

Advances in global bioavailable strontium isoscapes

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Abstract

Strontium isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) are a popular tool in provenance applications in archeology, forensics, paleoecology, and environmental sciences. Using bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ in provenance studies requires comparing the $^{87}\text{Sr}/^{86}\text{Sr}$ of a sample of interest to that of $^{87}\text{Sr}/^{86}\text{Sr}$ baselines. Historically, these baselines required building empirical datasets from plants or local animals to characterize the $^{87}\text{Sr}/^{86}\text{Sr}$ available to local ecosystems (bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$). However, researchers are increasingly relying on modeled bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ maps (called isoscapes). We review the advantages and limitations of existing approaches to mapping bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ for provenance studies and propose a globally applicable, scalable, and editable framework for creating bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes. This framework relies on: 1) Compiling global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data; 2) Mapping $^{87}\text{Sr}/^{86}\text{Sr}$ variability in rocks; 3) Leveraging global environmental covariates; and 4) Applying a random forest regression method that integrates these data to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. When the random-forest model is applied at the global scale it performs well (explaining 60% of the variance of the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset), and accounts for geological, geomorphological and atmospheric controls. In data-rich regions (e.g., Europe), the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape can be successfully extrapolated to broad regions without bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data. However, we also show that this extrapolation may not be valid in exceptionally geologically complex and data-poor regions (e.g., Madagascar). We suggest research directions to improve the accuracy of global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes, which include: 1) Increasing the collection of bioavailable datasets in data-poor regions; 2) Harmonizing data management practices and metadata collection for bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data; and 3) Relying on advances in remote sensing and geological mapping techniques to improve geological covariates. While significant potential to refine $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes remains, the data products provided in this review form a basis for using $^{87}\text{Sr}/^{86}\text{Sr}$ data in large-scale provenance studies, opening new research avenues in a range of fields.

Keywords: Provenance, Random Forest, Regression, Madagascar, Migration, Isotopes;

1. Introduction

Strontium (Sr) isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) display a unique and predictable patterns of variability on the Earth's surface that follow the geological age and lithology of bedrocks (Bataille and Bowen, 2012). As rocks interact with the hydrosphere, atmosphere and biosphere, bedrock Sr is transferred to other reservoirs on the Earth's surface, such as soils and plants. Geologists have long recognized and capitalized on this natural $^{87}\text{Sr}/^{86}\text{Sr}$ variability to trace the provenance of geological materials (Reviewed in Banner, 2004; Capo et al., 1998b; Peucker-Ehrenbrink and Fiske, 2019). In the last few decades, researchers have also recognized the potential for $^{87}\text{Sr}/^{86}\text{Sr}$ data to solve new questions in ecology, paleoecology, and archeology (Reviewed in Åberg, 1995; Bentley, 2006; Crowley et al., 2017a; Hobson et al., 2010; Makarewicz and Sealy, 2015). This uptick of interest in $^{87}\text{Sr}/^{86}\text{Sr}$ geochemistry has coincided with analytical advances and the development of multi-collector inductively coupled plasma mass spectrometers (MC-ICPMS). This instrumentation and its greater global availability has made $^{87}\text{Sr}/^{86}\text{Sr}$ analysis more mainstream by accelerating throughput and enhancing cost-effectiveness while also facilitating the development of new applications in the life sciences such as laser ablation of incrementally growing tissues (e.g., fin rays and otoliths; Brennan et al., 2015b; Willmes et al., 2016). With these advances, $^{87}\text{Sr}/^{86}\text{Sr}$ geochemistry has become a critical tool for tracing the mobility and/or geographic origin of biological material in ecology (Reviewed in Hobson et al., 2010), paleoecology (Reviewed in Crowley et al., 2017a), archeology (Reviewed in Bentley, 2006), forensic sciences (Reviewed in Makarewicz and Sealy, 2015), and food sciences (Reviewed in Coelho et al., 2017). All of these applications rely on comparing the $^{87}\text{Sr}/^{86}\text{Sr}$ of a given substrate with the isotopic signatures of its potential sources. To facilitate the interpretation of $^{87}\text{Sr}/^{86}\text{Sr}$ data in these applications, it is critical to constrain the spatial variability of $^{87}\text{Sr}/^{86}\text{Sr}$ in the geosphere, hydrosphere and biosphere.

Isoscapes are spatially explicit predictions of isotopic variations. These predictions can be produced either through geostatistical interpolation of observed isotopic data, or through mechanistic model based on first principles of isotope geochemistry (Bowen and West, 2008). Over the last few decades, isoscapes of hydrogen, carbon, oxygen, and nitrogen have been developed, building upon the growing number of isotopic observations (Bowen and Wilkinson, 2002; Still and Powell, 2010; West et al., 2010a). These isoscapes have become a routine tool to understand movement patterns of animals and humans and environmental and biological processes (West et al., 2010b). Isoscape science has recently contributed to research on many high-profile science questions, from partitioning the global hydrological cycle (Good et al., 2015) to assessing the population dynamics of critical species (Brennan et al., 2019). As such, the field of isotope provenancing is rapidly expanding and entering the realm of data science, for example through large initiatives to integrate relevant data in centralized repositories (Pauli et al., 2017), and community efforts to make modeling products widely accessible (Bowen et al., 2014). Historically, interest in, and development of, Sr isoscapes has lagged hydrogen, oxygen or carbon isotopic systems. The primary reasons are that $^{87}\text{Sr}/^{86}\text{Sr}$ analysis is challenging, relatively expensive, and relies on instrumentation that is not as widely available as that needed for conducting light stable isotope analyses. However, $^{87}\text{Sr}/^{86}\text{Sr}$ analyses have progressively emerged as a powerful complementary tool in

provenance studies due to their unique spatial patterns of isotopic variability, with pioneering work having been conducted in archeology (Ezzo et al., 1997; Price et al., 1994; Sillen et al., 1998), paleoecology (Hoppe et al., 1999), ecology (Chamberlain et al., 1997; Kennedy et al., 2002, 2000; Koch et al., 1995b, 1995a; Thorrold and Shuttleworth, 2000), and ecosystem dynamics (Blum et al., 2000; Gosz et al., 1983). In the last decade, the development and application of $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes has grown exponentially, driven by high-profile applications in archeology (e.g., Copeland et al., 2011), paleoecology (e.g., Price et al., 2017), ecology (e.g., Brennan et al., 2019; Glassburn et al., 2018), and forensic science (e.g., Bartelink and Chesson, 2019; Kramer et al., 2020).

This review synthesizes the current state of the rapidly evolving and interdisciplinary research associated with $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes. We begin by reviewing spatial $^{87}\text{Sr}/^{86}\text{Sr}$ trends on the Earth surface with a focus on large-scale patterns derived from the interactions of the geosphere, hydrosphere, atmosphere and biosphere. We then compare different approaches for making $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes in terrestrial and freshwater environments. In an effort to better integrate interdisciplinary $^{87}\text{Sr}/^{86}\text{Sr}$ data, we present the first global compilation of $^{87}\text{Sr}/^{86}\text{Sr}$ data from different environmental substrates. We use this compilation to produce a global model for predicting bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and demonstrate the potential of using this approach to generate $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes at the regional scale in two regions: Europe and Madagascar. We conclude by discussing key knowledge gaps and new research avenues opened by this global data science approach.

2. Strontium isotope cycling

2.1 Strontium isotopes geochemistry

Strontium is a divalent alkaline earth trace element with four naturally occurring isotopes: ^{84}Sr (~0.56%), ^{86}Sr (~9.87%), ^{87}Sr (~7.04%) and ^{88}Sr (~82.53%). ^{84}Sr , ^{86}Sr , ^{87}Sr and ^{88}Sr are all stable isotopes (i.e., do not radioactively decay). Unlike the other Sr isotopes, ^{87}Sr is the radiogenic daughter product of rubidium 87 (^{87}Rb ; decay constant $\lambda = 1.42 \times 10^{-11} \text{ year}^{-1}$; Steiger and Jäger, 1977). The ratio of ^{87}Sr to the other isotopes is therefore a function of the variable abundance of ^{87}Sr . In provenance studies, Sr isotope variations are typically represented using the ratio of ^{87}Sr relative to ^{86}Sr after correction for any mass-dependent fractionation by normalization to a fixed $^{86}\text{Sr}/^{88}\text{Sr}$ (0.1194; Nier, 1938). The resulting $^{87}\text{Sr}/^{86}\text{Sr}$ is thus not a function of isotopic fractionation processes but only reflects the mixing of isotopically distinct Sr sources.

