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Mapping biosphere strontium isotope ratios across major lithological boundaries. A systematic investigation of the major influences on geographic variation in the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of bioavailable strontium above the Cretaceous and Jurassic rocks of England.

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CHAPTER 1

INTRODUCTION

Within archaeology the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of skeletal tissue (chiefly, dental enamel) is used to provide direct biological evidence for the geographic origins of people and animals. This application of isotope analysis is based on the assumption that the strontium that is present within the food-chain ultimately represents a rock-weathering product. Governed by the radioactive decay of ^{87}Rb to ^{87}Sr , geochemical differentiation within the lithosphere (i.e. the crust and upper-mantle of the Earth) and the on-going rock cycle have produced numerous, isotopically contrasting reservoirs of strontium, the compositions of which are reflected within the biosphere. As strontium is not thought to fractionate significantly during low-temperature processes, systematic geographic variations in $^{87}\text{Sr}/^{86}\text{Sr}$ composition develop between biota, because of the mixing of strontium derived from locally available nutrient sources. Essentially, archaeologists seek to compare skeletal isotope data with $^{87}\text{Sr}/^{86}\text{Sr}$ ranges that are characteristic of the ecosystems that have developed above contrasting rock formations.

The surface geology of the British Isles effectively encompasses the entire history of the Earth's crust. Thus, when applied within this intensely variable geological setting, $^{87}\text{Sr}/^{86}\text{Sr}$ analysis potentially provides numerous opportunities to investigate questions related to residential mobility and past landscape use. However, human and animal variation in $^{87}\text{Sr}/^{86}\text{Sr}$ composition cannot be accurately constrained using geological maps. Firstly, as some minerals weather more rapidly than others do, the strontium released from a poly-mineralic substrate is not likely to resemble the bulk $^{87}\text{Sr}/^{86}\text{Sr}$ composition of that material. Secondly, most terrestrial ecosystems are supported by soils rather than bedrock or raw drift deposits, and these may become enriched in the most stable rock minerals, with respect to underlying geology. Finally, the strontium that is circulated within an ecosystem is likely to incorporate some form of atmospheric input, which may contrast with local geogenic $^{87}\text{Sr}/^{86}\text{Sr}$ values.

This thesis builds on a substantial body of work, which has established 'biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation' as the key concept in understanding the influence of environmental factors on the distribution of strontium from different sources within the surface environment (Price *et al.* 2002). The biosphere is characterised by ongoing processes of recycling, involving living organisms (Vernadsky 1926). Plants are particularly important mediators of these cycles, affecting the chemical availability and physical distribution of elements locked within the lithosphere. They drive, among other processes, Goldschmidt's plant-pump, enriching and redistributing certain chemical elements at the Earth's surface (Goldschmidt 1937).

Across the British Isles the reported biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values obtained from vegetation cover a substantial national range, from as low as 0.7050 above the Tertiary (65–2.6 Ma) volcanic terrain of the Isle of Sky (Evans *et al.* 2009), to 0.7251 above some schistose Proterozoic (2500–542 Ma) rocks in Scotland (Evans *et al.* 2010a). Broad regional contrasts can be detected between major geo-tectonic divisions of the landmass, such as the ancient Precambrian and Palaeozoic upland terrain of Scotland and Wales (dominated by igneous and metamorphic rocks), the Palaeozoic formations of northern England, and the recent Mesozoic and Cenozoic sediments of the English lowland zone. Such divisions link lithology and structure, to relief and weather patterns. These variables have a profound influence on the distinctive character of regional environments. Thus, within each broad bio-geographical package there may be considerable potential for biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation related to transport and weathering processes as well as lithology and bedrock age (Evans *et al.* 2010b).

An understanding of the degree to which it is possible to characterise and resolve ambient biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation within a given geological terrain is a key step towards realising the full potential of $^{87}\text{Sr}/^{86}\text{Sr}$ analysis. Even so, it would be naive to assume that even the most detailed biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ map would allow individuals to be matched to a particular geographic location, as no given range of values is likely to be geographically

unique. Thus, $^{87}\text{Sr}/^{86}\text{Sr}$ analysis cannot provide positive evidence that any one individual is of local origin, but it can be assumed that individuals that fall outside a given range of values are unlikely to be local. Although, accurate and increasingly precise biosphere constraints may reduce the frequency with which local individuals are misidentified as non-local, it is likely that any local range will incorporate both geographically and geologically non-local individuals.

When attempting to move beyond the identification of occasional, geologically exotic people and animals the potential insights into past life-ways that $^{87}\text{Sr}/^{86}\text{Sr}$ analysis may be able to provide are dependent on understanding biosphere variation at an appropriate geographic scale. For example, if a large proportion of individuals within a mortuary assemblage exhibit comparable $^{87}\text{Sr}/^{86}\text{Sr}$ compositions, does this mean that these represent the dead of a corporate community that exploited a geographically limited resource-base, or disparate and independent family groups that exploited a much wider, but isotopically homogenous terrain? That is to say, differences in the $^{87}\text{Sr}/^{86}\text{Sr}$ characteristics of archaeological skeletal assemblages cannot be attributed to differences in social organisation unless the underlying character of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation is also understood. Without access to some form of external control it is not possible to construct hypotheses that are both robust and testable, within the context of the archaeological record.

