Science of the Total Environment 839 (2022) 156083

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The forest effect: Biosphere $^{87}Sr/^{86}Sr$ shifts due to changing land use and the implications for migration studies

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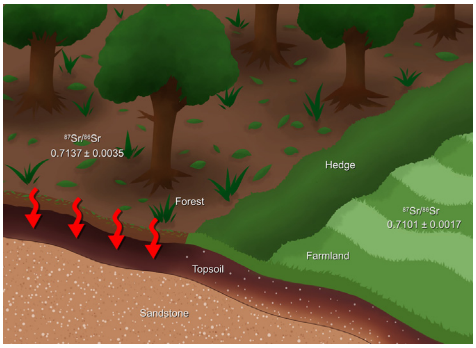
HIGHLIGHTS

- Long term forestation changes the $^{87}Sr/^{86}Sr$ isotope signature of the biosphere.
- Forests create acidic soil conditions that leach out the carbonate component.
- This may lead to erroneous interpretations of past provenance.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Mae Sexauer Gustin

Keywords:
Sr-isotopes
Bioavailable
Ancient forests
Deforestation
Pannage

ABSTRACT

This study documents a transect of $^{87}Sr/^{86}Sr$ values from a variety of plant, soil and rock samples across the ancient woodland of the Sherwood Forest National Nature Reserve (SFNNR) and into adjoining farmland in Britain. All samples were collected from the Triassic Sherwood Sandstone Group. A shift of +0.0037 in $^{87}Sr/^{86}Sr$ values is observed between the average plant from the biosphere of the ancient forest and that of the farmland. This shift is caused by the leaf litter accumulation in the forest, through time, leading to soil acidity that leaches out the carbonate component of the soil. This results in the forest floor soil reflecting only the silicate minerals from the original Sandstone rock formation. We have named this process "the forest effect". Rock samples from boreholes of the Sherwood Sandstone Group, as well as water samples from aquifers and mineral waters from previous studies, further indicate that the change in biosphere $^{87}Sr/^{86}Sr$ is a result of the wooded environment rather than the anthropological addition of lime to farmland.

The extent of the forest effect will vary with differing lithologies with the most susceptible terrains being those with mixed carbonate-silicate composition, and it may be sufficient to impact the interpretation of animal and human $^{87}Sr/^{86}Sr$ in studies of mobility and migration. The model provides an opportunity to understand and assess food procurement strategies and animal management practices in the past, as well as the interaction of humans with their natural environment.

1. Introduction

Understanding the geographical variation of strontium isotope ratios ($^{87}Sr/^{86}Sr$) available in the biosphere is a key issue when using strontium (Sr) isotope analysis to track the movement and provenance of modern and ancient humans and animals. The characterisation of Sr inputs to a local environment enables bioarchaeologists to define local biosphere ranges and determine whether they have migrants in their human and animal populations or assemblages. Since the first map was published defining isotope domains on the Isle of Skye (Evans et al., 2009), several biosphere, or baseline, maps of $^{87}Sr/^{86}Sr$ have been produced across several

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http://dx.doi.org/10.1016/j.scitotenv.2022.156083
Received 24 January 2022; Received in revised form 16 May 2022; Accepted 16 May 2022

regions and countries around the world (Evans et al., 2010, 2018; Frei and Frei, 2011; Hartman and Richards, 2014; Willmes et al., 2014; Laffoon et al., 2017; Adams et al., 2019) and these have become a valuable tool for migration and mobility studies (Holt et al., 2021). Mapping and using this geographical variation relies heavily on the fundamental principle that the ratio of radiogenic ^{87}Sr to stable ^{86}Sr remains essentially unchanged as elemental Sr is transferred from the source rock into soils and then into water and plants at the base of the food-chain. Thus, surface rocks of different ages and lithology will produce geographical variation in the biosphere ^{87}Sr/^{86}Sr and can be mapped by use of proxy samples such as plants, water and animal remains (e.g. Åberg, 1995; Capo et al., 1998; Beard and Johnson, 2000; Bentley, 2006; Evans et al., 2010, 2018).

However, the age and lithology of the underlying geology is not the only influence on the ^{87}Sr/^{86}Sr available in the biosphere. There are several other inputs that need to be considered which can vary depending on location. For example, in temperate environments such as Britain, atmospheric deposition (in the form of heavy rainfall) as well as the sea-spray effect, if located near to the coast, can lead to areas being saturated with a marine ^{87}Sr/^{86}Sr value (~0.7092) instead of reflecting the underlying geology (Montgomery et al., 2003; Bentley, 2006; Evans et al., 2009; Montgomery, 2010). In arid environments the Sr-contribution of wind-blown terrestrial aerosols, or dust, can cycle depending on climate. In particularly dry periods, dust can be transported great distances, potentially inputting exotic ^{87}Sr/^{86}Sr vastly different from the contributions of the local underlying bedrock geology (Capo et al., 1998; Chadwick et al., 1999; Yaalon, 1997; Ganor and Foner, 2001). Human activities such as the application of commercial fertilisers and lime and their Sr-contribution to the ancient and modern biosphereare still being debated (Probst et al., 2000; Åberg, 2001; Vitòria et al., 2004; Bentley, 2006; Frei and Frei, 2011; Maurer et al., 2012; Crowley, 2015; Thomsen and Andreasen, 2019).