2.2 Strontium isotopes in the geosphere

The $^{87}\text{Sr}/^{86}\text{Sr}$ in modern rocks and minerals is both mineral-dependent (initial ^{87}Rb , ^{87}Sr , and ^{86}Sr abundance), and time-dependent (radioactive decay of ^{87}Rb to ^{87}Sr). At the time of our planet's formation, the bulk Earth reservoir had a relatively homogeneous $^{87}\text{Sr}/^{86}\text{Sr}$ signature of around 0.699 (Wetherill et al., 1973). As geochemical differentiation progressed, Sr and Rb concentrated in melts that preferentially contributed to oceanic and continental crusts. This partitioning resulted in increased $^{87}\text{Rb}/^{86}\text{Sr}$ in the continental crust relative to the oceanic crust and the residual mantle (Faure and Powell, 1972), and over time, this led to differences in $^{87}\text{Sr}/^{86}\text{Sr}$ among geologic pools with the progressive decay of ^{87}Rb into ^{87}Sr .

These combined effects of geochemical partitioning of Rb and Sr and radioactive decay explain the large range of $^{87}\text{Sr}/^{86}\text{Sr}$ in igneous, sedimentary, and metamorphic rocks. With equal initial $^{87}\text{Rb}/^{86}\text{Sr}$, older igneous rocks have higher $^{87}\text{Sr}/^{86}\text{Sr}$ than younger rocks because ^{87}Rb has had more time to decay in the older reservoir. At equal age, more felsic

rocks (with higher $^{87}\text{Rb}/^{86}\text{Sr}$) have higher $^{87}\text{Sr}/^{86}\text{Sr}$ than mafic rocks (with lower $^{87}\text{Rb}/^{86}\text{Sr}$) because more ^{87}Rb is available to decay into ^{87}Sr . Old felsic igneous rock units (e.g., cratonic shields) have the highest measured $^{87}\text{Sr}/^{86}\text{Sr}$ (>0.720), while newly formed mafic igneous rock units (e.g., basalts, volcanic arcs) have the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ (~ 0.703) (Peucker-Ehrenbrink and Fiske, 2019).

Siliciclastic sediments inherit $^{87}\text{Sr}/^{86}\text{Sr}$ from their parent rocks but are usually composed of a mixture of minerals with distinct parent rock, and thus different isotopic ratios. Because local bedrock sources dominate the $^{87}\text{Sr}/^{86}\text{Sr}$, recently deposited siliciclastic sediments from young igneous rocks (e.g., volcanic arcs) tend to have lower $^{87}\text{Sr}/^{86}\text{Sr}$ than those forming in older felsic environments (Bataille et al., 2014). However, it becomes challenging to assess the original parent rock for older sediments due to tectonic and geomorphological evolution of the surface. As sediments are exposed and reworked on the surface, the preferential removal of Sr relative to Rb in weathering and metamorphic processes can subsequently modify their $^{87}\text{Sr}/^{86}\text{Sr}$ leading to additional $^{87}\text{Sr}/^{86}\text{Sr}$ variability in older sedimentary units (Bataille et al., 2014). Carbonate rocks, on the other hand, primarily inherit their $^{87}\text{Sr}/^{86}\text{Sr}$ from seawater (the exceptions being metamorphic, igneous or soil carbonates). They have a narrow range in $^{87}\text{Sr}/^{86}\text{Sr}$ because 1) seawater $^{87}\text{Sr}/^{86}\text{Sr}$ has remained within a tight range throughout the Phanerozoic (0.707 and 0.709; McArthur et al., 2001), and 2) carbonates have small amounts of Rb but large amounts of Sr (Sr readily substitutes for calcium), which means their $^{87}\text{Sr}/^{86}\text{Sr}$ does not evolve significantly through time.

Metamorphism can alter the $^{87}\text{Sr}/^{86}\text{Sr}$ of igneous and sedimentary rock units, which can lead to highly variable $^{87}\text{Sr}/^{86}\text{Sr}$ in metamorphic rocks. For example, metalimestones in the Himalayas have much higher $^{87}\text{Sr}/^{86}\text{Sr}$ than non-metamorphosed marine carbonates due to exchange of ^{87}Sr between silicates and carbonates during metamorphism (Bickle et al., 2001). Similarly, hydrothermal metamorphism can homogenize the $^{87}\text{Sr}/^{86}\text{Sr}$ of lithologically distinct sedimentary rock units (Bickle et al., 1988). Ultimately, the combination of igneous, sedimentary and metamorphic processes leads to considerable variability of $^{87}\text{Sr}/^{86}\text{Sr}$ in the geosphere. Bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ ranges from 0.702 in ophiolites to more than 1 in old felsic Archean rocks (Faure and Powell, 1972). The $^{87}\text{Sr}/^{86}\text{Sr}$ distribution in rocks is positively skewed with average upper crust $^{87}\text{Sr}/^{86}\text{Sr}$ around 0.716.

2.3 Strontium isotopes in soils and plants

Soils primarily inherit their $^{87}\text{Sr}/^{86}\text{Sr}$ composition from parent rock, and consequently $^{87}\text{Sr}/^{86}\text{Sr}$ patterns in soils follow those of the underlying geology (Fig. 1). However, the observed relationship between soil and local bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ is rarely 1:1. This is because soil $^{87}\text{Sr}/^{86}\text{Sr}$ is buffered by the influence of additional sources of Sr with distinct $^{87}\text{Sr}/^{86}\text{Sr}$ compositions (Fig. 1). For example, unconsolidated sediments, such as fluvial terraces and glacial till or loess, typically have different $^{87}\text{Sr}/^{86}\text{Sr}$ than the underlying bedrock (Börker et al., 2018). In the Midwestern USA, glacial till coming from the Canadian craton covers much of the bedrock and is the dominant source of Sr to soils (Hedman et al., 2009; Widga et al., 2017). Downwind of desert zones, dust often recharges soil primary minerals and can even serve as the dominant parent material in some cases (Aarons et al., 2017; Frumkin and Stein, 2004; Grousset et al., 1992; Miller et al., 2014; Naiman et al., 2000; Widory et al., 2010). Tephra deposition near volcanic centers can also provide easily weatherable and Sr-rich primary minerals to soils and ecosystems that will dominate over bedrock sources (Chadwick et al., 2009). Last but not least, near the coast, sea salt aerosol deposition can influence the exchangeable Sr fraction of soil (Hartman and Richards, 2014; Quade et al., 1995; Whipkey et al., 2000).

Different soil fractions can also have distinct $^{87}\text{Sr}/^{86}\text{Sr}$ compositions due to differential weathering and soil mixing processes (Fig. 1). In particular, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the soil exchangeable Sr fraction (defined as the fraction that is extractable using ammonium nitrate leaching from a top soil sample) can differ considerably from the bulk digested soil mineral fraction or from soil samples collected at different depths (Gryschko et al., 2005; Poszwa et al., 2004). The soil exchangeable Sr corresponds primarily to Sr dissolved in soil water and available to plants but also includes some Sr weakly absorbed on clays (Capo et al., 1998). The $^{87}\text{Sr}/^{86}\text{Sr}$ of this soil exchangeable Sr is strongly influenced by soil age. In a young soil, Sr-rich and rapidly weathering primary minerals (e.g., carbonates, plagioclases) will be the dominant contributors to the exchangeable Sr pool (Chadwick et al., 2009; Vitousek et al., 1999). However, as a soil matures, these rapidly weathering minerals can be exhausted and other primary minerals with distinct $^{87}\text{Sr}/^{86}\text{Sr}$ will dominate the Sr budget. As soils become more and more weathered, fewer primary minerals are available, and other sources (e.g., dust and other aerosols) increasingly contribute to the exchangeable Sr budget (Chadwick et al., 2009; Vitousek et al., 1999). The contribution of these atmospheric sources to the dissolved soil $^{87}\text{Sr}/^{86}\text{Sr}$ budget will depend in the rate of deposition, weatherability, Sr content, and $^{87}\text{Sr}/^{86}\text{Sr}$ (Pett-Ridge et al., 2009). In some cases, atmospheric deposition will dominate over bedrock sources even when soils are young (Miller et al., 2014), whereas in other cases, bedrock remains the dominant Sr source even when soils are highly weathered (Bern et al., 2005; Porder et al., 2006).

Plants take up Sr from the exchangeable soil fraction (Fig. 1). However, differences between $^{87}\text{Sr}/^{86}\text{Sr}$ in plants and the exchangeable Sr fraction can occur, particularly in regions where multiple sources of Sr mix into soils (e.g., atmospheric vs. parent rock Sr; Hartman and Richards, 2014; Laffoon et al., 2012; Snoeck et al., 2016). As mentioned above, differential weathering and soil mixing processes can lead to variable $^{87}\text{Sr}/^{86}\text{Sr}$ along the soil profile (Reynolds et al., 2012). This variability is propagated among plants with different rooting depth (Poszwa et al., 2004).

