OPENQUATERNARY

Widga, C et al 2017 Variability in Bioavailable ^{87}Sr/^{86}Sr in the North American Midcontinent. *Open Quaternary*, 3: 4, pp. 1–7, DOI: https://doi.org/10.5334/oq.32

RESEARCH PAPER

Variability in Bioavailable ^{87}Sr/^{86}Sr in the North American Midcontinent

Chris Widga*, J. Douglas Walker† and Andrew Boehm‡

Strontium (Sr) isotope tracers are useful for understanding provenance and mobility in biological materials across multiple disciplines. However, the impact of these techniques is highly dependent on the construction of appropriate comparative baselines (i.e., an isoscape). We present the results of a systematic survey of ^{87}Sr/^{86}Sr values from grasses in the North American Midcontinent with a particular emphasis on sedimentary systems. Although ^{87}Sr/^{86}Sr values are highly variable across the region, the Sr isoscape shows multi-scalar patterns that are dependent on local-to-regional trends in surficial geology. High values are found in bedrock-dominated areas such as the Black Hills (SD) and Ozark Uplift (MO), or formerly glaciated areas where surface deposits are dominated by ice-transported Precambrian clasts. The lowest values are found in river valleys that incorporate eroded Neogene sediments into terrace formation. Intermediate values are found in upland loess and alluvial deposits which blanket much of the study area. We demonstrate trends in large-scale variability of the Midcontinent's ^{87}Sr/^{86}Sr isoscape and suggest that future refinement focus on sub-regional trends in Sr isotope variability.

Keywords: ^{87}Sr/^{86}Sr; isoscapes; isotopic tracers; Midcontinent

Introduction

Isotopic tracers are useful in provenance and mobility applications across many different disciplines (Bentley, 2006; Crowley et al., 2015). The ratio of ^{87}Sr/^{86}Sr has proven invaluable to these studies since it reflects geological context and does not exhibit fractionation across trophic levels (Hoppe et al., 1999; Price et al., 2002). In addition, because the Rb to Sr ratio is so low and the time involved in studies so short (typically on Holocene or Late Pleistocene materials), the ^{87}Sr/^{86}Sr ratio does not evolve by radioactive decay and can be treated as essentially a stable isotope (Bowen and West, 2008). However, the impact of these techniques is highly dependent on the quality of baseline comparative data (i.e., isoscapes), complicating its use in areas dominated by allochthonous surface sediments of alluvial, eolian, or glacial origin.

The North American Midcontinent is one of these areas (**Figure 1**). Extensive glaciation during the Late Pleistocene and the subsequent geomorphic history of the region means that bedrock is often deeply buried and contributes little to surface soil parent material. Surface deposits are a heterogeneous mix of eolian, alluvial, and glaciogenic origin. Although there have been previous attempts to model regional ^{87}Sr/^{86}Sr composition in the region (Beard and Johnson, 2000; Bataille and Bowen, 2012), the resolution of surface geological mapping and the wide variability in surface geomorphology, even within small areas, have complicated efforts to resolve a Sr isoscape for surface materials. Although direct surface sampling of plants, animals, and sediments (Price et al., 2007; Hedman et al., 2009; Widga et al., 2010; Slater et al., 2014) offers some insight into regional Sr distribution patterns, these efforts have been limited to small parts of the study area and are opportunistic surveys undertaken independent of local geomorphic information.

Variability in surface ^{87}Sr/^{86}Sr is controlled by geological processes operating at two levels. First-order variability is determined by the source rock of surface deposits. Generally, ^{87}Sr/^{86}Sr of glaciogenic deposits is determined by the provenance of glaciogenic materials and subsequent landscape evolution (Taylor and Faure 1981, Widga et al., 2010), whereas ^{87}Sr/^{86}Sr of pre-glacial bedrock sources may be influenced by the age of parent rocks (Beard and Johnson, 2000), degree of crustal recycling (Bataille et al., 2014), progressive evolution of marine Sr reservoirs (McArthur et al., 1994), regional volcanism (Leeman 1982), or a number of other processes. Second-order variability is determined by local-scale geomorphological processes occurring within a geologic unit (Carpenter et al., 2003, Faure 1986, Widga et al., 2010).