One factor that has been commented on in the literature, but not addressed in more recent biosphere papers, is the effect of the plants themselves on the bioavailable Sr in the soil. The long-term growth of trees can deplete the minerals in the soil and lead to a gradual change in the Sr isotope signal being passed into the biosphere. Biosphere ^{87}Sr/^{86}Sr shifts in forest environment were observed by both Åberg et al. (1990) and Evans and Tatham, 2004. Soil profiles from Precambrian granite and gneiss lithologies showed a loss of Ca-minerals through weathering within the first 20 cm depth of the soil (Åberg et al., 1990). This loss was associated with an enrichment in more resistant K-minerals (rich in radiogenic ^{87}Sr) which resulted in higher ^{87}Sr/^{86}Sr values when compared to the deeper soil in their forested study areas. Evans and Tatham, 2004 measured plant and soil ^{87}Sr/^{86}Sr from a well-established woodland site (Locality 9) and these demonstrated an approximate increase of 0.002 compared to an unforested site (Locality 8) on the same Jurassic bedrock in eastern England. Speculative reasoning by the authors suggested that the soil within the woodland site was well-equilibrated and the carbonate component in this soil may have been washed out over time by the slightly acidic conditions of the organic-rich soil leading to higher biosphere ^{87}Sr/^{86}Sr values. These studies also demonstrate why plants such as trees, shrubs and grasses offer the best understanding of the bioavailable Sr that will be consumed by humans and animals at a location.

Organic acid production and increased soil acidity are mechanisms that can influence soil weathering in forests (Dijkstra et al., 2003; Poszwa et al., 2004) and can result in the decalcification seen in the above studies. As most Ca-bearing minerals in the soil have low ^{87}Sr/^{86}Sr values, the depletion in Ca can also be seen as a depletion of low ^{87}Sr/^{86}Sr values, resulting in the biospheres of forests increasing their overall ^{87}Sr/^{86}Sr value with time. Again, this can be observed in Åberg et al. (1990), where the ^{87}Sr/^{86}Sr in the uppermost layers of the soil (0.05–0.10 m) from the mature forest have distinctly higher ^{87}Sr/^{86}Sr values of approximately 0.013–0.001 compared to the younger forest even though they are growing on similar soils. Throughfall, the part of rainfall or other precipitation that falls to the forest floor from the canopy, can also have higher ^{87}Sr/^{86}Sr and Sr concentrations compared to normal precipitation due to the collection of Sr via terrestrial dust from leaf surfaces (Gosz and Moore, 1989; Poszwa et al., 2002; Probst et al., 2000). Differences between throughfall ^{87}Sr/^{86}Sr and normal precipitation ^{87}Sr/^{86}Sr often show an increase greater than 0.001 (Gosz and Moore, 1989; Probst et al., 2000). This can result in the upper part of the soil profile in a forest also being saturated with throughfall ^{87}Sr/^{86}Sr values. The combination of the effects on forest ^{87}Sr/^{86}Sr as a result of organic acid production, increased soil acidity and addition of throughfall, will be here-on in known as the forest effect.

Forested, or woodland, areas are not commonly considered as a specific domain during the baseline mapping of ^{87}Sr/^{86}Sr. In this study, we investigated whether this forest effect could produce significantly different biosphere ^{87}Sr/^{86}Sr values in forests compared to unforested land on the same bedrock geology. A transect of plant, soil and rock ^{87}Sr/^{86}Sr data are presented from across the Sherwood Forest region, one of the oldest ancient forests in Britain.

1.1. Anthropological changes through time: from forest to farmland

There is no escaping the fact that humans have had a major influence on their environment and that land-use has changed over time. Today, agriculture accounts for approximately 75% of all the land-use in Britain (Khan and Powell, 2011). Forest and woodlands were believed to be extensive in the Mesolithic period. They were not necessarily dense, and ranged in form from uneven and patchy to continuous closed canopies, but nevertheless they were present throughout Britain (Rackham, 2006, p.60ff; Whitehouse and Smith, 2010; Noble, 2017). Following the Mesolithic-Neolithic transition and the introduction of agriculture and domestic animal husbandry to Britain c. 4000 BCE, humans began to have a larger impact on the environment through processes of deforestation, forest management and agricultural practices. Several lines of archaeological evidence, pollen analyses and other environmental proxy studies (such as beetle assemblages) show varying stages of deforestation across Britain (Rowley-Conwy, 1982; Darvill, 1987, p.51–54; Greig, 1996; Chambers, 1996; Birks, 1996; Brown, 1997; Robinson, 2000; Bell and Walker, 2005, p.164–168; Rackham, 2006, p.60ff, 77ff; Thomas, 2008; Whitehouse and Smith, 2010; Noble, 2017, p.45–68). For example, it has been estimated that England may have lost 50% of its woodland cover by the Iron Age i.e. c. 800 BCE (Bell and Walker, 2005, p.164–168). From this transition point onwards, British agriculture intensified and expanded.

How much influence deforestation and agricultural practices over the years have had on the Sr-isotope biosphere of Britain is not well known. Validation for using modern proxy data to establish biosphere values for the past has been provided by Evans et al. (2010). This was multi-layered but primarily achieved through the comparability of bone and dentine values, which are highly susceptible to diagenetic alteration and have been shown to equilibrate with labile soil ^{87}Sr/^{86}Sr (Montgomery et al., 2007) during burial, with modern proxy values, including, significantly, mineral water (Montgomery et al., 2006), which by definition must be free of modern pollutants and the clustering tendencies of the values of archaeological children who have a small migration window (e.g. Montgomery et al., 2005). However, whether liming, modern fertilisers and other anthropological activities are disrupting this validation in certain locations is still debatable. For example, Frei and Frei (2011) calculated that the application of modern fertilisers contributes minimally, if at all, to the ^{87}Sr/^{86}Sr of surface waters in Denmark. They concluded that any animals eating plants from, or drinking from water sources that percolate through, agricultural land applied with fertilisers, would have the same negligible effect on their skeletal strontium values. Conversely, Thomsen and Andreasen (2019) and Thomsen et al. (2021) have more recently suggested that the biosphere map of Denmark was based on samples already contaminated with agricultural lime. Sr-isotope analyses of tree cores from agricultural land in Ireland and a forest in Germany have shown a change of ±0.0003–0.0016 over the last century, these shifts being compatible with anthropological influences such as the commencement of the application of modern commercial fertilisers (Crowley, 2015), forest liming and/or mining activities (Maurer et al., 2012).