2.4 Strontium isotopes in surface waters

Water inherits $^{87}\text{Sr}/^{86}\text{Sr}$ from rock weathering (Fig. 1). Consequently, spatial patterning of $^{87}\text{Sr}/^{86}\text{Sr}$ in the hydrosphere reflects that of rocks exposed on the surface and in aquifers, with high $^{87}\text{Sr}/^{86}\text{Sr}$ in rivers draining cratons and low $^{87}\text{Sr}/^{86}\text{Sr}$ in rivers draining mafic rock units (Peucker-Ehrenbrink and Fiske, 2019). However, the contributions of different minerals and rock units to the dissolved Sr pool vary broadly based on their weathering rate and Sr content, which in turn leads to distinct $^{87}\text{Sr}/^{86}\text{Sr}$ between the hydrosphere and geosphere (Blum et al., 1993). During igneous rock weathering, plagioclases contribute more than radiogenic minerals to the dissolved Sr flux due to their higher Sr content and weathering rate (Bain and Bacon, 1994; Clow et al., 1997; Pett-Ridge et al., 2009). Similarly Sr-rich and easily weatherable carbonates and evaporites contribute disproportionately to the dissolved Sr in the hydrosphere (Peucker-Ehrenbrink and Fiske, 2019). Even trace amounts of calcite in silicates can dominate the entire Sr budget in large rivers (Clow et al., 1997). At the scale of a catchment, the flux of Sr from isotopically distinct rock units is also modulated by geomorphological, climatic and environmental conditions (Bataille et al., 2014). For example, sediments from mountainous zones will contribute disproportionately to the total Sr flux and strongly influence the $^{87}\text{Sr}/^{86}\text{Sr}$ as they weathered along the entire course of a river (Galy and France-Lanord, 1999) and its floodplains (Bickle et al., 2018; Lupker et al., 2012).

Allochthonous surface sediments, such as glacial till, may also impact river $^{87}\text{Sr}/^{86}\text{Sr}$ (Curtis and Stueber, 1973). In boreal and arctic regions, seasonal land cover changes (i.e., vegetation, ice cover) can lead to strong seasonal $^{87}\text{Sr}/^{86}\text{Sr}$ fluctuations because of variable contribution of rock units within a catchment (Douglas et al., 2013; Voss et al., 2014). Ultimately, spatial $^{87}\text{Sr}/^{86}\text{Sr}$ trends in the hydrosphere follow those of the geosphere but with lower ratios and variability due to the buffering effect of carbonate weathering (Palmer and Edmond, 1992).

2.5 Strontium isotopes in animal tissues

Primary terrestrial consumers obtain the majority of their Sr from diet (Fig. 1; Gloirennec et al., 2016). As plants are at the base of many terrestrial food chains, $^{87}\text{Sr}/^{86}\text{Sr}$ for animals usually reflects that of local plants (Willmes et al., 2014). However, drinking water can contribute significantly to the Sr inputs when (1) water is Sr-rich (e.g., carbonate landscapes), or (2) animals drink frequently (Lewis et al., 2017). Different taxa sample Sr differently on the landscape depending on their feeding habits and feeding ranges (Lengfelder et al., 2019). For example, at a given location, small herbivores with a local feeding range (e.g., rodents) often have distinct $^{87}\text{Sr}/^{86}\text{Sr}$ signatures from larger herbivores (e.g., deer) because the larger animals integrate Sr sources over a larger area (Feranec et al., 2007; Lengfelder et al., 2019). Aquatic animals (e.g., tapirs) may also have distinct $^{87}\text{Sr}/^{86}\text{Sr}$ if they feed on riparian or freshwater foods (e.g., Hedman et al., 2009; Wallace et al., 2019). There are more challenges associated with predicting how different environmental sources contribute to consumer Sr with an increase in the complexity of animal feeding behaviors (e.g., large migratory herbivores, omnivores and carnivores) (Hoppe et al., 1999). At the extreme, humans can eat local terrestrial resources, hunt migratory mammals, harvest marine resources, inheriting a potentially very complex mixture of $^{87}\text{Sr}/^{86}\text{Sr}$ sources (Fig. 1; Bentley, 2006). Humans can also obtain resources from distant localities via trade (reviewed in Bentley, 2006).

Sr is integrated over variable timescales in different animal tissues (Makarewicz and Sealy, 2015). Incrementally growing tissues, such as tusks, otoliths or hair segments, sequentially record dietary Sr, allowing high-resolution reconstruction of mobility histories of the sampled individuals (e.g., Brennan et al., 2015a; Vautour et al., 2015). Other tissues, such as bulk (whole) tooth samples or bones, preserve a snapshot of a specific period in the organism's life, or integrate Sr over longer time periods, respectively, and can therefore provide information about dietary signatures at different stages of an individual's life (reviewed in Makarewicz and Sealy, 2015).

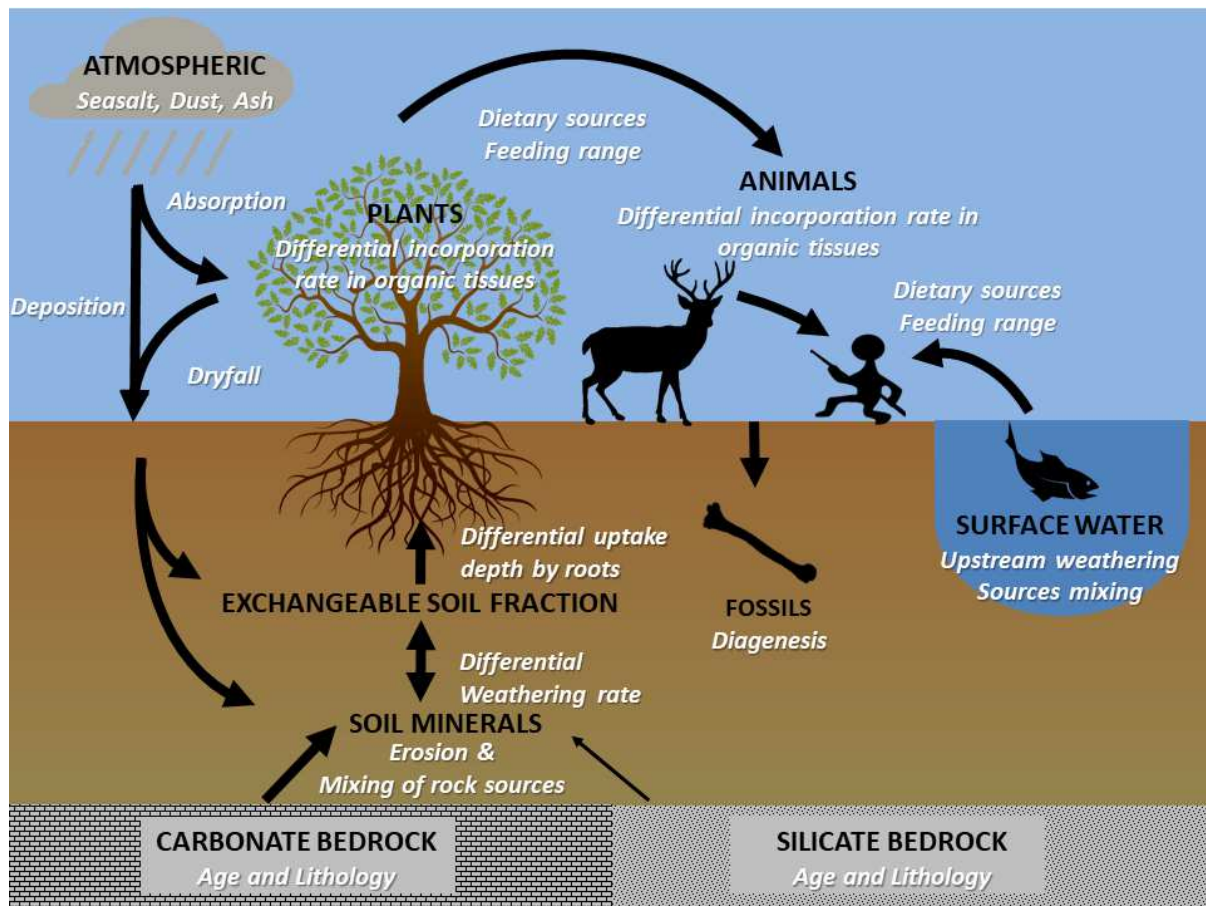


Figure 1: Simplified schematic of strontium isotope cycling from rocks to ecosystems. Capitalized black words correspond to Sr reservoirs and italicized white words correspond to process modifying $^{87}\text{Sr}/^{86}\text{Sr}$.

3. Strontium isoscapes

3.1 Empirical isoscapes

The most commonly used strategy to create biologically available (bioavailable) $^{87}\text{Sr}/^{86}\text{Sr}$ baselines is to analyze $^{87}\text{Sr}/^{86}\text{Sr}$ in a series of biological samples representing bioavailable Sr pools in a study area (Lengfelder et al., 2019). However, as outlined above, different substrates (e.g., soils, plants, animals, or waters) can integrate $^{87}\text{Sr}/^{86}\text{Sr}$ at different spatiotemporal scales depending on the local geo-environmental conditions. Thus, identifying appropriate substrates can be challenging. We review the pros and cons of each substrate type below.