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* Center of Excellence in Paleontology, East Tennessee State University, 1212 Suncrest Dr., Gray TN, 37615, US

† Department of Geology, University of Kansas, 1475 Jayhawk Blvd., Lawrence, KS 66045, US

‡ Museum of Natural and Cultural History, University of Oregon, Eugene, OR, 97403, US

Corresponding author: Chris Widga (widgac@etsu.edu)

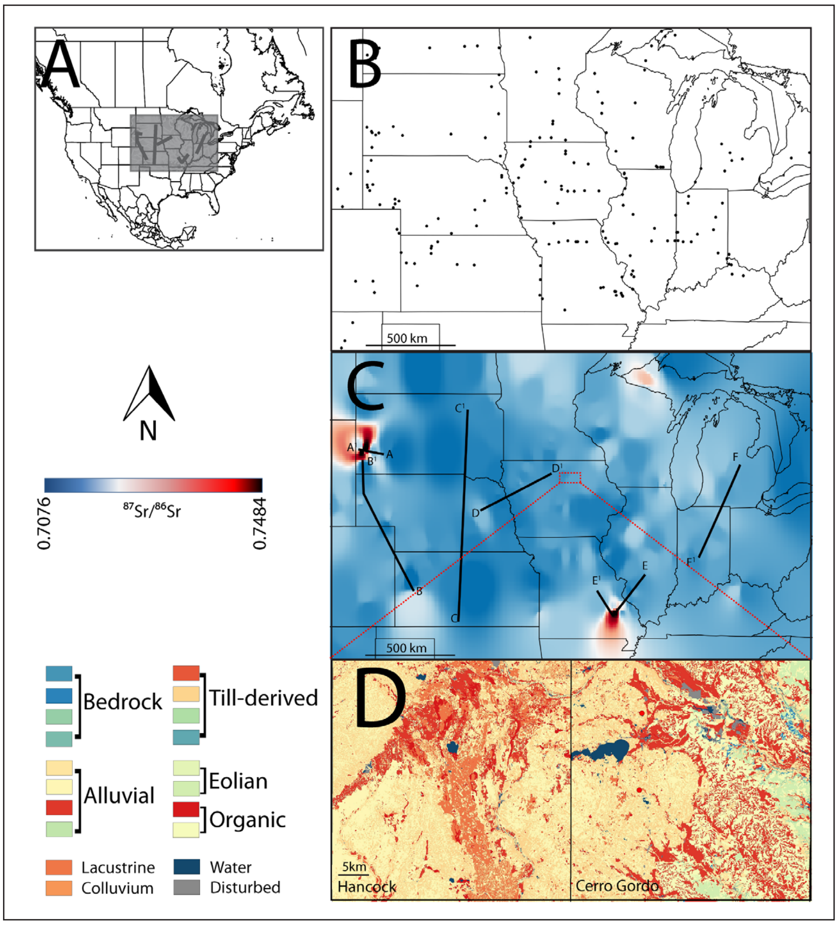

Figure 1: A) Location of study area. **B)** Sample collection sites. **C)** Rendered ^{87}Sr/^{86}Sr surface, with profile locations. **D)** Surface geology of the Des Moines Lobe as expressed in Hancock and Cerro Gordo counties, northern Iowa (USA) showing variability in parent material of surface sediments (Miller et al., 2008).

Strontium is a common element found in geological, hydrological, and ecological systems at the Earth's surface. It has four, naturally occurring stable isotopes (Faure, 1986): ^{84}Sr, ^{86}Sr, ^{81}Sr, and ^{88}Sr. The isotope ^{87}Sr, however, is the radiogenic daughter isotope of ^{87}Rb which has a half-life of 48.8×10^9 yr. The ratio of this decay product (^{87}Sr) to the stable isotope ^{86}Sr increases as the age of a rock formation increases. Due to its relatively large atomic mass, ^{87}Sr/^{86}Sr changes little as it passes from weathered bedrock, to soil parent material, and ultimately to plants and primary consumers (Flockhart et al., 2015). This ratio is dependent on the age and Sr content in soil parent material and atmospheric inputs (Beard and Johnson, 2000; Bataille et al., 2012; Bataille and Bowen, 2012; Reynolds et al., 2012). Geomorphic processes such as weathering and transport of parent rocks complicates surface ^{87}Sr/^{86}Sr distribution in many parts of the world where allochthonous sediments comprise soil parent material.