Such studies documenting recent changes in $^{87}Sr/^{86}Sr$ are important if they are of sufficient magnitude to introduce a disconnect in the transfer of $^{87}Sr/^{86}Sr$ from the bedrock through the food chain to animals and humans. Usually when interpreting and discussing $^{87}Sr/^{86}Sr$ in migration and mobility studies in Britain, any change to the 4th significant figure of the ratio can be considered within the population uncertainty: local populations can vary by 0.0002–0.002 (Evans et al., 2009, p.627–628), even siblings can vary by 0.0002 (Montgomery, 2002, p.146) and cattle born and raised in the same estate herd by 0.0006 (Towers, 2013, p.123–124). Therefore, the small Sr isotope ratio shift (+0.0003) seen in Crowley (2015) is unlikely to be a major concern for archaeological studies in Britain using modern proxy data to determine local $^{87}Sr/^{86}Sr$ for their animal and human populations. However, the larger shifts >0.001 in Sr-isotope ratios seen in Maurer et al. (2012) may be of greater concern.

Modern commercially-produced fertilisers were not available to pre-industrial populations. However, alternative types of natural fertilisers were available: manure use has been recorded from 6000 BCE from Neolithic sites across Europe (Bogaard et al., 2013) and the application of seaweed to improve and fertilise soils in island locations also appears to have a long history (Kvamme et al., 2004; Montgomery et al., 2007). The application of natural lime is documented in the Roman period (Goulding et al., 1989; Goulding, 2016), and, as seen in the previous examples, has the potential to change the biosphere $^{87}Sr/^{86}Sr$ of an area (Maurer et al., 2012; Thomsen and Andreasen, 2019). Consequently, growing crops and grazing animals on recently deforested farmland and historical liming may need to be factored into interpretations when determining 'local' Sr-isotope ratios for past populations. This may be of particular importance in regions where extensive forests or woodlands were known to be present or in particular time periods when land is likely to have been recently deforested and thus the principle of uniformitarianism may not hold for modern mapping proxy data.

In Britain, an ancient forest or woodland is defined as having been present since 1600 CE, but many are far older and unlikely to have been used for agriculture in the last few centuries. Approximately 2% of Britain is still ancient woodland (Hirst et al., 2015. These ancient wooded areas have had centuries to develop their own soil profile, which for silicate terrains may include depletion of carbonate mineral phases and their associated low $^{87}Sr/^{86}Sr$ values. This study looked at the $^{87}Sr/^{86}Sr$ difference measured in soils and plants, from two abutting environments (Ancient Forest and open farmland), both of which are founded solely on the Triassic Sherwood Sandstone Group, in the Sherwood Forest National Nature Reserve (SFNNR), Nottinghamshire that covers approximately 4.23 km$^{2.}$ This area has been forested since the early Holocene c. 11,550 BP following the end of the last glacial period in Britain (Stone et al., 2010) and was once part of the medieval 40 km^2 Royal Forest of Sherwood. It is currently home to over a thousand ancient oak trees, with many known to be >500 years old, the most famous being the Major Oak aged between 800 and 1000 years old (Natural England, 2014; The Sherwood Forest Trust, n.d.).

1.2. Geological and Sr-isotope summary of Sherwood Forest (Nottinghamshire)

The Triassic sediments that form the majority of the bedrock in the county of Nottinghamshire were deposited in a series of fault-bounded basins during the subsiding and breaking of the supercontinent Pangaea (Carney, 2010). Sherwood Forest and the transect extending across the surrounding agricultural land are located on the Sherwood Sandstone Group that consists primarily of red, yellow and brown sandstones interbedded with conglomerates indicative of fluvial origins. These were deposited in the East Midland Shelf that is linked to the Hinckley Basin (BGS, 1966, Sheet 113; Warrington et al., 1980; BGS, 1998, Sheet 101; Howard et al., 2008). Quaternary superficial (unconsolidated) deposits occur sporadically across the Sherwood Sandstone, which in the study region, mainly consist of alluvium, glacial till, and river terrace deposits (BGS, 1966, Sheet 113; BGS, 1998, Sheet 101).

The Triassic bedrock in Britain currently yields an mean biosphere $^{87}Sr/^{86}Sr$ of 0.7107 ± 0.0014 (2SD, n = 33) (Trickett, 2007; Leach et al., 2009; Chenery et al., 2010, 2011; Evans et al., 2010; Johnson, 2018; Montgomery, unpublished data: these data are compiled in Evans et al., 2018). Aquifer samples and mineral waters from Triassic sources give a similar result of 0.7095 ± 0.0004 (2SD, n = 22; Spiro et al., 2001; Montgomery et al., 2006). As mineral waters are defined as uncontaminated by pollutants, this suggests that the combined result of 0.7104 ± 0.0026 (2SD, n = 55: Evans et al., 2018) is a realistic estimate of British Triassic biosphere range.

2. Materials and methods

In the summer of 2016, a first set of eight plant samples were collected from a transect that stretched from around Farnsfield, north of Sherwood Forest National Nature Reserve, and towards Blyth in Nottinghamshire (SFL1 to SFL8). In the autumn of 2019 the rest of the plant and soil samples were collected from nine locations focusing on a smaller transect through the Sherwood Forest National Nature Reserve and the adjoining farmland (plants SWF-01 to SWF-18, soils Sher-01 to Sher-06). The foliage of long-established tree species, predominantly oak (*Quercus robur*), and more short-lived, shallow rooted, plants predominantly bracken (*Pteridium aquilinum*), were collected to investigate if trends were consistent between differing plant types. All the sites were specifically chosen to avoid areas with superficial cover. On collection the samples were sealed in Kraft paper bags and allowed to dry naturally. The soil samples were comprised of 200 g of material collected from the top 10 cm section of soil. Approximately 20–200 g of rock were collected from exposed rock outcrops outside Sherwood Forest National Nature Reserve (WAL-01, WAL-04 to WAL-07) and 1 cm length core samples of rock, from the Sherwood Sandstone Group, were sampled from each of two borehole cores (Weedon Camp and Kirkham boreholes), housed at the National Geological Repository at the British Geological Survey (BGS, Keyworth, Nottingham.) (Table 1). The outcrop and borehole rock samples were crushed, and a sand fraction prepared using standard rock preparation techniques in the Mineral Separation Laboratory at the British Geology Survey. The chemical preparation and Sr-isotope analysis of all these samples were conducted within the clean lab facilities at the National Environmental Isotope Facility (NEIF) at the British Geological Survey.

