

CONTRIBUTION OF STRONTIUM TO THE HUMAN DIET FROM QUERNS AND MILLSTONES: AN EXPERIMENT IN DIGESTIVE STRONTIUM ISOTOPE UPTAKE*

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The question of whether rock grit ingested unintentionally from querns, metates or millstones, or deliberately through pica or geophagy, is bioaccessible in the human gut has not been addressed in archaeological strontium (Sr) isotope studies. This study employed the unified bioaccessibility method and determined that ingested rock grit can provide bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$, but that unintentional consumption is unlikely to constitute > 1% of the diet (by mass) and will not significantly change, that is, by > 0.001, human skeletal $^{87}\text{Sr}/^{86}\text{Sr}$. The use of locally or non-locally sourced querns or millstones will not affect the interpretation of archaeological human $^{87}\text{Sr}/^{86}\text{Sr}$ values in Britain.

KEYWORDS: STRONTIUM, $^{87}\text{Sr}/^{86}\text{Sr}$, BIOACCESSIBLE, HUMAN DIET, UNIFIED BIOACCESSIBILITY METHOD (UBM), GRINDING STONES, QUERNS AND MILLSTONES

INTRODUCTION

Strontium (Sr) isotope analysis is used to track the movement and provenance of modern and ancient humans and animals. The fundamental principle upon which this rests is 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 $^{87}\text{Sr}/^{86}\text{Sr}$ (e.g., Aberg 1995; Capo *et al.* 1998; Beard and Johnson 2000; Bentley 2006; Evans *et al.* 2010, 2018). This variation is then traceable through the diet and into body tissues.

Animals and humans are not passive receptors of environmental Sr, but make choices about when and where they source the components of their diet. Once ingested, different dietary components may contribute more or less to the Sr in the body tissue that is measured. Geological and environmental processes, such as differential weathering, mixing of Sr from different sources and atmospheric deposition, also affect the available Sr. This multiplicity of options and events are distilled into the single measured $^{87}\text{Sr}/^{86}\text{Sr}$ that is obtained from tissues such as tooth enamel.

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Attempting to untangle and reconstruct the environmental constraints and culturally determined actions of people in the past that may lie behind an Sr isotope ratio is complex, but is the ultimate goal of archaeologists.

There are aspects of human activity that are not currently taken into account in our interpretations of Sr isotope ratios. For example, it is widely recognized that tooth wear in archaeological populations was often far greater than today—one of the main reasons for this is believed to be the coarser diet and, particularly, the presence of soil and rock in the food, included either accidentally via dirt or grit from grinding, or intentionally as a result of geophagy or pica (Eshed *et al.* 2006; Deter 2009). It has also been suggested by Aberg *et al.* (1998) that the grit from the milling process may have been a major contributor to human $^{87}\text{Sr}/^{86}\text{Sr}$ values, particularly during the medieval period.

This study addresses the possible contribution that rocks, used to grind grain, might have made to the $^{87}\text{Sr}/^{86}\text{Sr}$ of the human diet in the past from both locally derived and imported grinding materials. A wide range of rock types such as basalts, granites, schists and sandstones have been used as querns and, later, metates and millstones to grind food. Most ancient peoples would have used the hardest or coarsest local rock available to them for their grinding stones, but such stones were also traded and transported. For example, the Mayen Lava querns and millstones from quarries in Germany were being imported into Britain by the Roman period (Peacock 2013, 151–153) and thus have the potential to introduce non-local Sr into the diets of local people *if* they provide bioaccessible Sr and *if* that Sr is metabolized in the human gut.

The current model for the origin of Sr metabolized from the omnivore diet is that it is dominated by dietary plants rather than by water or animal products. This is due to complex antagonisms and synergisms with other dietary components such as calcium and protein that suppress, or conversely in the case of fibre and phytate, enhance elemental Sr absorption (see Montgomery 2010 for a summary). Whether ingested non-food components such as soil and rock contribute has not been explored and is rarely, if ever, taken into account in interpretations. Would ground rock from a quern or millstone, or individual minerals such as radiogenic micas (as found in the intestine of the Alpine Ice Man; Müller *et al.* 2003), provide a substantial Sr component when ingested regularly and made bioaccessible by the strong acids of the human digestive system? If so, the direct consumption of rock grit could bypass the biopurification of plants and animals and has the potential to disrupt the biosphere–human link, that is, the fundamental underlying assumption all applications using Sr isotopes to identify migrants is based on.

The nature and type of rocks used for grinding grain

Rocks have been used as tools to grind grains, nuts and seeds across the globe, but have been used in Britain since at least the start of the Neolithic in the fourth millennium BCE (Peacock 2013, 17–21; Watts 2014, 19). These grinding tools can be categorized into two main forms: querns and millstones. Their production rates vary depending on the type of quern or millstone being used, its lithology, its size and the type of grain or food being ground. In an assessment of flour production experiments, Peacock (2013, 126–128) concluded that approximately 0.5–4.0 kg of flour per hour can be produced from a quern. A set of millstones, on the other hand, can produce approximately 10–50 kg of flour per hour (128–129).