Modern plants and exchangeable Sr from top soils are the most commonly used substrate to represent local bioavailable Sr because they are easily collected. However, they may sample isotopically discrete pools depending on plant rooting depths (Poszwa et al., 2004). It is also increasingly recognized that application of agricultural fertilizers and limes impacts the natural bioavailable Sr pool (Frei et al., 2020; Maurer et al., 2012; Thomsen and Andreasen, 2019). The extent and frequency of agricultural contamination in baseline samples remains unknown but could seriously complicate the development of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ baselines in regions with active or historical agriculture.

Animals with relatively small foraging ranges, such as rodents or deer, are also used to represent the local bioavailable Sr pool (e.g., Burton and Price, 2013). Although animals

integrate Sr over variable spatiotemporal scales, adding some uncertainty in the interpretation, they also offer the advantage of integrating local bioavailable Sr from multiple sources. Isoscapes derived from local animals might therefore be more suitable than those derived from plants or soils to represent the bioavailable Sr sampled by large mammals, including humans (Crowley et al., 2017a). Fortunately, in many cases, the $^{87}\text{Sr}/^{86}\text{Sr}$ of local animals are similar to those of plants and soils, indicating that, at local scales, bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ signatures are relatively invariant (Flockhart et al., 2015).

Water samples taken from small streams, lakes and rivers have also been used to represent bioavailable Sr pools. As otoliths inherit their $^{87}\text{Sr}/^{86}\text{Sr}$ from the river or lake water in which fish live (Brennan et al., 2015a; Faure et al., 1967), water samples are particularly relevant in freshwater ecology applications (e.g., Brennan et al., 2019). In terrestrial applications, however, water can integrate Sr from larger areas than plants or local animals (Frei and Frei, 2011). Although this spatial integration might help solve some of the lime and fertilizer contamination issues found with other substrates (Frei et al., 2020), it can also complicate the comparison with biological samples. Nevertheless, water samples can provide a good baseline for the $^{87}\text{Sr}/^{86}\text{Sr}$ of terrestrial species exploiting aquatic or riparian habitats (Hamilton et al., 2019).

Once collected, bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data are usually interpolated using geostatistical algorithms (e.g., kriging; Copeland et al., 2016; Frei and Frei, 2011; Willmes et al., 2018). Geostatistical algorithms are based on the idea that points closer to each other tend to resemble each other (i.e., spatial autocorrelation). These geostatistical models can also integrate covariates (e.g., geological maps) to further constrain $^{87}\text{Sr}/^{86}\text{Sr}$ patterns (Willmes et al., 2018). Maps derived from geostatistical methods, however, can give ambiguous and inaccurate results due to the challenges of incorporating non-normal and skewed distributions of $^{87}\text{Sr}/^{86}\text{Sr}$ data (Bataille et al., 2018). Another method widely used to produce $^{87}\text{Sr}/^{86}\text{Sr}$ maps is to average $^{87}\text{Sr}/^{86}\text{Sr}$ data over regions with similar bedrock geology (e.g., Evans et al., 2010; Voerkelius et al., 2010). However, this leads to overlap and lack of precision in constructed isoscapes, which limits their usefulness for provenance studies. These limitations can be overcome by increasing the sampling density, but this becomes prohibitively expensive at regional to continental scales.

3.2 Mechanistic isoscapes

A second approach to developing $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes relies on mechanistic models. This approach leverages geochemical knowledge to predict the evolution of $^{87}\text{Sr}/^{86}\text{Sr}$ in rocks using the radiogenic equation:

$$\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{rock}} = \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_i + \left(\frac{^{87}\text{Rb}}{^{86}\text{Sr}}\right)_{\text{rock}} (e^{\lambda t} - 1), \quad (1)$$

where $^{87}\text{Sr}/^{86}\text{Sr}$ variability in rocks $(^{87}\text{Sr}/^{86}\text{Sr})_{\text{rock}}$ is a function of: 1) The initial $^{87}\text{Sr}/^{86}\text{Sr}$ $[(^{87}\text{Sr}/^{86}\text{Sr})_i]$, which depends on the geological history of the parent rock; 2) Bedrock age (t), which controls the fraction of ^{87}Rb that decayed into ^{87}Sr ; and 3) The $^{87}\text{Rb}/^{86}\text{Sr}$ of the rock $[(^{87}\text{Rb}/^{86}\text{Sr})_{\text{rock}}]$, which varies with lithology.

To simplify the equation, it was first assumed that $^{87}\text{Sr}/^{86}\text{Sr}$ variability in rocks is only a function of rock age (Beard and Johnson, 2000). Using geological maps to estimate the age of bedrock units, these authors predicted $^{87}\text{Sr}/^{86}\text{Sr}$ for bedrock across the USA. Later, Bataille and Bowen (2012) revisited this approach by accounting for distinct $^{87}\text{Rb}/^{86}\text{Sr}$ among rock types. They separated rocks into major lithological categories, specifically distinguishing

between carbonates and silicates, and leveraged geochemical databases to calculate the average $^{87}\text{Rb}/^{86}\text{Sr}$ for each group. These authors also predicted $^{87}\text{Sr}/^{86}\text{Sr}$ variability in rivers across the USA using a first-principles model of chemical weathering to propagate $^{87}\text{Sr}/^{86}\text{Sr}$ from rock to water. This modeling approach yielded notable improvement over the approach used by Beard and Johnson (2000), but the accuracy of both the rock and water isoscapes were not sufficient for most provenance applications.

Using a case study in Alaska, Bataille et al. (2014) improved this mechanistic model by incorporating more detailed lithological categories, including a sub-model for siliciclastic sedimentary rocks. They also integrated an empirically-calibrated chemical weathering model. The resulting model performed well against bedrock and water data. However, the averaging approach, uncertainty of geological maps, and difficulty in calibrating certain rock categories remained major obstacles to the development of $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes sufficiently accurate for provenance applications.

As mentioned previously, a multitude of coupled processes contribute to mix isotopically distinct Sr sources from rocks to ecosystems. Properly calibrating the interactions of those geological, environmental or biological processes to predict $^{87}\text{Sr}/^{86}\text{Sr}$ in the terrestrial biosphere is a substantial challenge. Bataille et al. (2012) attempted to calibrate a mechanistic bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ model in the circum-Caribbean region. They demonstrated that bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ predictions diverged from bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data in this region (Laffoon et al., 2012) and attempted to calibrate a multi-source model that accounted for the contribution of atmospheric sources. While the model performed well on the existing bioavailable data for the Antilles (Laffoon et al., 2012), additional sampling showed that the contribution of atmospheric deposition was not accurately calibrated in the Bahamas (Schulting et al., 2018).

Developing accurate mechanistic bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ models requires a coupled parameterization of a multitude of complex processes. Integrating Sr isotope cycling in global land surface models might be a productive avenue to advance the mechanistic approach. However, to date, the lack of accurate, detailed global geological maps remains the major hurdle to more accurate mechanistic $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes.

3.3 Process-based statistical isoscapes

A third, intermediate approach has been proposed to overcome the limitations of both mechanistic and geostatistical modeling. This approach relies on multivariate random forest regression, a tree based machine-learning algorithm, to integrate empirical data and other geo-environmental covariates into the modeling framework. Bataille et al. (2018) first used random forest regression to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ across Western Europe. This study integrated a compilation of empirical bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data, bedrock model products, and geo-environmental covariates to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. The resulting model yielded greatly improved accuracy over mechanistic models while also overcoming the issues encountered with traditional geostatistical methods. More recently, Hoogewerff et al., (2019) used random forest regression to predict $^{87}\text{Sr}/^{86}\text{Sr}$ for agricultural top soils across Europe using previously collected geochemical soil data and newly generated $^{87}\text{Sr}/^{86}\text{Sr}$ data from selected soils of the GEMAS project (i.e., Geochemical Mapping of Agricultural and Grazing Land Soil). This method yielded excellent results, predicting bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ with both high precision and accuracy. However, the model was limited by the availability of high-resolution geochemical soil surveys.

In freshwater environments, Brennan et al. (2016) proposed using spatial stream network models to predict $^{87}\text{Sr}/^{86}\text{Sr}$ in river water. This statistical approach accounts for the unique spatial correlation structure in rivers (e.g., flow direction, branching) while also providing a quantitative framework for assessing the contribution of in-stream processes (e.g., downstream transport) and landscape processes (e.g., climate, geology) on observed $^{87}\text{Sr}/^{86}\text{Sr}$ patterns. The application of this method was a break-through in freshwater $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes, improving accuracy by an order of magnitude over both mechanistic (Bataille et al., 2014) and geostatistical models (Hegg et al., 2013).