Chemical preparation for the plant samples was based on the microwave-assisted method of vegetation digestion developed by Warham (2011, p.42), from the method originally described in Evans et al. (2010). Approximately 200 mg of each plant sample was transferred into Teflon® microwave dissolution tubes and 2 mL of 8 M HNO_3 added to each. These were transferred to a hotplate (60 °C) to react overnight and then 5 mL of 8 M HNO_3 and 100 μL of H_2O_2 were added respectively. The dissolution vessels containing the samples were transferred into the microwave system (CEM MARS Xpress Xtraction) and microwaved at 175 °C for 20 min using a slow ramp up. Once the microwave system had completed and cooled, 500 μL of H_2O_2 was added to each, and samples were returned to the hotplate (60 °C) overnight. New, clean Savillex© beakers were labelled and the samples decanted into them. These were placed on a hotplate (100 °C) and dried down. If organics were still present, a further 2 mL of 8 M HNO_3 and 100 μL of H_2O_2 was added and the sample dried down again.

To approximate the extraction of the labile Sr from the soil, and sand fractions of the rock samples, approximately 1 g of each was placed in a centrifuge tube and 10 mL of 10% Romil© UpA grade acetic acid was added. Each sample was agitated gently throughout the day and then left overnight. The fluid was poured off and centrifuged to remove particulate matter. This supernatant fluid was then dried down and analysed.

All of the samples were then converted to chloride form using Teflon distilled 6 M HCl and centrifuged before the Sr was separated from them using Eichrom AG 50 W-X8 resin columns (Dickin, 1995, p.452). The Sr was then loaded onto single Re Filaments with TaF following the method of Birck (1986), and the isotope composition was determined by Thermal Ionisation Mass Spectroscopy (TIMS) using a Thermo Triton multi-collector mass spectrometer at the British Geological Survey. Mass-

Table 1
^{87}Sr/^{86}Sr isotope data for samples in this study.

Sample	Type	Species	Setting	^{87}Sr/^{86}Sr	Latitude	Longitude
SF L1	Plant	Oak	Ancient Woodland	0.71358	53.20433	−1.07786
SF L2	Plant	Oak	Ancient Woodland	0.71788	53.20289	−1.08625
SF L3	Plant	Oak	Ancient Woodland	0.71757	53.20792	−1.08578
SF L4	Plant	Hawthorne	Farmland	0.71218	53.17781	−1.09467
SF L5	Plant	Ash & elder	Farmland	0.70932	53.09522	−1.05783
SF L6	Plant	Oak & elm	Farmland	0.70959	53.36633	−1.06583
SF L7	Plant	Oak & beech	Farmland	0.70987	53.31131	−1.05236
SF L8	Plant	Lime	Farmland	0.71028	53.24506	−1.10267
SWF-01	Plant	Oak	Ancient Woodland	0.71380	53.20330	−1.06650
SWF-02	Plant	Bracken	Ancient Woodland	0.71422	53.20330	−1.06650
SWF-03	Plant	Oak	Ancient Woodland	0.71256	53.20483	−1.07411
SWF-04	Plant	Bracken	Ancient Woodland	0.71279	53.20483	−1.07411
SWF-05	Plant	Oak	Ancient Woodland	0.71254	53.20420	−1.07809
SWF-06	Plant	Bracken	Ancient Woodland	0.71251	53.20420	−1.07809
SWF-07a	Plant	Oak	Ancient Woodland	0.71254	53.20331	−1.08487
SWF-08	Plant	Bracken	Ancient Woodland	0.71277	53.20331	−1.08487
SWF-09	Plant	Oak	Ancient Woodland	0.71130	53.20268	−1.08947
SWF-10	Plant	Bracken	Ancient Woodland	0.71459	53.20268	−1.08947
SWF-11	Plant	Oak	Boundary hedge	0.71270	53.19733	−1.09274
SWF-12	Plant	Bracken	Boundary hedge	0.71127	53.19733	−1.09274
SWF-13	Plant	Oak leaves	Farmland	0.70968	53.19825	−1.08819
SWF-14	Plant	Wheat stubble	Farmland	0.71112	53.19825	−1.08819
SWF-15	Plant	Oak	Farmland	0.70941	53.19738	−1.08770
SWF-16	Plant	Wild flowers	Farmland	0.70986	53.19738	−1.08770
SWF-17	Plant	Oak	Farmland	0.71004	53.19475	−1.08577
SWF-18	Plant	Wild flowers	Farmland	0.70936	53.19475	−1.08577
Sher-01	Soil		Farmland	0.70918	53.19644	−1.08755
Sher-02	Soil		Farmland	0.70908	53.19738	−1.08770
Sher-03	Soil		Farmland	0.70929	53.19747	−1.08663
Sher-04	Soil		Ancient Woodland	0.71352	53.20354	−1.08255
Sher-05	Soil		Ancient Woodland	0.71315	53.20445	−1.07752
Sher-06	Soil		Ancient Woodland	0.71398	53.20479	−1.07491
WAL-01	Rock		Sherwood Sandstone	0.71120	53.22880	−1.00450
WAL-04	Rock		Sherwood Sandstone	0.71121	53.22957	−1.00469
WAL-05	Rock		Sherwood Sandstone	0.71009	53.22957	−1.00469
WAL-06	Rock		Sherwood Sandstone	0.71351	53.22997	−1.00489
WAL-07	Rock		Sherwood Sandstone	0.71316	53.22997	−1.00489
Weedon camp	Rock		Sherwood Sandstone	0.70959	53.78390	−2.87178
Kirkham	Rock		Sherwood Sandstone	0.71037	53.82009	−2.93582

dependant fraction of ^{87}Sr/^{86}Sr was corrected during each run using ^{86}Sr/^{88}Sr = 0.1194. Samples were run for 180 scans to an internal precision of $\leq \pm 0.00001$ (2SE). The international standard NBS 987 analysed over the period of this study gave ^{87}Sr/^{86}Sr of 0.71025 ± 0.00001 (n = 146, 2SD). Procedural blanks were in the region of 150 pg for microwaved samples and 80 pg for standard dissolutions.