The lithology of the quern or millstone is important for this study, as it is the rock tool itself that contaminates the food that humans digest. Grinding stones of many lithologies have been archaeologically documented in Britain, including granite, gneiss, mica schist, gabbro, basalt, sandstone, conglomerate, greensand, silicified limestone, quartzite and andesite (Peacock 2013,

62–65; Watts 2014, 29–30). Some of these rocks, such as gabbro and basalt, will give low whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ typically between 0.704 and 0.708, whereas granites and schists can give very high whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ values, sometimes > 1.0 (Faure and Powell 1972; Peterman and Hildreth 1978; Rundle 1979; Faure 1986). Granites and gabbros (including their finer crystal grained equivalents such as rhyolites and basalts) represent the extremes of isotope composition, but they were not commonly used in Britain, with the possible exception of the imported Mayen Lava from Germany. Instead, the native abrasive sandstones and gritstones were particularly prized and, hence, more commonly used across all periods in Britain (Peacock 2013, 2–3, 62–65,



Figure 1 Locations of the rock samples: Millstone Grit, Stoke Hall Quarry, Peak District (Derbyshire); Pennant Sandstone, Great Berry Quarry, Forest of Dean (Gloucestershire); and Eskdale granite, Beckfoot Quarry, Lake District (Cumbria).

151–154). All the lithologies used in this study are of British origin: Millstone Grit from the Peak District; Pennant sandstone from the Forest of Dean, near the southern borders of Wales; and Eskdale granite from the Lake District, Cumbria (Fig. 1).

CURRENT BRITISH BIOSPHERE AND HUMAN $^{87}\text{Sr}/^{86}\text{Sr}$

The current understanding of the Sr isotope biosphere in Britain is based on the analysis of plants as the primary source of Sr isotopes in the diet. The distribution mapping, although a work in progress, suggests that the $^{87}\text{Sr}/^{86}\text{Sr}$ biosphere of Britain ranges from 0.707 to 0.720, but this highest value is only found in a few isolated localities (Evans *et al.* 2018). The current published range for archaeological humans excavated in Britain is narrower (0.7078–0.7165), as would be expected as a result of averaging: the median is 0.7096, with an interquartile range of 0.0014 (Evans *et al.* 2012). Since Evans *et al.* (2012) published their review, more individuals with higher values have been found (e.g., Parker Pearson *et al.* 2016; Neil *et al.* 2017; Montgomery *et al.* 2019), but there are still no human populations with values > 0.7165 that are deemed to be of local origin and can be explained by the local geology or biosphere.

MATERIALS AND METHODS

This study focuses on three lithologies: Millstone Grit, Pennant sandstone and Eskdale granite. The abrasive gritstones and sandstones are well established rocks for milling (Peacock 2013, 62–65; Watts 2014, 29–30) and granite is one of the most radiogenic rocks found within Britain to be used as a grinding stone and thus has the potential to provide a high $^{87}\text{Sr}/^{86}\text{Sr}$ bioaccessible value. The sample locations are given in Figure 1.

This study also employs the *in vivo* validated unified bioaccessibility method (UBM) developed by the Bioaccessibility Research Group of Europe (BARGE) for examining the bioaccessible inorganic components in soils (Hamilton *et al.* 2015). The UBM simulates the human digestive system *in vitro*, including digestive fluids, body temperature, timings at each stage of, and agitation produced by, the digestive system.

The geographical source and the mineralogy of the selected rock samples

Millstone Grit describes a succession of Carboniferous gritstones found mainly in the Peak District and Pennines of northern England. This lithology is dominated by the feldspathic minerals of microcline, plagioclase, orthoclase and also the mica mineral muscovite; calcite can be another common component and some samples of Millstone Grit can have up to 8% apatite (Muir 1963). All these minerals are Sr-bearing phases.

Millstone Grit is believed to have been extensively quarried since the Iron Age, but many known quarries are abandoned and difficult to access (Butcher 1970; Tucker 1985; Pearson 2000; Peacock 2013, 65). The Millstone Grit used in this study was collected from the still-active Stoke Hall Quarry near Grindleford, Derbyshire (opened in 1835).

The Pennant sandstone is a Carboniferous rock described as being lithic and rich in the minerals feldspar and mica (BGS Lexicon). The main Sr-bearing phases of Pennant sandstone are predominantly the feldspars such as plagioclase and K-feldspar and the micas which are mainly muscovite (Roe 1987, 1988).

The Pennant sandstone was collected from the Great Berry Quarry, Brierley, Gloucestershire, and is part of the Coal Measures found in the Forest of Dean, near the southern borders of Wales.

This sandstone has been used as building stone since the Roman period (Welch and Trotter 1961; Williams 1971; Price 2002) and was used for a variety of other stone tools including honestones, whetstones, querns and millstones (Moore 1978; Fowler 1981; Roe 1987, 1988; Allen 2014).

Granites are felsic, intrusive, igneous rocks and will often contain combinations of biotite and muscovite micas, K-feldspar and plagioclase as the main Sr-bearing phases (Hatch *et al.* 1974). The Eskdale granite was collected from Beckfoot Quarry in the Lake District. The Beckfoot Quarry is situated in the Eskdale pluton and a sample of the 'normal' granite was taken as it is rich in muscovite (Young 1999).