4. Building process-based global bioavailable strontium isoscape

4.1 Global compilation

4.1.1 Compilation description

To date, no researchers have attempted global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ modeling. Here we leverage a new global data compilation, mechanistic models, and auxiliary variables integrated in a multivariate random forest regression framework to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ at the global scale. The dataset used in our study is a compilation of 17,240 published and unpublished $^{87}\text{Sr}/^{86}\text{Sr}$ analyses from 278 individual studies spanning 8,476 individual locations across the globe (Fig. 2). Unpublished data include 279 bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ from Madagascar (see supplementary material Appendix B) and 70 surface waters from Cook Inlet Alaska (see supplementary material Appendix C). The compiled database is available in supplementary material Table S1 (along with the full citations of the original sources from which the data sets were derived), and is being integrated in the IsoBank repository (Pauli et al., 2017). The data can be filtered by country, sample type (several levels), or analytical substrate. Local data come from 3,249 plants, 2,598 soils, 2,335 local animals (bone, dentine, enamel, snail shell), and 4,813 surface waters. The remaining samples include migratory or highly mobile animals, humans, and dust that could be useful in other applications. The database includes latitude and longitude data for each individual sample. In the majority of cases, geographic coordinates were reported by the authors in the publication. When these were not included, we used Google Earth to georeference reported geographic information (e.g., maps or locality names). When necessary, authors of publications were contacted to clarify locality information. The method of georeferencing and its associated uncertainty are reported in the database (Table S1).

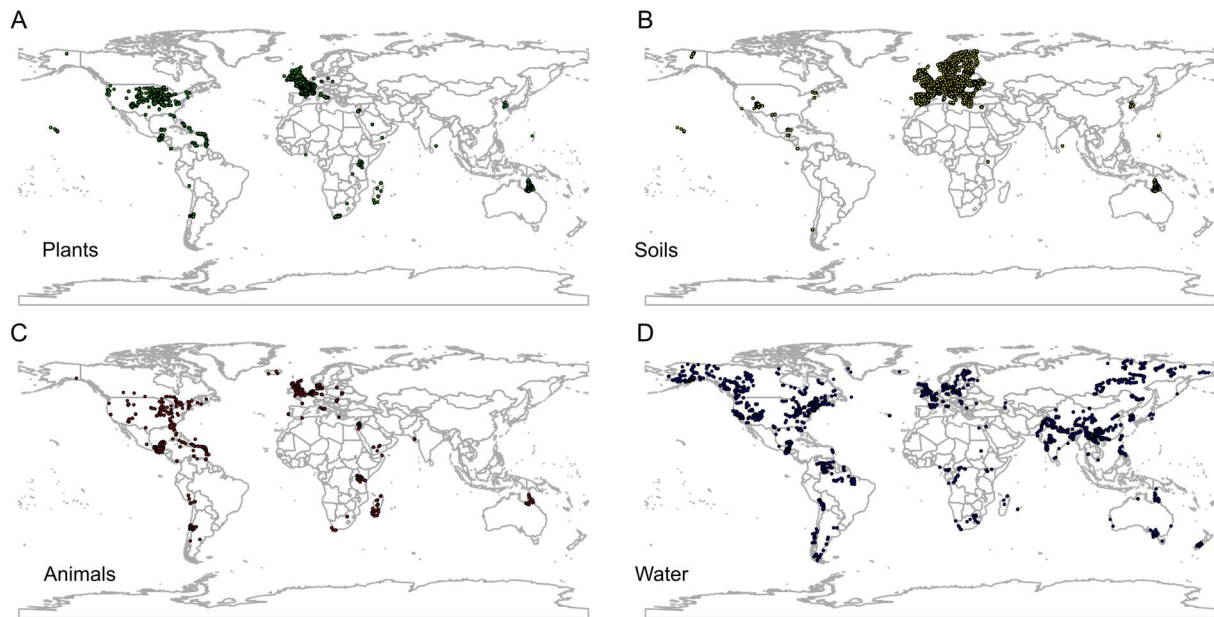


Figure 2: Global compilation of $^{87}\text{Sr}/^{86}\text{Sr}$ data by substrate. A) Plants, B) Soils, C) Animals and D) Water.

4.1.2 Geographic biases and sample distribution

The sampling density, and geographic distribution of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data vary tremendously across the globe. Most samples are from regions with a high archeological interest (e.g., Mesoamerica) or strong research funding programs (e.g., Europe, USA). Plant and local animal samples are concentrated in Europe, the USA and the circum-Caribbean region. Soil samples are highly concentrated in Europe with close to half coming from the published GEMAS dataset (Hoogewerff et al., 2019). Water samples are more broadly distributed and include most large rivers of the world; this reflects the 50 years of hydrological research using $^{87}\text{Sr}/^{86}\text{Sr}$ geochemistry. The sampling strategy also varied among studies. In some, samples from different substrates were collected opportunistically depending on research funds and accessibility (e.g., Laffoon et al., 2012), while in others, samples from one substrate were collected systematically on a regular grid (e.g., Hoogewerff et al., 2019). Additionally, even though the global compilation is extensive and covers areas representing different climatic zones, many regions remain strongly under-sampled due to restricted access, extreme climate conditions, or political reasons. These geographic biases will propagate in models calibrated using this global compilation (e.g., regions with low sample density will be poorly calibrated relative to regions with high sample density).

4.1.3 Descriptive statistics

Plants, soils and local animals display similar distributions of $^{87}\text{Sr}/^{86}\text{Sr}$, with Quartile 1 = 0.7084 ± 0.0001 , median = 0.7095 ± 0.0003 , and Quartile 3 = 0.711 ± 0.0004 (Fig. 3). More than 90% of the samples have $^{87}\text{Sr}/^{86}\text{Sr}$ that falls within a tight range of 0.706 ± 0.001 to 0.720 ± 0.001 (Fig. 3). Variability in water is slightly larger with Quartile 1 = 0.7089 , median = 0.7110 , Quartile 3 = 0.7155 , and a 90% interval range from 0.706 to 0.7396 . This difference in range between water and other substrates is likely due to spatial sampling biases (Fig. 2). Many of the water samples compiled in the database are from older radiogenic geological regions including the Himalayas, Canadian craton, Scandinavian cratons and Russian cratons (Fig. 2). This geographic distribution contrasts with that of plants, soils and

local animals that were mostly sampled from flatter areas, agricultural regions and younger geological regions.

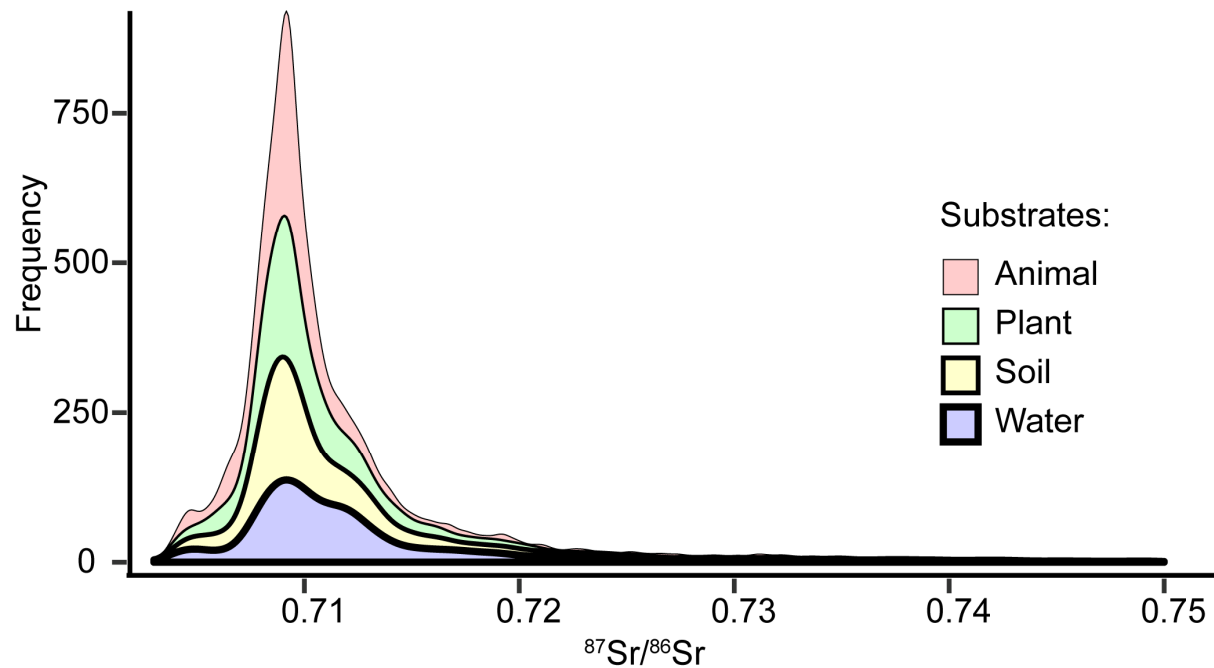


Figure 3: Distribution of $^{87}\text{Sr}/^{86}\text{Sr}$ variability by substrate (stacked). The x-axis is cut at 0.750 to facilitate visualization but more some water samples fall beyond that range.