3. Results

The data from this study are displayed on a map (Fig. 1) and tabulated (Table 1). The ^{87}Sr/^{86}Sr values from the biosphere, hydrosphere and the labile components of the pedosphere and lithosphere from this study are presented in Fig. 2, along with previously published data for water and biosphere samples from areas underlain by Triassic rocks (Table 2).

Borehole samples of Sherwood Sandstone lithologies provide a sample of unweathered, uncontaminated rock that give a data range for the labile components in these rocks. These borehole rock leaches give ^{87}Sr/^{86}Sr of 0.70959 and 0.71037 (from Weedon Camp and Kirkham boreholes respectively). Weathered Sherwood Sandstone rock outcrops produce the widest range of values in this study with a mean ^{87}Sr/^{86}Sr of 0.7118 ± 0.0029 (2SD, n = 5).

The soil leaches from farmland (this study) provide the lowest range of values with a mean ^{87}Sr/^{86}Sr of 0.7092 ± 0.0002 (2SD, n = 3) and the plant samples from farmland have a mean ^{87}Sr/^{86}Sr of 0.7104 ± 0.0022 (2SD, n = 13.) Two samples (SWF 11 &12) oak and bracken were collected from the boundary hedge separating the farmland and forest and are designated as transition zone samples. If these two samples are excluded the remaining eleven farmland samples give an ^{87}Sr/^{86}Sr value of 0.7101 ± 0.0017 (2SD, n = 11). These data are similar to previously published data giving a mean of 0.7104 ± 0.0026 (2SD, n = 55) (Evans et al., 2018).

Data from the forested areas contrast with that of the unforested areas giving significantly higher values: soil leach ^{87}Sr/^{86}Sr values of 0.7135 ± 0.0008 (2SD, n = 3) and plant ^{87}Sr/^{86}Sr values of 0.7137 ± 0.0035 (2SD, n = 16). There is a gradational drop in ^{87}Sr/^{86}Sr values from the forest into the adjacent farmland, and this is seen in both shallow rooted plants and the deeper-rooted trees (Fig. 3), with values below 0.710 from both plant types at one site recorded 140 m from the boundary hedge. The shift in mean ^{87}Sr/^{86}Sr values between the forest and farmland plants is approximately +0.0037 that represents a significant effect on the over lying biosphere ^{87}Sr/^{86}Sr due to land use across a single lithology.

4. Discussion

4.1. The biosphere ^{87}Sr/^{86}Sr variation of the Sherwood Sandstone Group

The Sherwood sandstone has previously been described as showing “variable weathering of outcrops near surface, and deeper dissolution of the cement binding the sand-sized grains.” (BGS 2). In the case of the Sherwood Forest environment, the cement dissolution is attributed to the long-term action of humic acid derived from the leaf litter and other organic detritus (Dijkstra et al., 2003; Poszwa et al., 2004). This means that it is possible for a well-established forest to change its own geochemical biosphere signature through time. It could be argued that it is the farmland that has changed through the anthropogenic addition of lime, however, the agreement of ^{87}Sr/^{86}Sr values from the pristine borehole data, the water samples, soils and plant samples argues for the farmland having the “pristine”