Granites, although not as popular and extensively used as the abrasive sandstones and gritstones, have been used as querns or millstones in Britain when locally available (Watts 2014, 77, 104, 132–133). They are very difficult to shape and grind due to their large crystal sizes (Coope 1979, 100), but Bronze Age and Roman querns of Eskdale granite have been recorded at Drigg, Cumbria (Cherry and Cherry 1968; Cherry 1988) and several post-medieval millstones have been found in Cumbrian mills (Davies-Shiel 1978). Nicholson and Burn, writing in the 18th century about the massive granite glacial erratics used to construct the monumental Neolithic Shap Avenue (Waterhouse 1985, 125), recorded that the 'country people [...] cut these stones (but with difficulty) for millstones' (Nicholson and Burn 1777, 477).

Sample preparation

All the rocks were crushed and ground using standard rock preparation techniques in the Mineral Separation Laboratory at the British Geological Survey (BGS, Keyworth). The < 250 µm fraction of each rock was selected for the UBM to mimic the rock that could potentially be ground and make its way into food products. A whole-rock powder was also prepared for each rock.

Large detrital muscovite micas were hand-picked from the > 500 µm fraction of the Millstone Grit. Because of their large size, these micas have the potential to be easily plucked from a quern or millstone. Micas were only present in the Millstone Grit and the amount collected was too small to run through the UBM. Instead, they were subjected to a simplified method that mimicked the gastric fluid stage of the UBM and thus the effect only of stomach acid on the minerals.

Unified bioaccessibility method (UBM)

Bioaccessibility extractions were undertaken according to the method outlined in Hamilton *et al.* (2015) within the Inorganic Geochemistry Laboratories at the BGS (Keyworth). Briefly, 0.6 ± 0.01 g of the ground rock sample (< 250 µm fraction) were accurately weighed into 85 ml Nalgene® oak ridge tubes (ThermoScientific, UK). Simulated saliva and gastric fluid were added to each tube, the pH was adjusted to 1.2 ± 0.05 , followed by 1 h of end-over-end agitation in a temperature-controlled water bath held at 37°C. The samples were then taken through the stomach and intestine extraction using simulated duodenal and bile fluids (pH adjusted to 6.3 ± 0.5 where necessary to account for natural buffering of the sample material). The stomach and intestine extraction involved 4 h of end-over-end agitation at 37°C. The samples were then extracted through centrifugation at 6500 RPM for 15 min. At the end of each extraction, 10 ml of the supernatant were collected and preserved with 0.2 ml concentrated (15.9 M) HNO₃ before analysis and diluted 100-fold with 1% v/v HNO₃. A total of 0.5% v/v HCl before analysis by inductively coupled plasma mass spectrometry (ICP-MS).

The determination of Sr concentration was carried out using an Agilent 7500cx ICP-MS fitted with a CETAC ASX-520 autosampler. Sample introduction from the autosampler to the ICP-MS

was controlled by a CETAC ASXpress+ vacuum pump. A quality control (QC) check standard (ULTRA Scientific, USA), containing Sr at $25 \mu\text{g l}^{-1}$, was analysed at the start and end of each run and after no more than every 20 samples. To overcome polyatomic interferences the ICP-MS collision cell was operated in He mode at a flow rate of 5.5 ml min^{-1} . Sample values were calibrated using the standard SpexCertiPREP at 1, 10 and $100 \mu\text{g L}^{-1}$. The procedural blank for the UBM procedure was 33 ng, which represents $< 1\%$ of total Sr yield.

Simplified leach method and standard whole-rock dissolution method

Approximately 0.08 g of detrital muscovite mica from the Millstone Grit were exposed to 10 ml of stomach acid strength HCl (2.5 M) in a Savillex[®] beaker in a temperature-controlled water bath at 37°C for 1 h. The sample was shaken approximately every 5 min to mimic the agitation process of the UBM method. After 1 h the supernatant, termed leach, was collected by centrifuging at 3000 RPM for 7 min and decanting into a newly labelled Savillex beaker. The leach was then dried down. The HCl-insoluble residues were dissolved using a standard HF-HNO₃ method and converted to chloride form in preparation for Sr separation.

Approximately 100 mg of each of the whole-rock powders were dissolved using a standard HF-HNO₃ in labelled Savillex beakers and converted to chloride form. All samples were centrifuged before Sr separation.

Sr was separated from all the samples using Dowex resin columns (Dickin 1995, 452). The Sr was then loaded onto single Re Filaments with TaF following the method of Birck (1986), and the isotope composition and concentrations were determined by thermal ionisation mass spectroscopy (TIMS) using a Thermo Triton multi-collector mass spectrometer. The international standard for $^{87}\text{Sr}/^{86}\text{Sr}$, NBS987, gave a value of 0.710251 ± 0.000005 ($n = 19$, 2 SD) during the analysis of these samples. Procedural blanks were in the region of 100 pg.