Within the Midcontinental study area, ^{87}Sr/^{86}Sr values are determined by the sediment source(s), local weathering processes, and surface geomorphology. The region is drained by a number of large rivers. Valley alluvium reflects a variety of local and non-local sources, including upstream deposits. Major basins in the study area include the Mississippi, Illinois, Ohio, Wabash, Missouri, Platte, Kansas, and Arkansas River basins. In the Great Plains, these rivers begin in the foothills of the Rocky Mountains and carry a sediment load reflecting these origins. For instance, the upper Missouri River drains extensive Neogene deposits in Montana and western North Dakota. The potential contribution of these young rocks on downstream alluvial deposits is evident in the lower Missouri valley ^{87}Sr/^{86}Sr in Nebraska and Iowa (Widga et al., 2010). Alternatively, erosion of local shale or carbonate units may contribute significantly to valley Sr ratios. In eastern parts of the study area, terrace fill is commonly derived from glaciogenic sediments such as reworked glacial till or loess deposits.

Loess covers a large portion of the study area and can reach depths of >40 m (Bettis et al., 2003). The source for loess deposits is in nearby river valleys, although long-distance transport of clay and silt-sized particles can occur (Muhs and Bettis, 2000; Muhs et al., 2013). The prevailing wind direction for most of the study area is from the northwest. Therefore the source area for loess deposits in the Great Plains are typically from non-glaciogenic sources (Mason, 2001) while loess in areas covered by recent glacial deposits reflects a glacial origin via large river valleys (Muhs and Bettis, 2000). Both regions may be susceptible to long-distance transport of dust (Mason and Jacobs, 1998).

Spatial variation in surface Sr isotope ratios provide the basis for many recent studies of human and animal mobility, as well as source estimates for sedimentary deposits. Small mammals and animals with limited mobility are typically used to assess the range of local variation in archaeological mortuary populations (Price et al., 2007; Hedman et al., 2009; Slater et al., 2014). This study however, relies on the expression of surface $^{87}Sr/^{86}Sr$ in regional grasses, an approach previously used to infer mobility middle Holocene bison populations from the eastern Great Plains (Widga et al., 2010).

Materials and methods

Sampling occurred opportunistically over the course of more than a decade. The distribution of samples reflects large-scale differences in regional landscape evolution, such as differences in loess deposition (Bettis et al., 2003), the evolution of alluvial systems (Mandel, 2008), and glaciogenic landscapes (Ehlers and Gibbard, 2004). Grass samples were collected from lithologically defined locations along roadsides or within natural areas. Locations were selected to avoid the effects of agricultural runoff, non-local rock products (e.g., roads surfaced with crushed limestone), or modern earth moving activities. Grasses are an ideal material to assess levels of bioavailable Sr because 1) they form the base of regional terrestrial foodwebs and incorporate $^{87}Sr/^{86}Sr$ in similar proportion to the soil parent material (Capo et al., 1998); 2) they are minimally impacted by atmospheric inputs of Sr due to low leaf area; and 3) they are widespread and common elements of the regional flora.

The emergent portion of grasses was collected and rinsed in DI water. Dried grass samples were then ashed at 425°C in platinum crucibles. The ashed grasses were dissolved in 3.5 N HNO_3 and eluted through Eichrom Sr-spec ion-exchange columns. The purified Sr was then analyzed for $^{87}Sr/^{86}Sr$ ratios on a Thermal Ionization Mass Spectrometer