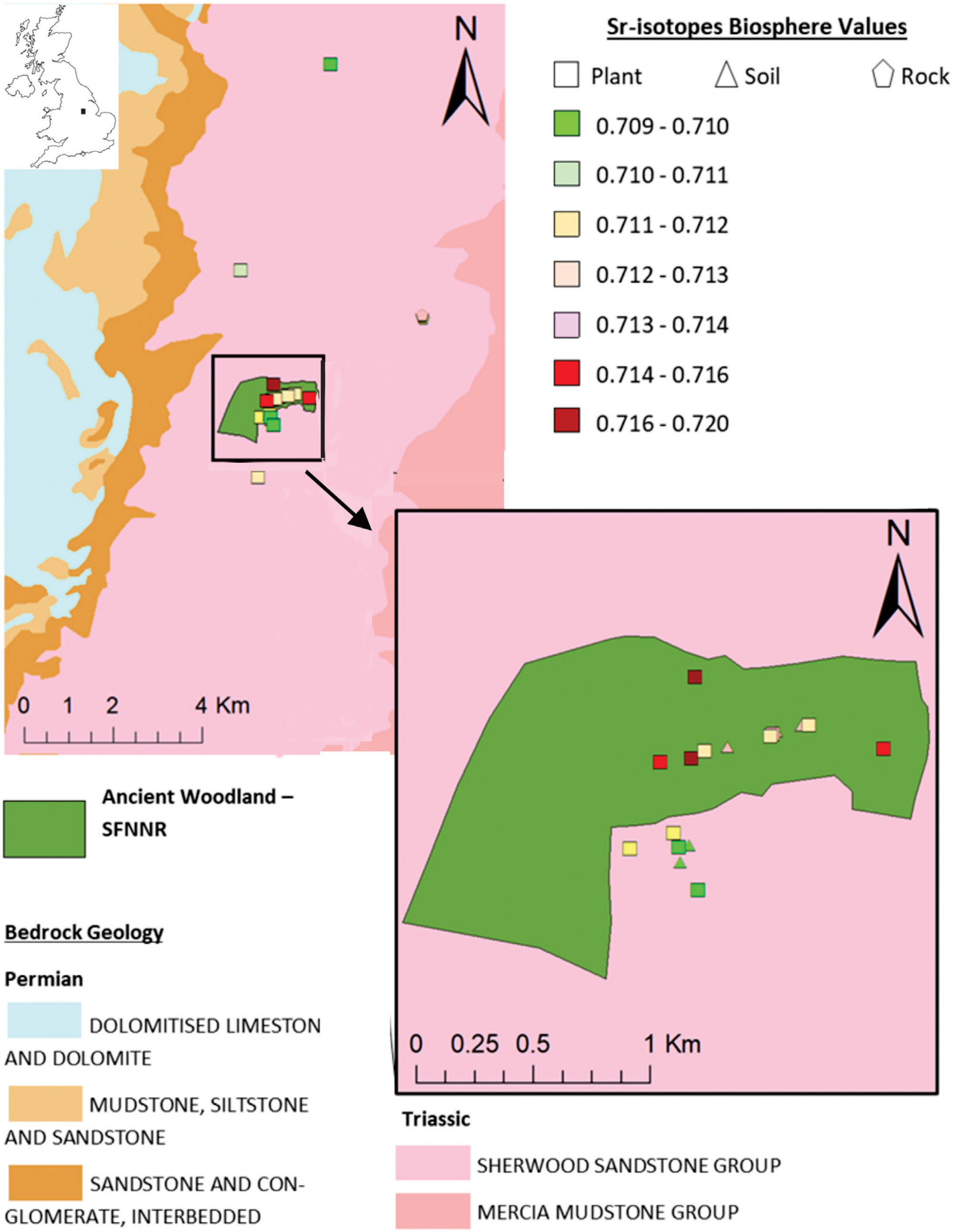

Fig. 1. The plant, soil and rock $^{87}Sr/^{86}Sr$ from across the Sherwood Forest study area along with GIS data of the 1:625000 Bedrock map (BGS, DiGMapGB, 2007) and Ancient Forest Inventory, England (AWI, 2013).

$^{87}Sr/^{86}Sr$ values around 0.7101 whereas the forest-induced deep weathering and leaching of carbonate cement from the sandstone (BGS 2) leads to the elevated, silicate-only derived biosphere values in the forest plants.

During the Medieval period, this forest covered an area approximately 10 times greater than today (~40 vs. ~4 km^2: Natural England, 2014; The Sherwood Forest Trust). As such, Sherwood Forest alone covered approximately 20% of the Sherwood Sandstone Group on the East Midlands Shelf. It is unlikely that all of this area was covered by dense forest; a significant part was open land, heathland and grassland. However, larger areas of woodland existed in the Medieval period than at present and can be assumed to have a biosphere $^{87}Sr/^{86}Sr$ of 0.7137 ± 0.0034 (2SD, n = 16, plants and soil) due to the forest effect as shown in this study. Any other unforested land, from open to agricultural, on the Sherwood Sandstone Group should have a more typical Triassic biosphere of $^{87}Sr/^{86}Sr = 0.7103 \pm 0.0024$ (2SD, n = 71; Evans et al., 2018; this study). Therefore, humans and animals living in the region of Sherwood Forest during the Medieval period and procuring their food locally, could have $^{87}Sr/^{86}Sr$ values that lie in between the forest and the unforested values depending on their food procurement and animal husbandry strategies and where they sourced the majority of their diet.

4.2. Implications for the past

The results of this study show that mature forests can alter the $^{87}Sr/^{86}Sr$ biosphere range that would be predicted from the lithology alone or obtained from an unforested landscape. However, the extent to which this happens will depend on the underlying lithology. Rocks such as the Sherwood Sandstone Group, which have a carbonate component, are the type of rocks most susceptible to this selective leaching process. Limestone

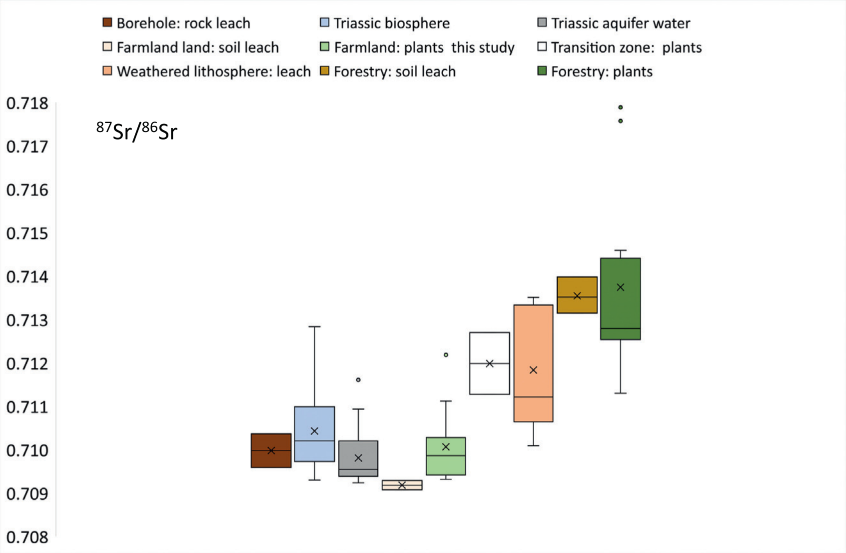

Fig. 2. A box and whisker plot of the plant, soil and rock $^{87}Sr/^{86}Sr$ from across the study area separated into Farmland, Ancient Woodland (Sherwood Forest) and Sherwood Sandstone Group rock samples for comparison. The box represents the interquartile range of the data with the median value marked as a bar within. The whiskers extend to 1.5 * the interquartile range and outliers beyond this are marked as points.

or chalk bedrock, consisting largely of carbonate minerals, will show very little to no difference in biosphere $^{87}Sr/^{86}Sr$ between forest and other types of land uses because of their homogenous nature and high carbonate content. Igneous and metamorphic lithologies are unlikely to be significantly affected by leaching processes as they compromise predominantly silicate minerals, which are more resistant to selective leaching (Faure, 1986). However, Sr-rich phosphate minerals such as apatite and monazite and the progressive removal of Sr bearing silicates such as feldspars (with lower $^{87}Sr/^{86}Sr$ values), through time, is possible in igneous/metamorphic lithologies and is likely one of the mechanisms observed by Åberg et al. (1990) although not explicitly stated.