RESULTS

The whole-rock Sr isotope compositions are given in Table 1. The whole-rock sample of the Millstone Grit has an Sr concentration of 106 ppm and $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.72079, which is within the range, and typical of, previously determined whole-rock values for this lithology, that is, 0.71300–0.74080 (Diskin 2002, 95–96). The Pennant sandstone has a lower Sr concentration of 57 ppm and a higher whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.73028. The Eskdale granite is the most radiogenic sample with $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.91373 and has the lowest Sr concentration of 27 ppm,

Table 1 *Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ of the whole-rock samples and the bioaccessible Sr derived from the rock grit determined from the unified bioaccessibility method (UBM)*

<i>Rock sample</i>	<i>Whole-rock Sr (ppm)</i>	<i>Whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$</i>	<i>Bioaccessible Sr (ppm)</i>	<i>Bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$</i>	<i>Whole-rock bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$</i>
Millstone Grit	106	0.72079	3	0.71147	0.00932
Pennant Sandstone	57	0.73028	4	0.71281	0.01747
Eskdale Granite	27	0.91373	3	0.72224	0.19149

comparable with data from Rundle (1979) who reported an $^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.76407–1.11430 for this granite.

The bioaccessible Sr from the Millstone Grit had $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.71147 and an Sr concentration of 3 ppm; Pennant sandstone yielded a higher bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.71281 and a similar Sr concentration of 4 ppm; and the Eskdale granite was the most radiogenic at 0.72224 with an Sr concentration of 3 ppm (Table 1). In all three cases there is a significant difference between the whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ and the bioaccessible component (Fig. 2); the bioaccessible Sr has a lower $^{87}\text{Sr}/^{86}\text{Sr}$ value and Sr concentration than the whole rock. The concentration of bioaccessible Sr recovered (3–4 ppm) is comparable between rock types despite different initial concentrations.

The results from the detrital muscovite mica of the Millstone Grit can be seen in Figure 3. The bioaccessible Sr is lower ($^{87}\text{Sr}/^{86}\text{Sr}=0.71450$) than both the whole-rock Millstone Grit ($^{87}\text{Sr}/^{86}\text{Sr}=0.72079$) and the indigestible mica residue ($^{87}\text{Sr}/^{86}\text{Sr}=0.74858$). Mica could realistically be expected to provide the most radiogenic contribution to the diet because of its high Rb/Sr ratio. Pure micas will typically contain < 10 Sr ppm (Eberlei *et al.* 2015). However, large micas can often contain inclusion of other minerals including readily acid-soluble phosphates such as apatite. The high Sr concentration of 270 ppm in the bioaccessible component of these micas suggests this is the case and that these minerals are not contributing highly radiogenic Sr. The bio-accessible $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7145 from the micas thus represents the maximum bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ that the Millstone Grit can contribute.

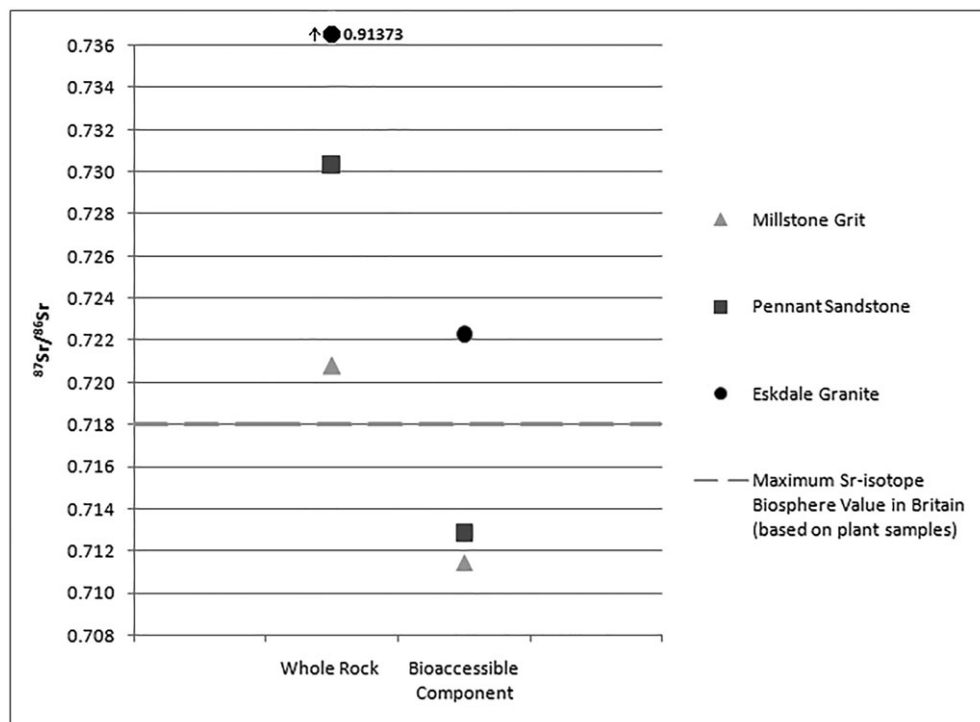


Figure 2 $^{87}\text{Sr}/^{86}\text{Sr}$ of the whole rock versus the bioaccessible component of the ground rock determined by the unified bioaccessibility method (UBM).