4.1.4 Intra-site variability

We explored intra-site variability in bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ by calculating the standard deviation of $^{87}\text{Sr}/^{86}\text{Sr}$ among samples at each location where more than one sample/substrate was collected. This intra-site variability is an important measure as it represents the maximum accuracy a model could reach. As noted in previous studies (Bataille et al., 2018; Hoogewerff et al., 2019), there is a clear trend of increasing variance with increasing $^{87}\text{Sr}/^{86}\text{Sr}$ for plants, soils and animals (Fig. 4). Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is most variable for sites dominated by very old felsic rock units (e.g., cratons) with ratios >0.710 . The minimum variance is found between 0.707 and 0.709, corresponding to $^{87}\text{Sr}/^{86}\text{Sr}$ of carbonate units (Fig. 4). Sites with $^{87}\text{Sr}/^{86}\text{Sr} < 0.707$ (typical of younger and/or mafic geologies) have slightly higher variance than sites dominated by carbonates. Combining plants, soils and local animals, the Root Mean Squared Error (RMSE) of intra-site variability is 0.001. However, this RMSE is lower than 0.0005 when using only bioavailable $^{87}\text{Sr}/^{86}\text{Sr} < 0.710$.

The positive correlation between predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and RMSE is consistent with the lognormal distribution of $^{87}\text{Sr}/^{86}\text{Sr}$ in the geosphere. As the age of a rock unit increases, the different minerals composing this unit have increasingly divergent $^{87}\text{Sr}/^{86}\text{Sr}$. Differential weathering of those minerals can considerably increase variability in $^{87}\text{Sr}/^{86}\text{Sr}$ for soils at radiogenic sites even at the very local scale (Clow et al., 1997). In addition, felsic rocks tend to weather slowly, which means there may be a higher relative contribution of bioavailable Sr from exogenous sources (e.g., dust) (Hoogewerff et al., 2019).

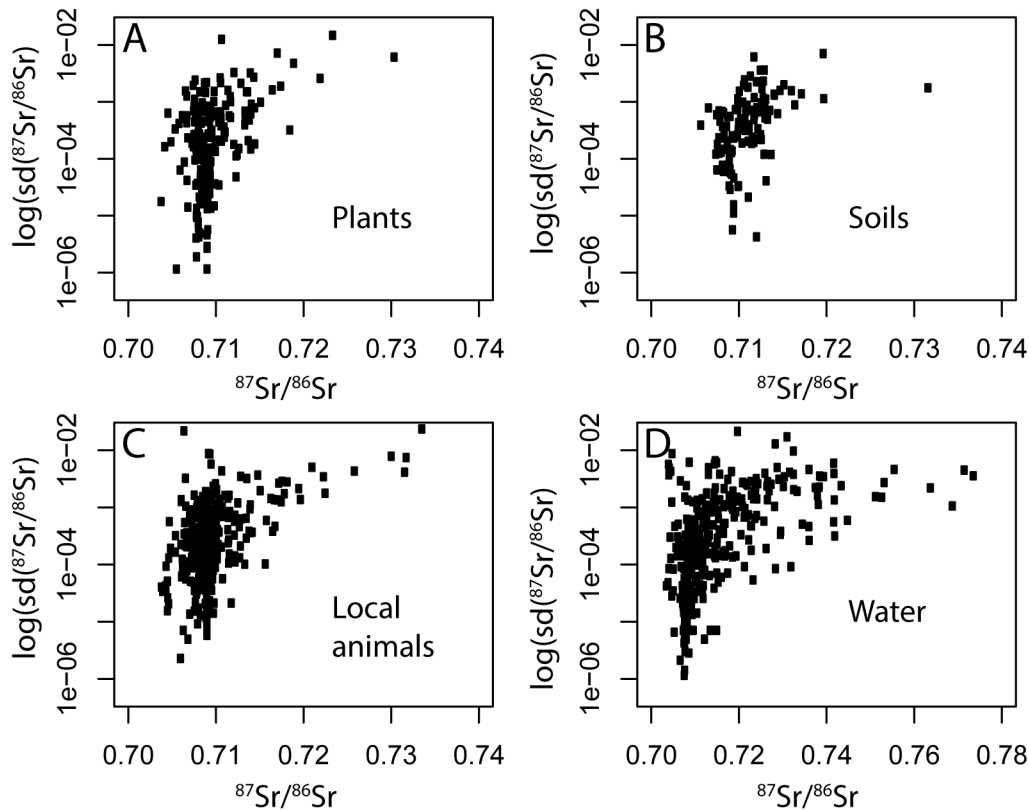


Figure 4. Within-site $^{87}\text{Sr}/^{86}\text{Sr}$ variability by substrate. A) Plants, B) Soils, C) Local animals and D) Water.

4.2 Mechanistic bedrock model

We applied the published model formulation of Bataille et al. (2014) at a global scale using the GLiM geological map as the primary source for geological information. (Fig. 5). This bedrock model predicts the median, Quartile 1 and Quartile 3 of $^{87}\text{Sr}/^{86}\text{Sr}$ for bedrock at present-day. As the GLiM geodatabase does not contain numerical age information (Hartmann and Moosdorf, 2012), we converted age descriptor to a range of numerical values using the geological timescale (Walker et al., 2013; see supplementary material Table S2). For more information on the exact parameterization steps of the bedrock model, refer to the supplementary material in Bataille et al. (2014), including Tables A1 and A3. Predicted bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ follows the expected trends, with higher $^{87}\text{Sr}/^{86}\text{Sr}$ in older cratonic settings and lower $^{87}\text{Sr}/^{86}\text{Sr}$ in arc settings (Fig. 5). However, the bedrock model predictions also reflect the limitations of the GLiM geodatabase. The GLiM was constructed by compiling geological maps at different resolutions with an "average" resolution of 1:3 750 000. It divides lithologies into 16 major classes, separating mafic, intermediate and felsic plutonic and volcanic rocks, metamorphic rocks, carbonates, evaporites, silicates, and mixed carbonate/silicate sediments. Of particular relevance for predicting $^{87}\text{Sr}/^{86}\text{Sr}$ in rocks, secondary lithological descriptors also report the presence of carbonates, evaporites and loess. However, reporting of secondary lithologies is inconsistent among countries (and frequently absent). Similarly, the definition of bedrock can differ between countries, with some surveys mapping surficial sediments (e.g., glacial till) as bedrock whereas other reserve bedrock mapping to the solid rock unit underlying unconsolidated sediments. Consequently, irregularities in predicted $^{87}\text{Sr}/^{86}\text{Sr}$, reflecting different mapping methods, resolution and

accuracy are often observed at political borders (e.g., Canada, USA). These inaccuracies cannot be resolved until high resolution harmonized geological maps are produced at the global scale. Another major source of uncertainty in this bedrock model is that the bedrock ages (Table S2) mean different things for different lithologies (e.g., crystallization age for intrusive rocks, eruptive ages for volcanic rocks, last higher-grade metamorphic overprint of metamorphic rocks, and depositional age for sedimentary rocks). Consequently, some lithologies (i.e., metamorphic, sedimentary), might contain significantly older minerals than the overall age of the geologic unit (e.g., a recently deposited sediment might have zircon grains that are billion years old). This can lead to large uncertainties in predicted bedrock $^{87}\text{Sr}/^{86}\text{Sr}$.

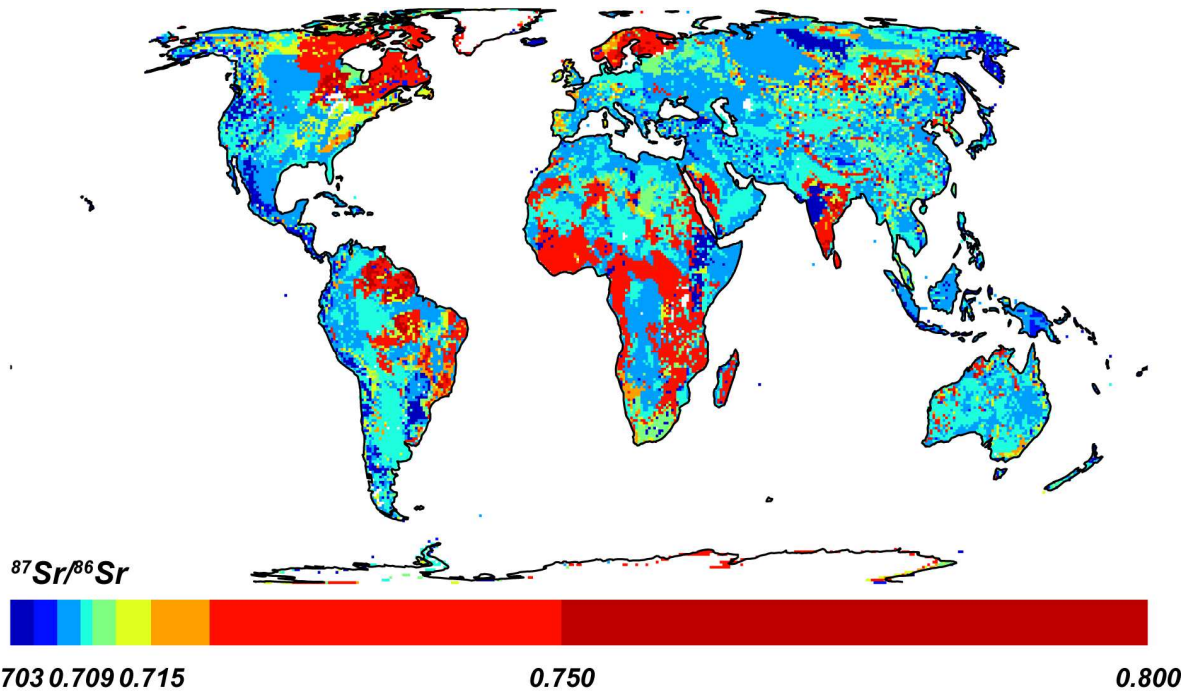


Figure 5: Global map of predicted $^{87}\text{Sr}/^{86}\text{Sr}$ in bedrock following the formulation of Bataille et al. (2014)