How does this forest effect affect the interpretation of biosphere data from the past? Areas of southern Britain that can support higher $^{87}Sr/^{86}Sr$ biosphere values are currently restricted to outcrops of igneous rock such as the Malvern Hills Igneous Complex and parts of the SW England granite. The possibility that forested areas could support a biosphere with values >0.713 opens the possibility that much larger areas of low-lying fertile land carry these values. The Malvern Hills complex occupies about 1 km^2 of land whereas Sherwood Forest may have occupied *c.* 40 km^2; however, the extent of ancient forests could be far more extensive as shown by the New Forest that currently occupies 566 km^2. These forests could supply substantial areas of accessible foraging and grazing for wild animals and domesticates such as cattle and pigs.

The forestry effect is going to be most critical to provenance studies that focus on periods before extensive deforestation, such as the Neolithic, and periods of recent forest clearance for agriculture. An example of such a human population comes from the early Neolithic long cairn at Penywyrlod, south Wales (Neil et al., 2017). The individuals at this site have human tooth enamel values ranging from 0.71323 to 0.71702 (n = 11), whereas the biosphere range for the underlying Devonian rocks in southern Britain is 0.71054 + 0.002 (2SD, n = 45). Neil et al. (2017) argued that while these people were clearly not sedentary, evidence from a sample USK-04 ($^{87}Sr/^{86}Sr$ = 0.71550) pointed at the potential for a wider range of values within foraging distance. It is notable that this sample was collected from a now-restricted area of well-established woodland and might hint at a more geographically extensive elevated $^{87}Sr/^{86}Sr$ biosphere if the area was wooded in the Neolithic period.

Table 2
Summary of statistical analysis of different domains referred to in this study.

Domain	Average	2SD	n
Pristine borehole: rock leach	0.7100	0.0011	2
published Triassic data-dentine soil and plants	0.7098	0.0017	48
Triassic aquifer water	0.7094	0.0002	11
Farmland land: soil leach	0.7092	0.0002	3
Farmland: plants biosphere this study	0.7101	0.0017	11
Transition zone: plants	0.7120	0.0020	2
Weathered lithosphere: leach	0.7118	0.0029	5
Forestry: soil leach	0.7135	0.0008	3
Forestry: plants	0.7139	0.0040	11
Overall mean of unforested area	0.7103	0.0024	71

Another area where this forest effect may have significance is the interpretation of herbivorous animal teeth. Since the Neolithic, cattle and sheep were foddered with forest vegetation particularly during the winter months, (Rowley-Conwy, 1982; Brown, 1997; Robinson, 2000; Bell and Walker, 2005, p.164–168) whereas their human counter parts are more likely to derive the bulk of their $^{87}Sr/^{86}Sr$ from grains, vegetables and pulses grown on open ground. Cattle from Roman Worcester (Gan et al., 2018) also displayed high Sr-isotopes ranging between 0.7139 and 0.7158 that were inconsistent with the current understanding of the biosphere $^{87}Sr/^{86}Sr$ of 0.7100 ± 0.0024 (2SD, n = 71). They thus proposed that some of the cattle had been raised elsewhere and, driven from areas of Britain such as Wales which can support biosphere values of >0.713 (Evans et al., 2018) to a regional cattle market at Roman Worcester in an attempt to identify the antiquity of a practice documented since Norman times. However, Shrawley Woods Forest, recorded in the Doomsday Book, is 15 km to the north of Worcester, and is situated on the same Triassic Sandstone as Sherwood Forest. This could, perhaps, have provided a local source for the values recorded in the teeth if the cattle were grazed in the forest.

Finally, the effect of forest in a modern environment might equally produce anomalous results when extrapolated back into the past. This may provide an explanation for the debate on the provenance of an iconic Bronze Age female - the Egtved girl - buried in an oak coffin in the Egtved area of Central Denmark. Frei et al. (2015) argue that the elevated $^{87}Sr/^{86}Sr$ measured from this adolescent female (up to 0.7155) exclude her from having an origin on mainland Denmark based on current mapping (Frei and Frei, 2011 and Frei et al., 2022). However, it has been suggested that the biosphere map of Denmark, which is based on surface waters, was contaminated with agricultural lime (Thomsen and Andreasen, 2019; Thomsen et al., 2021).

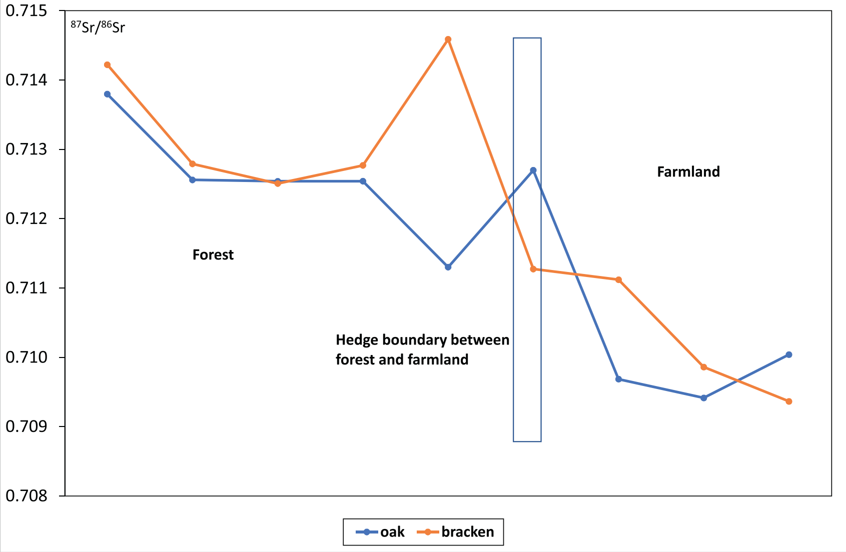

Fig. 3. Traverse of ^{87}Sr/^{86}Sr isotope compositions of oak and bracken samples from the forest into farmland.