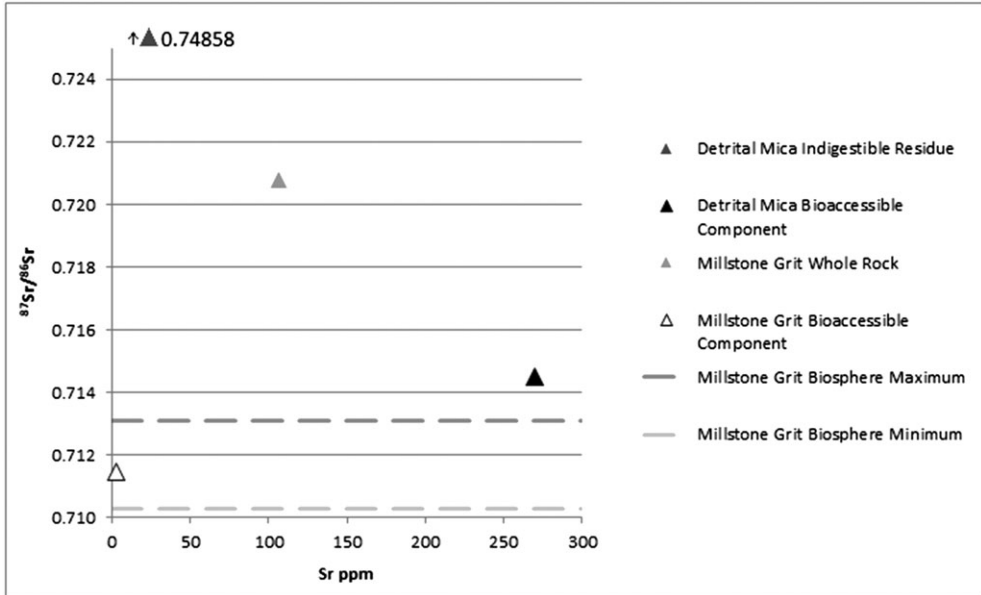


Figure 3 $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr concentration of the detrital muscovite micas from the Millstone Grit compared with the whole rock and bioaccessible component of Millstone Grit determined by the unified bioaccessibility method (UBM). Biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ on the Millstone Grit was determined using the methodology of Evans *et al.* (2018).

DISCUSSION

Comparison of bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from grinding stones with biosphere values contributing to the diet

The Millstone Grit crops out in the Pennines where it is interspersed with Carboniferous limestone giving bimodality to biosphere data in this area and the food procured by humans. The Carboniferous limestone is well constrained providing a plant biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7094 ± 0.001 ($n=22$, 2 SD; Evans *et al.* 2018) whilst the Millstone Grit biosphere provides 0.7117 ± 0.0014 ($n=11$, 2 SD; Evans *et al.* 2018). At early medieval Masham in North Yorkshire, one of the few cemeteries located on Millstone Grit with published human Sr isotope data, local human values (enamel and dentine) ranged from 0.7096 to 0.7112 (Buckberry *et al.* 2014), consistent with the biosphere data given above.

The bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from Millstone Grit is 0.71147 which is within the 2 SD range of predicted biosphere values for this rock type. Thus, the ingestion of Millstone Grit by people using grinding stones made of this rock or through ingestion of soil overlying Millstone Grit would not create any anomalous values in their skeletal tissues. If detrital mica were preferentially plucked out of a Millstone Grit quern or millstone during grinding and ingested (as was proposed for the micas in the Alpine Ice Man; Müller *et al.* 2003) the maximum bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ value that could be achieved is 0.71450, in the absence of any other mineral contribution.

The Forest of Dean (Gloucestershire) was a recognized source for querns (Peacock 2013, 62–65). The main lithologies in this area are the Siluro-Devonian Old Red Sandstones (ORS), the

Carboniferous Coal Measures and the Carboniferous limestones. The ORS in this area has a plant biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.712 ± 0.002 ($n=9$, 2 SD; Chenery *et al.* 2010; Johnson 2017) and the Coal Measures give a plant biosphere of 0.712 ± 0.001 ($n=4$, 2 SD; Chenery *et al.* 2010; Johnson 2017). As expected for marine carbonates formed primarily from shell, Carboniferous limestones in this location produce a lower and much less variable plant biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.7103 ± 0.0002 ($n=3$, 2 SD; Johnson 2017), but which nevertheless is relatively high for Carboniferous limestones (Evans *et al.* 2018).

The Pennant sandstone is part of the Carboniferous Coal Measures. The bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from Pennant sandstone has a value of 0.71281, which is comparable with the values obtained from plants growing on the Coal Measures and the ORS and medieval humans from sites such as Hereford Cathedral, whose $^{87}\text{Sr}/^{86}\text{Sr}$ values range predominantly from 0.712 to 0.713 (Evans *et al.* 2012). Therefore, as with the Millstone Grit, the use of locally sourced grinding stones would not cause any anomalously high $^{87}\text{Sr}/^{86}\text{Sr}$ values in human skeletal tissues.

The importation of Millstone Grit or Pennant sandstone grinding stones, or indeed any grinding stone, into a region of different geology may be problematic to provenance studies if the receiving biosphere is characterized by significantly lower or higher $^{87}\text{Sr}/^{86}\text{Sr}$. In this scenario there is the potential for the ingested rock to contribute non-local dietary $^{87}\text{Sr}/^{86}\text{Sr}$. In some localities, both the Millstone Grit and Pennant sandstone can occur in proximity to Carboniferous limestones regions and in this scenario the bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from these imported grinding stones will be approximately 0.002 higher than the $^{87}\text{Sr}/^{86}\text{Sr}$ of plants sourced from limestone lithologies, that is, 0.7094 ± 0.001 ($n=22$, 2 SD; Evans *et al.* 2018). Abrasive sandstones, particularly the prized Millstone Grit, have been extensively traded and transported as grinding stones in Britain (Peacock 2013, 62–65) and the potential thus exists for their rock grit to be unintentionally consumed in a variety of geological regions. For Millstone Grit or Pennant sandstone, however, neither would raise human skeletal $^{87}\text{Sr}/^{86}\text{Sr} > 0.71281$ and cannot thus explain human $^{87}\text{Sr}/^{86}\text{Sr}$ of ≥ 0.713 .