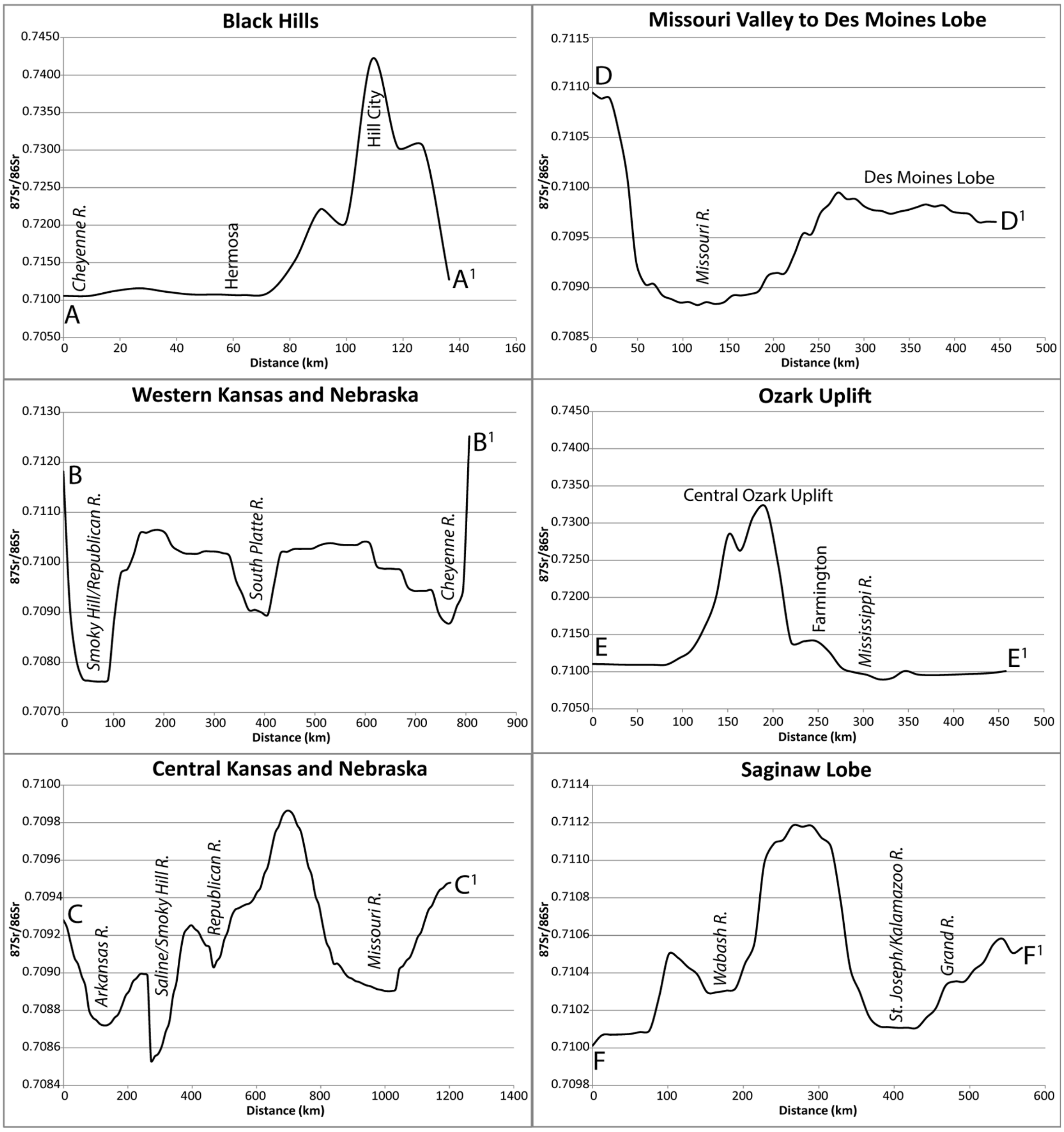

Figure 2: Profiles of surface $^{87}Sr/^{86}Sr$ values along well-sampled transects. See Figure 1C for profile locations.

(TIMS), an automated VG Sector or VG Sector 54 in the University of Kansas Isotope Geochemistry Laboratories. Isotope ratios were adjusted to correspond to a value of 0.71250 on NBS-987 for ^{87}Sr/^{86}Sr. Over 120 runs of NBS 987 were made along with the samples. Standard deviation of results is about 11 ppm on ratio, and standard error is miniscule. We also assumed a value of ^{86}Sr/^{88}Sr of 0.1194 to correct for fractionation.

The final ^{87}Sr/^{86}Sr dataset consists of 190 grass samples from all major physiological provinces in the study area. QGIS 2.10 (Pisa) was used to interpolate and render ^{87}Sr/^{86}Sr as a continuous surface. ^{87}Sr/^{86}Sr data was interpolated using an inverse-distance-weighted algorithm (power = 3; search radius = 0) with no smoothing. An IDW algorithm was preferred over an ordinary kriging method because ^{87}Sr/^{86}Sr of surface sediments throughout most of the study area vary continuously. Unfortunately, this method produces artifacts in areas of discrete variation such as the Ozark uplift and Black Hills. Future studies that seek to understand second-order variability in these areas should take this into account. Surface profiles in well-sampled areas were rendered with the Profile Tool (ver. 3.7.0) plug-in.