These authors argue that surface waters at a "pristine" forest site, provide sufficiently high ^{87}Sr/^{86}Sr to account for the human values obtained.

The forest effect model presented in this paper provides an alternative explanation for the Danish data and supports the original interpretation. The "pristine" radiogenic values of Thomsen and Andreasen (2019) come from a stream sourced in a mature coniferous forest. Irrespective of the question as to whether such a source would enter the food chain and contribute in any meaningful way to the plants consumed by animals and humans, the principal source of skeletal strontium (Montgomery, 2010). Using the forest effect, we would argue that the water coming from this area is atypical and caused by soil leaching due to humic acid build up under the forest canopy. The effects persist for only a few metres away from the forest, as with Sherwood Forest. This modern forest did not exist during the Bronze Age and, consequently, it could make no contribution to the Bronze Age biosphere.

The forest effect should not, and does not, provide an explanation for all animal and humans within England and Wales with values >0.714. Several ($n = 13$) Bronze Age and Iron Age humans with ^{87}Sr/^{86}Sr between 0.713 and 0.717 have been excavated from sites overlying the Carboniferous Dolomitised Limestones of central England. They include seven Bronze Age humans from the White Peak region of the Peak District (Parker Pearson et al., 2016; Montgomery et al., 2019) and six Iron Age humans from Ferry Fryston, West Yorkshire (Jay et al., 2007). The tooth enamel of two cows from the Iron Age chariot burial at Ferry Fryston have also produced high ^{87}Sr/^{86}Sr of approximately 0.715 (cattle 5) and 0.720 (cattle 3) (Jay et al., 2007). The Carboniferous Dolomitised Limestones host a mean ^{87}Sr/^{86}Sr biosphere of 0.7092 ± 0.0004 ($n = 11$, 2SD: Evans et al., 2010), while the nearby Carboniferous Millstone Grits host a mean ^{87}Sr/^{86}Sr biosphere of 0.7117 ± 0.0014 ($n = 11$, 2SD: Evans et al., 2018). As previously discussed, soils overlying Carboniferous Dolomitised Limestone, a marine carbonate, will not be affected by the forest effect. The effect of forestation will vary for different lithologies, being dependent on the composition of the silicates and the extent to which soluble Sr-bearing minerals can be removed. It is thus difficult to predict if forests on the Millstone Grits would display a difference as significant as seen in Sherwood Forest in this study. However, even with a theoretical increase of +0.0034 for forested domains on the Millstone Grits, a common lithology in northern England, this still would not account for the majority of the humans and cattle with ^{87}Sr/^{86}Sr > 0.716 from the Peak District and Ferry Fryston. Therefore, the conclusion that these prehistoric humans are migrants whose origins are inconsistent with their place of burial remains unchallenged.

5. Conclusion

The transect of forest to farmland ^{87}Sr/^{86}Sr displayed in this study highlights the importance of collecting and understanding 'local' biosphere ^{87}Sr/^{86}Sr for mobility and migration studies. Just like heavy rainfall, sea-spray and wind-blown terrestrial aerosols (Capo et al., 1998; Chadwick et al., 1999; Yaalon, 1997; Ganor and Foner, 2001; Montgomery et al., 2003; Bentley, 2006; Evans et al., 2009; Montgomery, 2010), ancient forests can produce biosphere ^{87}Sr/^{86}Sr values that are different from unforested land on the same lithology. As such they should be considered as a separate domain when producing a biosphere, or baseline, map of ^{87}Sr/^{86}Sr. A difference of +0.0037 has been observed between the average plant ^{87}Sr/^{86}Sr of the ancient forest of the Sherwood Forest National Nature Reserve and surrounding farmland, all of which are founded upon the Sherwood Sandstone Group bedrock. This shift is significant and is believed to be caused by organic acid production, increased soil acidity, and throughfall associated with forest environments: collectively termed the forest effect. The borehole rock leach samples from the Sherwood Sandstone Group, as well as aquifer and mineral waters from previous studies (Spiro et al., 2001; Montgomery et al., 2006; Evans et al., 2010) have been used to deduce that the Sherwood Sandstone Group naturally releases Sr with ^{87}Sr/^{86}Sr values between 0.709 and 0.710 and the alternative reason for the observed shift through the anthropological addition of liming to farmland can be rejected in this instance. The forest effect, and its associated change in biosphere ^{87}Sr/^{86}Sr, has the potential to impact the interpretation of animal and human ^{87}Sr/^{86}Sr, as such future mobility and migration studies should consider this when using modern proxy data in archaeological studies of residential mobility particularly in periods and places where agricultural land may have been recently deforested or forests and their clearings may constitute a significant location for food resources. However, it also opens up an opportunity to further understand food procurement strategies and animal husbandry practices such as pannage, in the past, as well as the interaction of humans with their natural environment.

It is expected that changes of different magnitude and direction will be observed between the average biosphere ^{87}Sr/^{86}Sr of ancient forests vs other land uses hosted on different bedrock lithologies. The greatest change will likely be seen on sedimentary lithologies where the soluble Sr-bearing minerals such as carbonates can easily be leached out over time, such as the Sherwood Sandstone Group investigated in this study. The existence, spatial extent, age of forest, and underlying rocks type are likely all factors that need to be considered when assessing the impact this effect may have on mobility studies along with the extent to which humans and animals might utilise the forest resource.

CRediT authorship contribution statement

L. Johnston: Conceptualization, Sampling, Analytical and Interpretive Analysis, Creation and writing the initial draft. J. A. Evans: Supervision, Sample analysis, Editing. J. Montgomery: Supervision and Editing, C. Chenery, Sampling and Data analysis.

Declaration of competing interest

The authors have no competing interests.

Acknowledgments

Dr. Lucie Johnson was supported by a NERC doctoral studentship (Award Reference No. 1345819). We thank the Sherwood Forest National Nature Reserve, Doris Wagner for analytical support, and Sal Kellett for producing the graphical abstract.

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