The Eskdale granite crops out in the Eskdale area of the Lake District, which has an old and complicated geological structure. The sedimentary rocks (Ordovician mud-, silt- and sandstones) host a plant biosphere $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.711 ± 0.002 ($n=5$, 2 SD) and the Ordovician lavas and tuffs give 0.710 ± 0.002 ($n=6$, 2 SD; Johnson 2017). Plants growing on the Eskdale granites gave $^{87}\text{Sr}/^{86}\text{Sr}$ values of 0.7105, 0.7130 and 0.7147 (Johnson 2017). The bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ of Eskdale granite is 0.72224, which considerably exceeds the $^{87}\text{Sr}/^{86}\text{Sr}$ range of plants naturally extracting from this lithology. Unlike the Pennant sandstone and Millstone Grit, this granite has the potential to raise human $^{87}\text{Sr}/^{86}\text{Sr}$ values beyond any current $^{87}\text{Sr}/^{86}\text{Sr}$ biosphere values found within Britain (Evans *et al.* 2018). However, the outcrop area of the Eskdale granite is insignificant compared with that of Carboniferous sandstones and gritstones and granites were not commonly used as grinding stones in Britain and were known for being difficult to cut (Nicholson and Burn 1777, 477; Coope 1979, 100); from a functional and practical viewpoint, why seek out a stone that is rare, less accessible and does not do the job as well as the abrasive sandstones, such as Millstone Grit, which were readily available in most regions?

Beyond the British Isles, other igneous lithologies, such as volcanic lavas, have been favoured for milling and grinding (Hunt and Griffiths n.d.; Ebeling and Rowan 2004). Grinding stones of basaltic Mayen Lava quarried in Germany are found across North-Western Europe (Pohl 2010; Peacock 2013, 151–153), while the metates quarried and transported across South and North America include basalts, rhyolites, andesites and their larger crystal and metamorphic equivalents, but quartzite, sandstones and limestone are also very common in Mexico and South America (Ebeling and Rowan 2004; Searcy 2011, 82). Basaltic rocks have the lowest whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.704–0.708 (Faure and Powell 1972) and will contribute this $^{87}\text{Sr}/^{86}\text{Sr}$ to both

the biosphere and the human gut. Conversely, granitic rocks can have very high whole-rock $^{87}\text{Sr}/^{86}\text{Sr}$ values, sometimes > 1.0 (Faure and Powell 1972; Peterman and Hildreth 1978; Rundle 1979; Faure 1986) and will, as shown by the Eskdale granite in this study, contribute higher bio-accessible $^{87}\text{Sr}/^{86}\text{Sr}$ in the presence of strong acid than when weathered through natural processes. The bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ of these multi-mineralic igneous rocks will thus depend on which minerals are contributing Sr during their passage through the gut. In such cases, their contribution to human $^{87}\text{Sr}/^{86}\text{Sr}$ needs to be considered carefully and an appraisal of the amount of ingested rock grit required to change human $^{87}\text{Sr}/^{86}\text{Sr}$ is expedient.

Food versus rock grit: modelling the amount of ingested rock grit required to change human $^{87}\text{Sr}/^{86}\text{Sr}$

Although the results demonstrate that there is potential for human $^{87}\text{Sr}/^{86}\text{Sr}$ to be changed by ingested grit, particularly if using imported ‘exotic’ stones, it will never be the case that grit is the only thing ingested. When, therefore, can ground rock (or minerals), produced from grinding stones, realistically affect the Sr isotope contribution to the human diet? The controlling factors are the difference in isotope composition between the bioaccessible component of the rock (or mineral) and the main Sr isotope composition of the diet, the relative concentration of Sr in both components, and the amount of rock that can sensibly be incorporated within the diet. In order to model such an eventuality, an assessment of the amount of ground rock that would be produced and incorporated into food such as bread is required.

Estimates of quern and millstone wear and erosion rates will depend on the hardness of the rock being used (Biot 2011; Buonasera 2015), but an estimated contribution of 1–2% grit from the grinding stones in milled flour seems to be realistic. Biot (2011) states that around 2% of greensand sandstone was present in grain ground by these querns. Given that 4 kg of flour is the maximum that can be produced from one quern in 1 h (Peacock 2013, 126–128), around 80 g (2%) could be rock grit. Once the flour is produced, the potential percentage grit component in the diet is further diluted by the other dietary components, including those used in bread making. The calculations that follow are based on bread in the medieval period of Britain and are a *theoretical model* of how the unintentional ingestion of grit from grinding stones in flour through the consumption of bread could contribute to human $^{87}\text{Sr}/^{86}\text{Sr}$ values. Although predicated on historically documented British diets, such calculations may be applicable for different diets and in other regions, for example, where the staple food is rice or maize.

