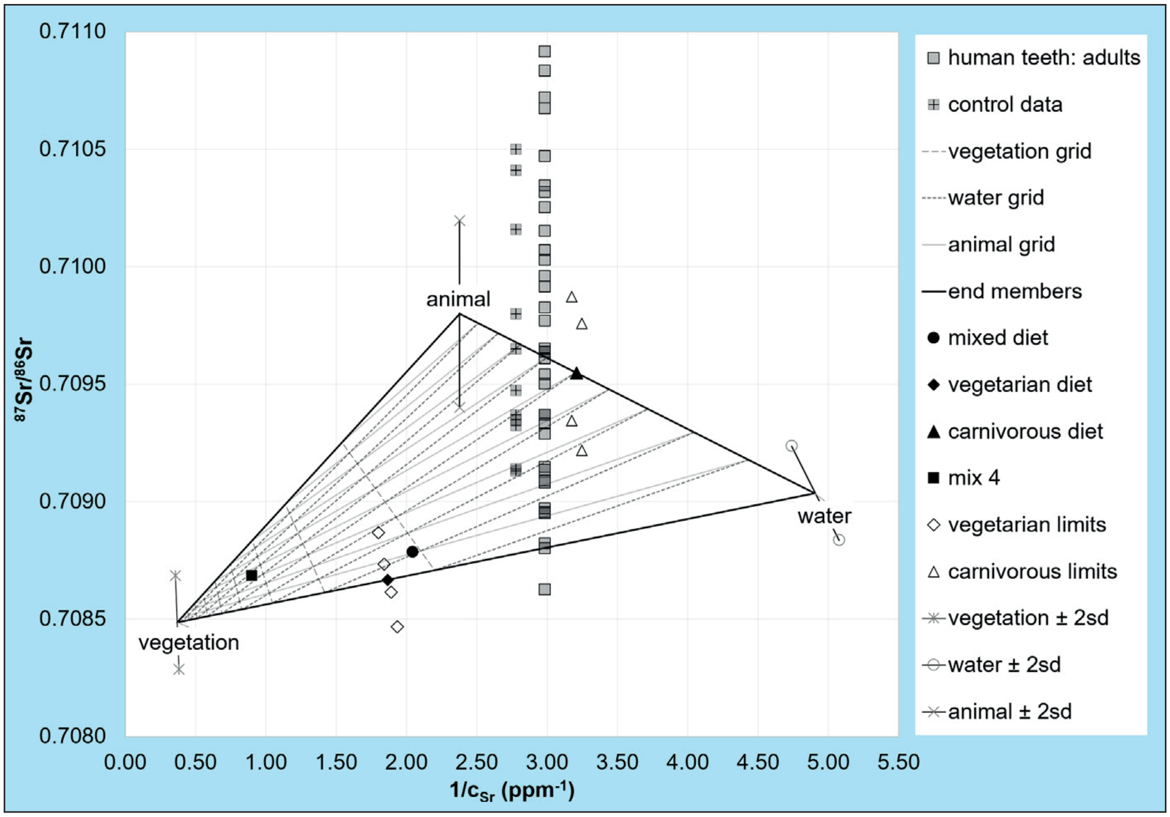
Contour maps are produced either through inverse distance weighting (Emery et al. 2018) or some form of kriging (Adams et al. 2019; Wang et al. 2018; Willmes et al. 2018). Prediction error surfaces for the maps can then be produced by plotting the root-mean-square error (RMSE; Adams et al. 2019; Willmes et al. 2018); some studies also use mean absolute error (MAE; Wang et al. 2018). Different types of kriging are often compared in an effort to minimize the RMSE (Adams et al. 2019; Willmes et al. 2018); however, low RMSE does not always mean better prediction (Adams et al. 2019). The error of a contour map is generally presented alongside the map itself as a second, shaded map. This allows researchers who are applying the contour map to assess how accurate the map is for the area they are trying to match. As with domain maps and MAD maps, contour maps and RMSE maps must be used together, making the error map an essential component of the publication of a contour mapped isoscape.