Results

Surface Sr isotope values vary significantly across the Midcontinent (**Figure 1**). The most radiogenic samples were from Precambrian igneous outcrops in the Black Hills and Missouri Ozarks (Harney Peak Granite, SD: 0.7484; Graniteville Granite, MO: 0.7330). Outlying areas of these bedrock-dominated landscapes show lower values consistent with younger carbonate rocks (0.7139–0.7179). Glaciogenic sediments in the upper Midwest containing shield-derived clasts also exhibit high values (0.7162–0.7122). The lowest ^{87}Sr/^{86}Sr values are found in the western part of the study area, where valley alluvium and uplands are dominated by Neogene sources and loess-covered landscapes along the Missouri River and the eastern prairies (0.7079–0.7120). Mississippian and Pennsylvanian limestones in Illinois, Missouri, and eastern Kansas (0.7093–0.7085) and Cretaceous limestone in western Kansas (0.7076) also exhibit low ^{87}Sr/^{86}Sr values. Although not mutually exclusive, these geologic packages represent large-scale Sr isotope variability in surface materials across the Midcontinent.

Sampling resolution in the study area was not high enough to establish sub-regional patterns within larger geologic packages. However, it is possible to illustrate local changes in well-sampled or highly variable areas (**Figure 2**). Although ^{87}Sr/^{86}Sr profiles constructed across the Sr isoscape show expected changes across older landscapes such as the Ozarks and Black Hills, locally significant variability in ^{87}Sr/^{86}Sr also occurred in other parts of the study area where alluvial valleys show local, negative excursions when compared to surrounding uplands. This is true for landscapes in the Great Plains, where uplands are loess-capped bedrock, as well as glaciated parts of the Midwest, where uplands consist of till and loess (**Figure 2**).

Discussion

Ancient landscapes comprised of Precambrian igneous and metamorphic rocks with little alluvial or eolian surface deposits are isotopically heterogeneous and exhibit the highest ^{87}Sr/^{86}Sr values in the study area. Widespread limestones outcropping near the surface in Kansas, Missouri, and Illinois show significantly lower ^{87}Sr/^{86}Sr values (consistent with the seawater curve; McArthur et al., 2001) than crystalline rocks in the Ozark Uplift and Black Hills. Since these rock types are easily eroded, they potentially comprise more of the bedload in regional rivers and streams.

In glaciated areas, glacial till exerts a strong influence on Sr isotope variability. For instance, glaciogenic deposits sourced in the granite-dominated Precambrian Canadian Shield show higher ^{87}Sr/^{86}Sr values than tills with source areas dominated by Cretaceous-aged Dakota sandstone. In these areas, loess and lacustrine sediments are also predominantly (although not entirely) derived from glacial sources. Local-scale variability in the characteristics of surface sediments from glaciated areas is much higher than the sample resolution in this dataset (**Figure 1D**). Therefore

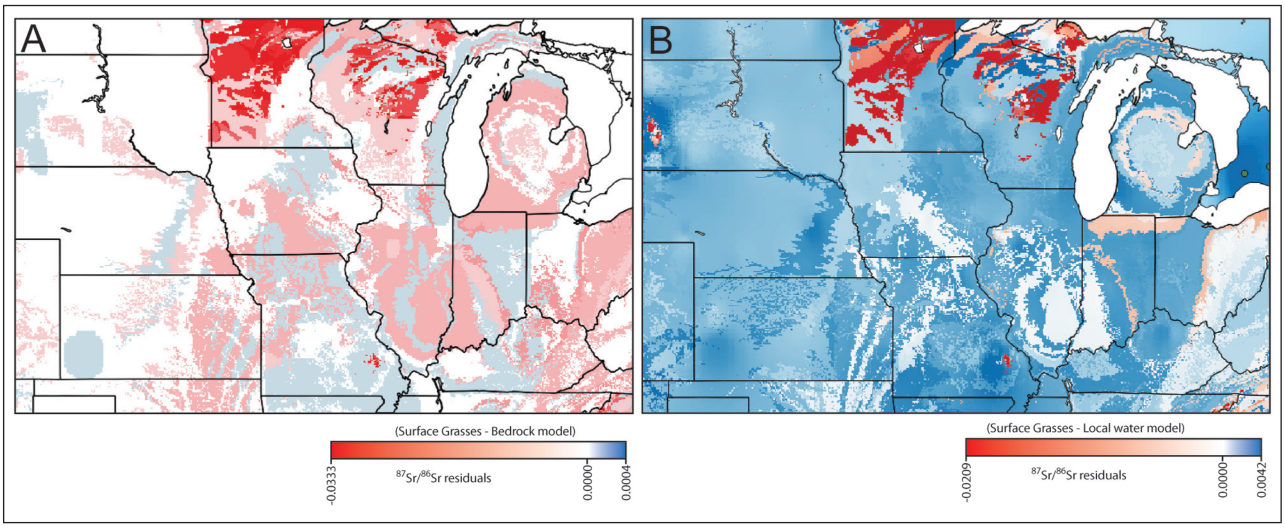

Figure 3: Residual analysis of observed ^{87}Sr/^{86}Sr (grass) and isoscapes derived from **A)** bedrock, and **B)** local water models (Bataille and Bowen, 2012).

second-order variability (e.g., variability of ^{87}Sr/^{86}Sr within geologic units) is beyond the scope of this study.