There are a variety of bread recipes known from medieval Britain, often ranked by the quality of flour used to make the loaves. The ingredients below produce one large wholemeal bread loaf, converted to decimal measurements (Brears 2015, 125–131):

- 900 g (strong) wholemeal flour.
- 550 ml water at 24°C.
- 15 g dried yeast (instead of the sourdough culture they would have used).
- 15 g salt.

In this loaf, a maximum of 2%, that is, 18 g, of the wholemeal flour can be considered rock grit. During the baking process approximately 10–20% of the water content can be lost through evaporation (Hamelman 2004), which is approximately 55–110 mL of the water in the loaf recipe above. Assuming that none of the other ingredients are contaminated with grit and 20% of the water is lost through evaporation in the baking process, the grit makes up approximately 1.3% of this bread loaf (by mass). Bread consumption in the medieval period was known to be particularly high. An upper limit for the proportion of bread in the medieval diet was about 74% (by

mass and calorific intake: Dyer 1988, 25–27) and if 1.3% of the bread was grit, about 0.96% of the total diet could be rock grit from the milling process.

Consequently, if this rock grit is to impact on dietary Sr and affect human $^{87}\text{Sr}/^{86}\text{Sr}$ values, it needs to be able to change significantly the Sr isotope composition of the diet when present at about 0.96%. The contribution to the diet can be calculated using a two-component, or end-member, mixing diagram (Faure 1998). For an individual who used stones composed of Millstone Grit (component 1) to grind grain, the bioaccessible component of the ground rock ingested would contribute $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.71147 at 3 ppm Sr concentration in the digestive tract. If, in this scenario, the individual were procuring and consuming plants grown in a region of Chalk (component 2), the plants consumed as the bulk of the diet can be estimated to have a mean $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.70808 and 9 ppm Sr concentration (based on Warham 2011, 88).

Table 2 shows the results of mixing component 1, the ground Millstone Grit, with component 2, plants grown in soil overlying Chalk bedrock from 0% to 100% contribution. At 0.96% grit the $^{87}\text{Sr}/^{86}\text{Sr}$ contribution to human diet has only been increased by 0.00001. At the fifth significant figure, this change in the $^{87}\text{Sr}/^{86}\text{Sr}$ value can be considered of little importance: it is largely within 2 SD measurement error. Moreover, for mobility and provenance studies a change to the fourth significant figure of the ratio can be considered irrelevant: co-habiting siblings can vary by 0.0002 (Montgomery 2002, 146) and cattle raised in the same herd by 0.0006 (Towers 2013, 123–124). Even if the Millstone Grit is replaced by the considerably more radiogenic Eskdale granite ($^{87}\text{Sr}/^{86}\text{Sr}$ of 0.72224 and 3 ppm) the change is still in the fifth significant figure at a 0.96% contribution, increasing the $^{87}\text{Sr}/^{86}\text{Sr}$ only by 0.00004.

Up to 8% of the diet needs to be from ground Millstone Grit before the $^{87}\text{Sr}/^{86}\text{Sr}$ is shifted by 0.0001. To achieve a significant change of 0.001 requires a 56% contribution, which is

Table 2 Results of the mixing model from 0% to 100% contribution of component 1 (ground Millstone Grit) to component 2 (plants grown in soil overlying chalk bedrock)

Percentage component 1	$^{87}\text{Sr}/^{86}\text{Sr}$ mixture
0.00	0.70808
0.50	0.70809
0.96	0.70809
1.00	0.70809
5.00	0.70814
8.00	0.70818
10.00	0.70820
20.00	0.70834
30.00	0.70850
40.00	0.70870
50.00	0.70893
56.00	0.70909
60.00	0.70921
70.00	0.70956
80.00	0.71002
90.00	0.71062
100.00	0.71147

The 0.96% contribution is highlighted in bold.

extremely unrealistic (Table 2). For the Eskdale granite, a 2% contribution of ground rock in the diet is needed to change the $^{87}\text{Sr}/^{86}\text{Sr}$ by 0.0001 and a 19% contribution to change it more significantly by 0.001. These are of course only approximated percentages and a 2–8% unintentional grit component in the diet may not seem unrealistic. However, the calculations are based on a maximum possible grit production in the milling process and medieval diet, when records document a high consumption of bread (up to 74%; Dyer 1988, 25–27). It would seem to be difficult to introduce unintentionally more than 1% contribution of rock into the human diet. Therefore, although ground rock (or minerals), via the use of querns and millstones, can provide a source of non-local bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ they would be very rarely consumed in sufficient quantity to have an impact on human $^{87}\text{Sr}/^{86}\text{Sr}$ values and result in the false identification of an individual as non-local.