This thesis focuses on resolving geographic biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation above Mesozoic rocks within the lowland zone of England (e.g. Rackham 2000: 3, Figure 1.3). The bedrock geology spans the Jurassic and the Cretaceous periods and is characterised generally by an alternating succession of carbonate-rich limestones, and clay-rich mudstones. The outcrops fall within a mild, temperate bioclimatic zone and generally support productive agricultural soils (Hodge et al. 1984; Jarvis et al. 1984). Unlike impermeable and acidic, hard-rock formations they are associated often with favourable preservation characteristics for archaeological skeletal tissues

(Campbell *et al.* 2011). In addition, the outcrops underlie some of the most populous regions of the UK, leading to pronounced, and widely acknowledged recovery biases in the archaeological record – e.g. see discussion of “Distribution of sites” in Hambleton (2008: 12–16). Closely related formations are also present across mainland Europe. Accordingly, an understanding of the character of biosphere variation above these rocks is of vital importance to the wider application of $^{87}\text{Sr}/^{86}\text{Sr}$ analysis within the archaeological record.

1.1 Aims and objectives

This research project investigates the levels of variation in biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ composition that occur at different geographic scales within the modern surface-environment. The overall aim is to elucidate the character of systematic biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation, and determine at what geographic scale it is possible to describe this in relation to underlying geology. Specific research aims and objectives are summarised below.

Aim 1: **To identify the most appropriate sampling and analytical techniques for biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ characterisation and mapping.**

Objectives:

- a) Based on a review of the literature, identify priority sampling strategies and biosphere sample media.
- b) Develop appropriate methods of sample preparation capable of producing reliable data applicable to the characterisation of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values.
- c) Compare $^{87}\text{Sr}/^{86}\text{Sr}$ values produced by different modern biosphere proxies, collected in a consistent manner at proximal locations.

Aim 2: To assess the benefits of using high-density and low density sampling methods to characterise geographic biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation.

Objectives:

- a) Select and analyse environmental sample media collected within a restricted study area, underlain by a series of contrasting solid geological formations, so that biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values can be compared at a local scale.
- b) Select and analyse samples collected across a wider region underlain by geological formations related closely to those identified in the course of achieving Objective a.
- c) Compare the sampling constraints and resulting ranges of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values obtained under the two sampling regimes.

Aim 3: To evaluate the influence of weathering, erosion, and sediment transport mechanisms on patterns of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation related to underlying geology.

Objectives:

- a) Select and analyse samples collected within alluvial systems capable of integrating a number of local sediment sources, so that these can be compared with $^{87}\text{Sr}/^{86}\text{Sr}$ values more closely associated with specific geological formations (see Aim 1, Objective a).
- b) Select and analyse samples collected above glacial till, in-situ Pleistocene weathering deposits and recent marine silts, so that a comparison can be made with biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values predicted on the basis of underlying solid geology.

Aim 4: To evaluate the potential influence of atmospheric transport on patterns of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation.

Objectives:

- a) Measure and compare biosphere sample media affected by different modes of atmospheric deposition to determine whether short-term inputs of atmospheric aerosols are likely to have a significant effect on biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values.

- b) Measure rainwater collected over a period from one location, in order to assess the degree to which the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of wet deposits is likely to represent a distinctive environmental end-member.
- c) Measure rainwater that has been collected at different locations across the UK in order to determine on what scale the $^{87}\text{Sr}/^{86}\text{Sr}$ compositions of wet deposits are likely to vary geographically.

Aim 5: To understand the implications of modern patterns of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation for the interpretation of archaeological human and faunal data.

Objectives:

- a) Within the area in which high-resolution biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ data are available (see Aim 1, Objective a), identify archaeological sites in locations that are thought to be associated with the exploitation of local resources, so that the $^{87}\text{Sr}/^{86}\text{Sr}$ of faunal material can be compared directly with modern biosphere values as a ground-truthing study.
- b) Analyse human material recovered from a burial context associated closely with a particular geological domain, so that these values can be compared with appropriate biosphere values and existing human data.

1.2 Thesis structure

In Chapter 2, this thesis begins with a literature review that focuses on the attempts that have been made to map and characterise biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ values for archaeological applications (Section 2.1). This is complemented in the sections that follow by a review of the sampling strategies that have been applied within the British Isles for the purposes of geochemical mapping (Section 2.2) and the reasons why certain sample media are used (Section 2.3). Geochemical mapping represents a mature area of research with a well-developed theoretical and metrological foundation that is used to support the biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ characterisation and mapping studies reported in later chapters of this thesis.

The methods of sample preparation and analysis developed for this project are detailed in Chapter 3. In the mapping and characterisation chapters that follow, the processes that influence biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ variation are investigated broadly in ascending order of geographic scale. An assessment of local variation using a high-density sample archive (Chapter 4) is followed by an investigation of broader geographic trends using low-density data (Chapter 5). Weather systems are potentially capable of transporting strontium across geological boundaries, over continental or even global distances; the influence of this process is evaluated in Chapter 6. The interpretive implications of an enhanced understanding of biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ are explored using faunal (Chapter 7) and human (Chapter 8) data.