Machine learning methods have used different approaches to error and uncertainty in refining models, expressing data, and presenting isoscapes. RMSE (Bataille et al. 2018; Bataille et al. 2020) and mean/maximum absolute error (Hoogewerff et al. 2019) between observed and predicted values have been used to optimise models and to compare the predictive performance of different modelling methods. In terms of expressing spatially explicit uncertainty, Hoogewerff et al. (2019) use two error sources (standard deviation of site-specific measured $^{87}Sr/^{86}Sr$ variance and random forest model residuals) to plot predicted standard deviations across a map of Europe. Bataille et al. (2020) build a function into models based on mean absolute residual values to predict standard deviations in different locations. This is relatively broad-brush and, as in Hoogewerff et al. (2019), reflects a general increase in uncertainty with increasing predicted $^{87}Sr/^{86}Sr$ (although uncertainties are not presented on a map). Areas with higher $^{87}Sr/^{86}Sr$ also frequently have fewer primary data, again emphasising the need for denser sampling. This approach to global mapping differs from that taken by Bataille et al. (2018) for Europe, in which inter-quartile ranges (IQR) are predicted for lithological zones. This is more fine-grained but also more computationally intensive. The predicted IQR can also be cross-checked against plotted standard deviations of primary analyses of different substrates from the same location on a bubble map of Europe (Bataille et al. 2018, p. 10).

As the discussion above shows, researchers using strontium isotopes to assign provenance need to critically assess the general approach to uncertainty or error of the isoscape they are using as well as the specific uncertainty or error associated with their region of interest. For clarity and transparency, it is advisable that researchers state the uncertainty or error of each isoscape region to which they are assigning provenance.

After accounting for uncertainty or error in the isoscape itself, strontium isotope analysis suffers from additional interpretative issues. For example, the biogenic reservoir of strontium represents an average of dietary resources (and thus locations) and this incorporates in tissues at an undefined, but relatively slow rate. Montgomery et al. (2010) found that incorporation in cattle molars occurred over a period in excess of 12 months, even when sampling at a microscopic scale. This gradual incorporation complicates interpretation. For example, if an animal feeds equally across a lithological boundary in two areas with differing bioavailable strontium, their averaged biogenic signature will not represent either location, and interpretation may therefore focus on a different area entirely. The potential for these averaged ratios to occur creates difficulties for provenancing human remains using isoscapes built from animal remains, whether modern or archaeological, with uncertain feeding regimes.

Mixing models have been proposed as a more accurate way of modeling expected human strontium isotope ratios for given areas (Lengfelder et al. 2019; Montgomery et al. 2007, see fig. 8). Because human diets are varied and do not consist of any single input, a model that mixes inputs is hypothesized to provide a better estimation of local strontium isotope signatures for humans. However, this method was found to greatly underestimate the range of strontium isotope signatures in humans that were considered likely to be local based on their very young ages (Toncala et al. 2020). The authors conclude that mixing models should not be applied and that local origins are better estimated from the population itself if it is large enough to support a Kernel density distribution analysis, an approach that can only be applied to archaeological populations. Whether mixing models can be useful in creating isoscapes for modern forensic analysis remains unexplored; however, the results of Toncala et al. (2020) suggest that it would not provide a more accurate range than modeling from other strontium archives.