4.3 Other auxiliary data.

We assembled data on selected covariates that represent the main factors that impact variability in bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$: Bedrock ages (see section 4.2), terrane age, surficial geology type, soil properties, aerosol deposition, relief, climate, and agricultural activity (Table 1). This series of covariates expands on Bataille et al. (2018) by including global nitrogen and phosphorus fertilization (Potter et al., 2010), surficial deposits (Börker et al., 2018), global mean annual temperature (Hijmans et al., 2005), and an updated raster of global sea salt aerosol deposition (Vet et al., 2014).

Table 1: List of geological, climatic, environmental and anthropogenic variables used in the multivariate regression. D = Discrete; C = Continuous; GLiM = Global Lithological Map; CCSM.3 = Community Climate System Model 3; SRTM = Shuttle Radar Topography Mission

Variables	Description	Resolution	Type	Source
r.maxage_geol	GLiM age attribute	1 km	D	(Hartmann and Moosdorf,

	(Myrs)			2012)
r.minage_geol	GLiM age attribute (Myrs)	1 km	D	(Hartmann and Moosdorf, 2012)
r.meanage_geol	GLiM age attribute (Myrs)	1 km	D	(Hartmann and Moosdorf, 2012)
r.age	Terrane age attribute (Myrs)	1 km	D	(Mooney et al., 1998)
r.GUM	Global unconsolidated sediment map	1 km	C	(Börker et al., 2018)
r.ssaw	Multi-models average sea salt wet deposition (kg.ha ⁻¹ .yr ⁻¹)	1°×1°	C	(Vet et al., 2014)
r.ssa	Multi-models average Sea salt wet+dry deposition (kg.ha ⁻¹ .yr ⁻¹)	1°×1°	C	(Vet et al., 2014)
r.dust	Multi-models average (g.m ⁻² .yr ⁻¹)	1°×1°	C	(Mahowald et al., 2006)
r.elevation	SRTM (m)	90 m	C	(Jarvis et al., 2008)
r.ccc	Cation Exchange Capacity	250 m	C	(Hengl et al., 2017)
r.ph	Soil pH in H ₂ O solution (x10)	250 m	C	(Hengl et al., 2017)
r.phkcl	Soil pH in KCl solution (x10)	250 m	C	(Hengl et al., 2017)
r.clay	Clay (weight %)	250 m	C	(Hengl et al., 2017)
r.orc	Soil organic carbon (weight %)	250 m	C	(Hengl et al., 2017)
r.bulk	Bulk density (kg m ⁻³)	250 m	C	(Hengl et al., 2017)
r.bouguer	WGM2012_Bouguer	2 min	C	(Balmino et al., 2012)
r.map	Mean annual precipitation (mm.yr ⁻¹)	30-arc sec	C	(Hijmans et al., 2005)
r.mat	Mean annual temperature (°C)	30-arc sec	C	(Hijmans et al., 2005)
r.nfert	Global Nitrogen Fertilization	30-arc sec	C	(Potter et al., 2010)
r.pfert	Global Phosphorus Fertilization	30-arc sec	C	(Potter et al., 2010)

4.4 Random forest regression and spatial predictions

4.4.1 Random forest regression procedure

All statistical analyses and figures from this manuscript are conducted in R programming language version Rx64 3.4.2. (<https://www.r-project.org/>). An example of R-script is available in supplementary material Script S1. We used random forest regression to predict bioavailable ⁸⁷Sr/⁸⁶Sr variability using the compiled database (Table S1), the bedrock model and the covariates described above following the framework developed by Bataille et al. (2018) and the caret package (Kuhn, 2008). Random forest is a tree-based machine-learning algorithm trained by bootstrap sampling and random feature selection. In a decision tree, a random subset of the dataset is entered, and then each predictor splits the original dataset into smaller and smaller sets at nodes in the tree. Random forest takes this idea to the next level by constructing an ensemble of trees (or forest) using bootstrapping. Specifically, random

forest creates multiple decision trees on different data samples where sampling is done with replacement to prevent overfitting. To make fair use of all potential predictors, the number of features split at each node of a tree is limited to some user-defined threshold. Ultimately, random forest aggregates the results of these decision trees to predict the mean value of the response variable, in our case the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. Random forest is a practical algorithm that requires very little pre-processing. No transformation is required for $^{87}\text{Sr}/^{86}\text{Sr}$ data as random forest makes no assumptions about the data distribution and residual heteroscedasticity. Random forest can also directly integrate categorical variables in multivariate regression so long as they do not have too many classes (Strobl et al., 2007).

In our approach, random forest regression models were optimized using root mean squared error (RMSE) as the primary metric and a 10-fold repeated cross-validation scheme with 5 repetitions using 80% of the data for training at each iteration. Variable selection was performed using the *VSURF* package (Genuer et al., 2015). Once a model is optimized, we used variable importance purity measure and partial dependence plots to describe the relationships between the selected predictors and predicted $^{87}\text{Sr}/^{86}\text{Sr}$. Ultimately, the $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes were generated using the best performing random forest regression model for each substrate (plant, soil, local animal, water) and the associated predictors.

Spatial uncertainty assessment is critical for using isoscapes in continuous-probability surface models of geographic assignment (Wunder, 2012). However, while random forest provides a mean $^{87}\text{Sr}/^{86}\text{Sr}$ prediction using the selected predictors, there are no built-in features to assess spatially explicit model uncertainty. To circumvent this issue, we calculated an uncertainty function for each trained model that relates the mean absolute residual values to predicted $^{87}\text{Sr}/^{86}\text{Sr}$ (e.g., Fig. 7D). This function reflects the observation that uncertainty tends to increase with increasing predicted $^{87}\text{Sr}/^{86}\text{Sr}$. While using quartile random forest regression to calculate the interquartile range would be ideal (as in Bataille et al. 2018), this method is computationally intensive. Our uncertainty function provides an average standard deviation at each pixel that that can be directly used in probabilistic provenance assignments (Wunder, 2012).

4.4.1 Comparison of random forest regressions among substrates

We applied random forest regression independently to the plant, soil, local animal, and water datasets (Fig. 6). All substrates show very similar predictors after the *VSURF* variable selection step (Fig. 7 and Appendix A). Random forest models applied to plant, soil, and local animals perform similarly, whereas the model using only water data has a lower performance (Fig. 6). In our approach, we extracted the values of covariates using site location and the closest underlying 1 km² pixel. This approach is appropriate for plants, soils and local animals because these substrates integrate Sr sources over local spatial scales. However, river water frequently integrates Sr sources over much larger spatial scales and from groundwater sources that are not represented in geological maps. Although extracting local environmental conditions at 1 km² pixel resolution might be appropriate for small streams, it becomes inaccurate for large rivers, which comprise a large proportion of the database. Future predictive work focused on surface waters could use watershed-integrated covariates to address this issue. Another reason for the lower model performance for water samples, and to a lesser degree soil samples, is their broader distribution of $^{87}\text{Sr}/^{86}\text{Sr}$ (due to samples coming from a larger variety of geologies; Fig. 6B and 6D).

To simplify visualization, we trained the main model using a dataset combining plant, soil and local animal $^{87}\text{Sr}/^{86}\text{Sr}$ (herein called the “combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset”). We excluded the water samples from this subset due to the difference in spatial integration represented by this substrate. Model predictors, results, and residuals for individual substrates including plant, soil, local animal, and water are available in supplementary material Appendix A.