There may, of course, be exceptions to this if an individual was intentionally ingesting soil and grit, for example, pica and geophagy, or if food were being deliberately contaminated. For example, it was not uncommon for 19th-century bakers to add chalk (CaCO_3) or other substances to their bread flour (Accum 1820) and the nutritionally beneficial Mesoamerican practice of nixtamalization involved cooking with lime or limestone (also CaCO_3) before grinding (Coe 1994, 14; Ellwood *et al.* 2013). Pica and geophagy, the practice of consuming non-food substances, is remarkably prevalent across human cultures and time particularly, and crucially given when deciduous and permanent tooth enamel is mineralizing, in pregnant women and children (Young 2012; Henry and Cring 2013, 189–191). The consumption of earth or soil-like substances via geophagy typically consisted of clays and chalks (Young 2012, 95; Henry and Cring 2013, 189–191). If certain individuals did intentionally consume clays or chalks in Britain, $^{87}\text{Sr}/^{86}\text{Sr}$ ranging from 0.7091 ± 0.0022 ($n=51$, 2 SD) for Cretaceous clays to 0.7106 ± 0.0022 ($n=10$, 2 SD) for Carboniferous clays and 0.7083 ± 0.0012 ($n=85$, 2 SD) for the chalks (Evans *et al.* 2018) could potentially be bioaccessible. The bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from clays and particularly the homogenous carbonate rocks in Britain, which include the chalks, are likely to follow the same pattern shown by the Millstone Grit and Pennant sandstone in this study. Thus, if clays or chalks were consumed locally, it is unlikely that they would create any anomalous $^{87}\text{Sr}/^{86}\text{Sr}$ values in the human skeletal tissues. If clays or chalks were consumed in different geological regions with a significantly lower or higher $^{87}\text{Sr}/^{86}\text{Sr}$ biosphere, then the human $^{87}\text{Sr}/^{86}\text{Sr}$ could diverge from the expected local biosphere. If the mixing model used above was rerun with Chalk rock grit consumed in a Millstone Grit biosphere, only 13% of the diet would need to be from the deliberate consumption of grit in order to lower the $^{87}\text{Sr}/^{86}\text{Sr}$ significantly, that is, by 0.001. However, the uptake of heavy metals such as Sr and lead (Pb) is suppressed in high-calcium (Ca) diets (Burton and Wright 1995; Underwood 1977) and hard (high-Ca) water areas (Mahaffey 1978) and thus even if ingested carbonate-dominated clays and chalks released Sr in the stomach, it will not necessarily be absorbed by the body if co-ingested with the high levels of calcium present in chalk. Indeed, in Britain, archaeological humans with enamel $^{87}\text{Sr}/^{86}\text{Sr}$ indicative of chalk and limestones tend to exhibit the lowest enamel Sr concentrations (Montgomery *et al.* 2014; Montgomery *et al.* 2019). A similar situation may arise when limestone or slaked lime is intentionally added to maize during the preparation and cooking process: this has been shown to increase the calcium content of the maize by approximately 750% (Serna-Saldívar *et al.* 1987).

The practice of geophagy can be extremely difficult to recognize in the archaeological record (Henry and Cring 2013, 190) and is not well documented in Britain. Therefore, if an archaeological individual excavated in Britain has a non-local $^{87}\text{Sr}/^{86}\text{Sr}$ value, it appears more realistic to infer that they are non-local rather than participating in geophagy or pica, unless evidence

strongly suggests otherwise. Such an observation may also apply to humans in many other regions of the world.

CONCLUSIONS

The UBM method has shown that Sr in rock grit, whether accidentally ingested via the use of grinding stones or deliberately through pica or geophagy, is rendered bioaccessible by the strong acids of the human gut. However, a significant alteration of human skeletal $^{87}\text{Sr}/^{86}\text{Sr}$ as a result of ingested rock Sr circumventing the transmission via plants is likely to occur only in unusual behaviours such as pica, geophagy or the deliberate contamination of food. In such circumstances, clays, chalks or limestones are commonly used and it is estimated that they would need to comprise > 13% of the diet (by mass and calorific intake) on a regular basis to significantly shift human skeletal $^{87}\text{Sr}/^{86}\text{Sr}$.

Rock grit unintentionally consumed as a result of using quern or millstones to grind grain is unlikely to equate to > 1% of the diet and bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ from Millstone Grit or Pennant sandstone falls within the observed biosphere ranges for these rocks. The Eskdale granite produces bioaccessible $^{87}\text{Sr}/^{86}\text{Sr}$ which exceeds that available from plants in granitic regions. Nonetheless, at the 1% level, regardless of the grinding stone's lithology or the local Sr isotope biosphere, ingested grit will not result in a significant change in human $^{87}\text{Sr}/^{86}\text{Sr}$ values. Consequently, the use of querns or millstones and the regular unintentional consumption of their grit, whether of locally derived or imported rock, will have a negligible effect on human $^{87}\text{Sr}/^{86}\text{Sr}$ data and will produce neither anomalously high skeletal $^{87}\text{Sr}/^{86}\text{Sr}$ values nor false migrants. Even the intentional consumption of rock, clays or soils through geophagy or pica, which is difficult to identify in the archaeological record, is unlikely to affect skeletal $^{87}\text{Sr}/^{86}\text{Sr}$ adversely. Moreover, this deliberate targeting of high-Ca clays and chalks or preparation of maize with limestone, which tend to be characterized by unradiogenic $^{87}\text{Sr}/^{86}\text{Sr}$, is also an unlikely source of either radiogenic and, due to suppression of Sr uptake in the presence of high-dietary Ca, unradiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ in archaeological humans.

The results of this study thus provide reassurance to researchers using Sr isotope analysis to identify migrants in Britain and beyond that non-local or unusually high $^{87}\text{Sr}/^{86}\text{Sr}$ values cannot be explained by the direct ingestion of rock grit, clays or soils.

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