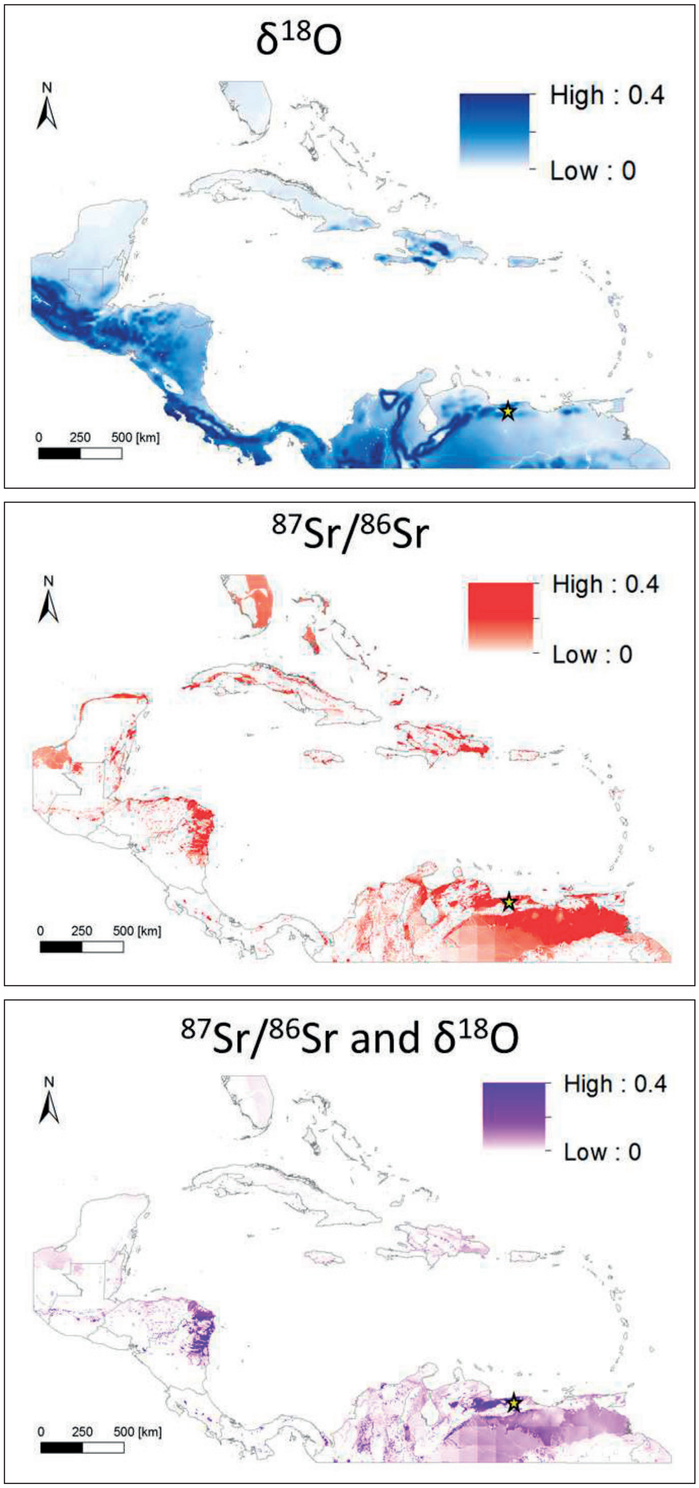


[**Figure 8:** An example of a mixing model. Toncala et al. 2020, fig. 5.]

Whatever archive is used to construct an isoscape, the complexity of human diet poses challenges for identifying when an individual is local. Because plant foods have a much stronger effect on the overall strontium ratio than animal foods (Burton and Hahn 2016), consuming a large proportion of imported plant foods could make an individual appear non-local even if they never left the area where they were born (Larsson et al 2020; cf. discussions in Grumbkow et al. 2013; Price et al. 2006). In addition, diets rich in marine resources have a major impact on biogenic strontium, averaging terrestrial values with the marine signal of 0.7092 (Lahtinen et al. 2021). Therefore, it is important to temper interpretation with dietary isotope proxies such as carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$) and sulphur ($\delta^{34}S$) (Madgwick et al. 2019a).

The globalization of food supply networks in the recent past may introduce complications for provenancing modern forensic samples as opposed to archaeological samples. The more non-local plant foods a person consumes, the less likely their strontium ratio is to match that of the area where they grew up. While imported foods may sometimes affect the strontium ratios of wealthy individuals in the past, making them appear non-local when they were not, this phenomenon is likely to be quite common in modern populations in developed nations where much of the food consumed is not produced locally. However, some modern forensics indicate that local bioavailable strontium is still expressed in human remains despite globalized dietary practice (Degryse et al. 2012). This issue still needs careful consideration and highlights the need for multiple isotopes for provenancing, especially in modern forensic studies.

Researchers have taken different approaches to provenancing remains to try to overcome interpretation challenges. Laffoon et al. (2017) tested both a simple Interval Approach and a Likelihood Approach (see fig. 9) as statistical methods of determining human provenance for three dental samples (one modern sample of known origin and two archaeological samples of unknown origin) in the Circum-Caribbean region. While the two methods gave broadly similar results, the authors argue in favor of the Likelihood Approach as more applicable because it does not enforce an artificial yes/no binary for each division of the map. Hoogewerff et al. (2019, p. 1041-1042) suggest a similar, likelihood-based interpretation approach where strontium isotope results are matched to the values of grid squares, with the more common a value is, the less significant the match would be as evidence of provenance. To apply this approach, they adapted an accepted Bayesian likelihood ratio approach to develop a tool that produced common source likelihood ratio maps. While the specific methods differ among studies, the use of likelihood approaches for interpreting strontium isotope results is effective because it acknowledges the fact that strontium isotope ratios are almost never unique and may have broad and/or discontinuous distributions within a region. Incorporating probability statistics in assigning provenance is a useful development and one that should be further advanced in the future.



[**Figure 9:** An example of a likelihood approach to using isotope results for provenance identification of a specimen. This study evaluated both the $\delta^{18}O$ (top) and $^{87}Sr/^{86}Sr$ (middle) for the specimen and cross-referenced the results (bottom) to narrow the possible areas of provenance. Adapted from Laffoon et al. 2017, fig. 5.]