The range of ^{87}Sr/^{86}Sr values in this study are similar to previously published data from the region utilizing small or non-migratory animals (Price et al., 2007; Hedman et al., 2009; Slater et al., 2014), sediments, and grasses (Widga et al., 2010). However, the spatial scale of this isoscape is much greater and demonstrates the potential for significant local variability in surface sediments.

Comparisons can be made between this surface dataset and bedrock-derived Sr isoscapes (**Figure 3**). Bataille and Bowen (2012) built two large-scale, process-driven isoscapes using widely available geological data (available at wateriso.utah.edu/waterisotopes, accessed on 21 April, 2017). The resulting coverages reflect 1) age of bedrock (Beard and Johnson, 2000; Figure 6B in Bataille and Bowen, 2012) and 2) local water (Figure 9A in Bataille and Bowen, 2012). Due to the presence of deep surficial sediments in the Great Plains and Midwest, residual maps of surface ^{87}Sr/^{86}Sr values presented in this study and these bedrock-derived isoscapes show significant differences. These differences are especially pronounced in areas of deep glaciogenic deposits in the upper Midwest where bedrock models overestimate ^{87}Sr/^{86}Sr values by up to 0.0333. The bedrock model more closely aligns with the surface data in unglaciated parts of the Great Plains, but both positive and negative residuals are present in the Midwest, where loess-dominated landscapes reflect the source of the loess, not the underlying bedrock.

These data illustrate the difficulty in constructing regional Sr isoscapes. Although continent-scale changes can be estimated through bedrock derived models, these models cannot presently account for highly variable surface expression of ^{87}Sr/^{86}Sr, especially in areas exposed to significant deposition of loess, alluvium, and glacial till. Provenance and mobility studies in these areas should take this into account.

Conclusion

Sr isotope tracers are increasingly useful for understanding a broad range of ecological and geological questions. In many applications, such as understanding wildlife movement patterns (Hoppe et al., 1999; Balasse et al., 2002; Hoppe, 2004; Hoppe and Koch, 2007; Britton et al., 2009; Britton et al., 2011) or forensic provenance (Beard and Johnson, 2000; Degryse et al., 2012), it is important to establish the maximum degree of variability in bioavailable Sr. We illustrate the structure of ^{87}Sr/^{86}Sr variability across the Midcontinent, encompassing a wide range of surface geology, vegetation, and terrain.

For mobility and provenance studies, bedrock and water models of ^{87}Sr/^{86}Sr distribution are inadequate for the region due to extensive coverage of allochthonous alluvial, eolian, and glaciogenic sediments. These surfaces are complex, requiring direct sampling of surface deposits, or vegetation growing on surface deposits. Unsurprisingly, the surface dataset presented here differs significantly from bedrock-derived models. Although we would expect the models to be more in agreement in areas without significant surface deposition, this is not always the case. Further refinement of all ^{87}Sr/^{86}Sr isoscape models for the Midcontinent region is required.

Additional File

The additional file for this article can be found as follows:

- **Table S1.** Midcontinental grass ^{87}Sr/^{86}Sr data. DOI: https://doi.org/10.5334/oq.32.s1

Acknowledgements

Kayla Kolis, Josh Feldman, and Lisa Stockli assisted with laboratory analyses. This project was funded by NSF #1050638, NSF #0532363, and the Quest Archaeological Research Program (Southern Methodist University, Dallas, TX).

Competing Interests

The authors have no competing interests to declare.

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How to cite this article: Widga, C, Walker, J D and Boehm, A 2017 Variability in Bioavailable ^{87}Sr/^{86}Sr in the North American Midcontinent. *Open Quaternary*, 3: 4, pp. 1–7. DOI: https://doi.org/10.5334/oq.32

Submitted: 08 March 2017 **Accepted:** 20 May 2017 **Published:** 07 June 2017

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