Finally, it is important to note that isoscapes capture bioavailable strontium at the moments in time that produced the analyzed archives. Isoscapes produced from archaeological materials may not be applicable in forensics or food science studies. Similarly, isoscapes constructed from modern plants or animals may not be applicable in archaeological studies, particularly if major environmental changes are likely to have occurred. For example, large shifts in surficial sediments occurred during the Ice Ages, meaning that strontium isoscapes made from contemporary data must be used with caution when provenancing remains from the distant past (cf. Willmes et al. 2018).

5. Discussion and conclusions

Approaches to strontium biosphere mapping have developed and diversified markedly in recent years. There has been an important shift in focus to a greater emphasis on creating and enhancing isoscapes, rather than an overwhelming investment in provenancing samples, as has previously been the dominant trend in research. However, the process of mapping bioavailable strontium is yet to reach full maturity. This discussion provides recommendations for best practice and suggests profitable directions for future research.

Firstly, strontium isoscapes for any region cannot be considered universal across the research spectrum. The basis for these isoscapes and the nature of their creation must be carefully considered and applicable isoscapes must be selected (or generated) dependent on research objectives. The validity of isoscapes, in terms of resolution, uncertainty and relevant archives, must be aligned with research questions and additional primary mapping will often be necessary. While it may be possible to exclude samples via broad categories like “potentially local” and “non-local” based on existing isoscapes, many projects will have more specific research goals. Very few isoscapes comprise a sampling density that can be relied on to provide a precise local range in areas of more radiogenic lithologies (e.g. >0.710, see Madgwick et al. 2019b), though younger lithologies tend to be much better resolved.

Addressing more specific research goals will require careful consideration of the strengths and weaknesses of the strontium archives as well as the mapping method(s) used to build the isoscape. Machine learning provides opportunities for researchers with the correct skillsets to refine existing isoscapes rather than starting from scratch; however, refining existing isoscapes requires additional strontium sampling. Researchers will still have to consider the most appropriate strontium archives to sample and the most useful geographical distribution of those samples. In almost all landscapes, researchers should assume that additional sampling will be necessary to achieve their research goals and biosphere sampling should be incorporated into research designs and budgets at the outset. These incremental advances in primary mapping will create higher resolution, more reliable isoscapes, even at the local level, in the long term.

Secondly, researchers should recognize that, even with an applicable isoscape, interpreting the results of strontium isotope analysis is not straightforward. Analyzed strontium isotope ratios cannot simply be matched to an isoscape. Strontium isotope ratios are almost never unique even at a regional scale, meaning that strontium isotope analysis alone is usually insufficient to answer research questions. Multi-isotope studies have been found to be more effective than single isotope studies for provenancing biological materials (Bartelink and Chesson 2019, p. 31; Fernández-Crespo et al. 2020; Koehler et al. 2019; Laffoon et al. 2017; Madgwick et al. 2019a, Madgwick et al. 2019c). Multi-isotope studies allow researchers to refine provenance as each proxy allows some areas of the landscape to be excluded as potential areas of origin. Not all isotopes are applicable in all areas or for all target samples, however, and researchers need to design multi-isotope studies around specific landscapes and research questions. Trace elements provide alternative proxies that can be used in conjunction with strontium isotope analysis for narrowing possible areas of origin (Asfaha et al. 2011; Shaw et al. 2010; Shaw et al. 2011).

Applying a likelihood approach to all isotope results and comparing the outcomes is one of the best methods for identifying possible areas of origin.

Finally, it should be stressed that strontium isotope evidence and isotope evidence in general should be interpreted in conjunction with other evidence when available, particularly in fields such as archaeology and forensics (Makarewicz and Sealy 2015). Wider proxies (e.g. associated provenanced artefacts in archaeology) can aid in guiding interpretation when equifinality remains a problem (e.g. Scorrer et al. accepted).

Future directions in strontium mapping research should focus on increased sampling of carefully targeted archives, especially in areas with sparse existing coverage or high diversity (generally areas of radiogenic lithology). In addition, the wider application and refinement of machine learning mapping methods will be of benefit, including regular iterations with the inclusion of new primary data. Current studies indicate a need for both refining contour mapping and machine learning techniques for building isoscapes as well as collecting more empirical data. There is a general consensus that large-scale isoscapes built using predictive models, geospatial statistics, and machine learning still need to be used in conjunction with local isotope data to be effective at answering specific questions (Bataille et al. 2018; Bataille et al. 2020; Willmes et al. 2018). Uncertainties in large-scale isoscapes can guide the collection of empirical data, creating a positive feedback loop between better models and targeted data collection.

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