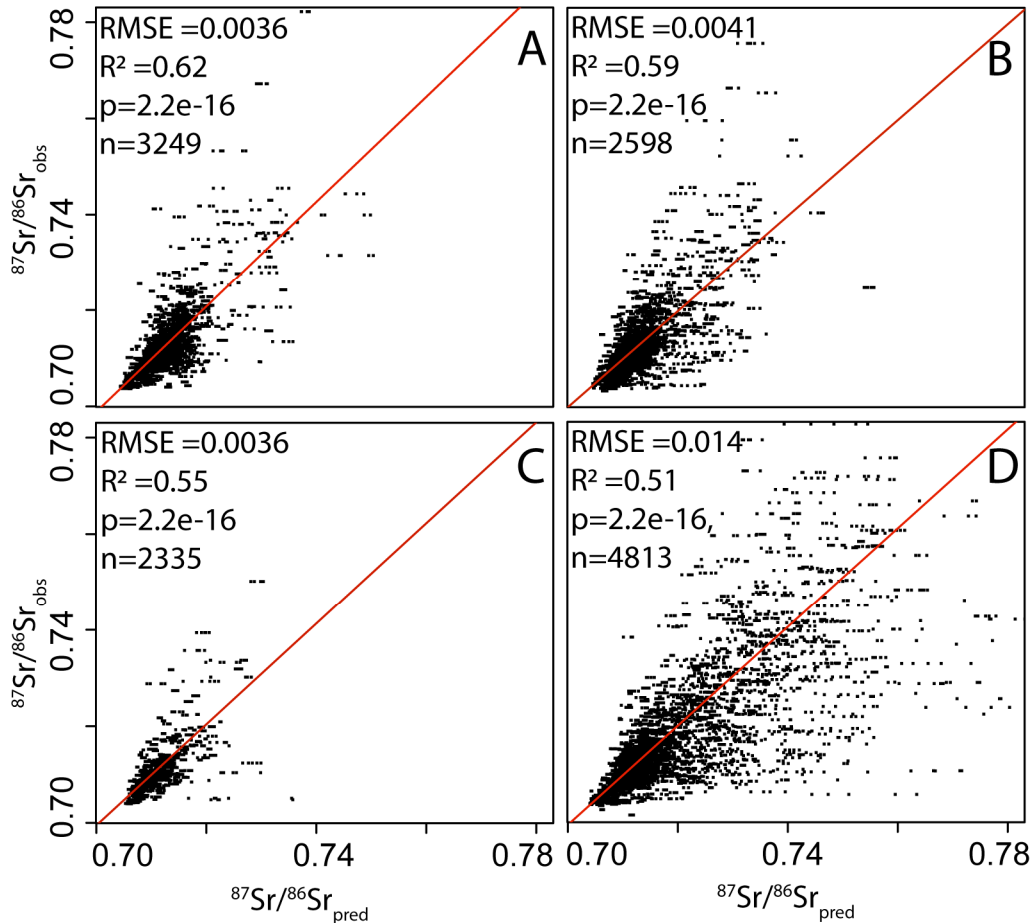


Figure 6: n-fold cross-validation for random forest model by substrate. A) Plants, B) Soils, C) Local animals, and D) Water. RMSE = Root Mean Square Error. Red lines are best fit linear models.

4.4.2 Global bioavailable model performance

After *VSURF* feature selection, Bataille et al. (2014)’s model products (*r.srsrq1* and *r.srsrq3*) and geological variables (*r.age*, *r.minage_geol*) from the GLiM database were the dominant predictors of the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset (Fig. 7A). Other important predictors of $^{87}\text{Sr}/^{86}\text{Sr}$ included dust and sea salt aerosol deposition (*r.dust* and *r.ssaw*), elevation (*r.elevation*), climate variables (*r.pet* and *r.mat*) and soil properties (*r.ph* and *r.clay*) (Fig. 7A). After n-fold cross validation, the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ model explains 60% of the variance, with a RMSE of 0.0034 over the dataset (Fig 7B). The value of 0.0034 represents < 10% of the full range of observed bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ over the compiled dataset. However, this uncertainty is not uniform across the prediction range. For low bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ (<0.710), the RMSE is low (<0.001), with lowest uncertainty values for

$^{87}\text{Sr}/^{86}\text{Sr} \sim 0.709$ (Fig. 7C and 7D). However, as $^{87}\text{Sr}/^{86}\text{Sr}$ increases, the absolute values of residuals increase (Fig. 7C and 7D). This observation conforms with previous studies (e.g., Bataille et al. 2018) that ecosystems developing on older, more felsic rock units (e.g., cratons, Precambrian metasediments) not only have higher intra-site $^{87}\text{Sr}/^{86}\text{Sr}$ variability (Fig. 4) but are also much harder to predict accurately. Ecosystems developing on carbonate units have the lowest intra-site $^{87}\text{Sr}/^{86}\text{Sr}$ variability (Fig. 7D).

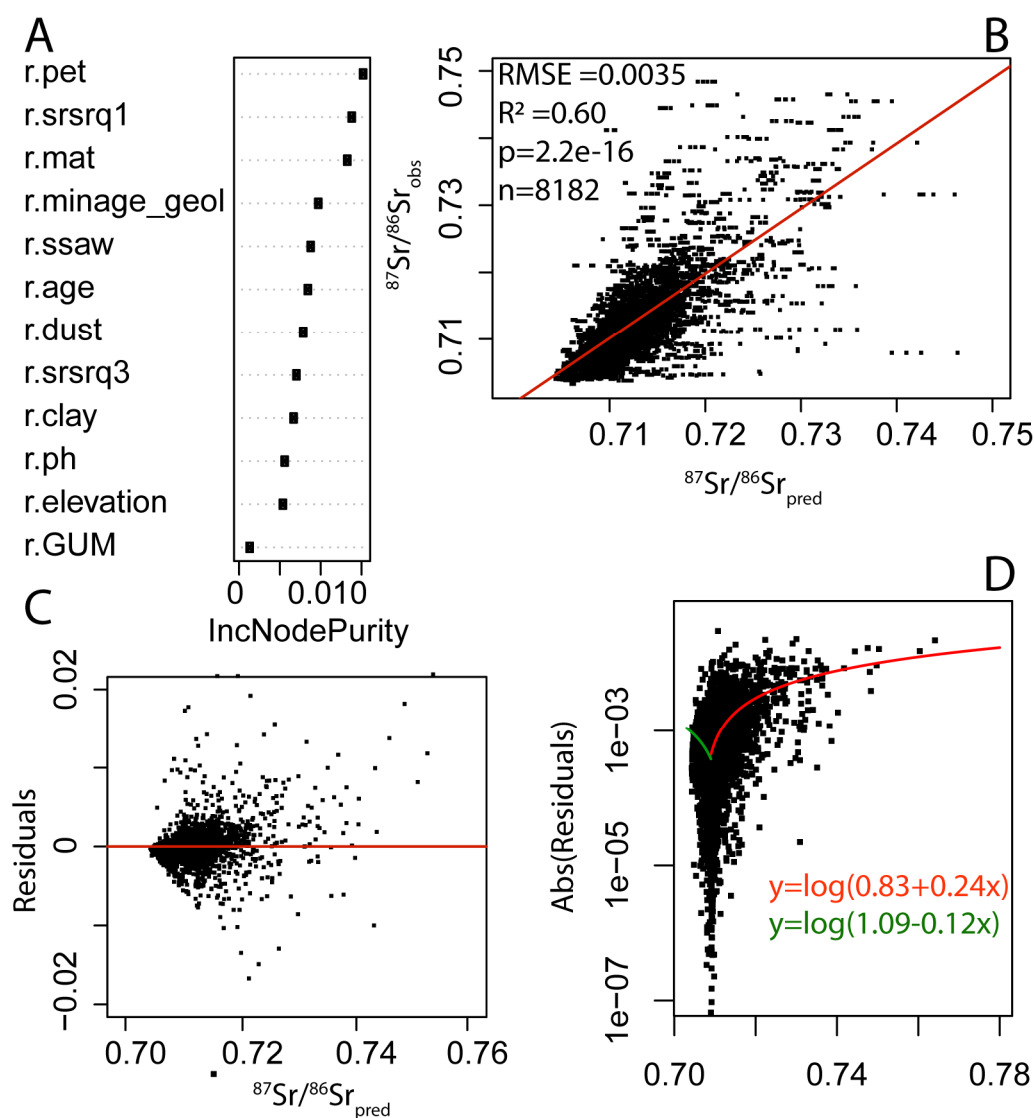


Figure 7: Random forest regression model for the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset combining plant, soil and local animal samples: A) Variable importance plot after selection of predictors by *VSURF*; B) N-fold cross-validation results with best fit linear model (red line); C) Residuals against $^{87}\text{Sr}/^{86}\text{Sr}_{\text{pred}}$; D) Absolute residual values (logscale) against $^{87}\text{Sr}/^{86}\text{Sr}_{\text{pred}}$. Green line indicates the best fit non-linear model between 0.703 and 0.709; red line indicates the best fit non-linear model between 0.709 and 0.780. Refer to Table 1 for predictor names.

4.4.3 Predictors of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ variability

We used a partial dependence plot to investigate the relationship between bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and the selected predictors for the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset (Fig. 8). As expected bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ increased with increasing bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ (r.srsrq1 and r.srsrq3), as well as the age of geological units (r.minage_geol) and terranes (r.age). These relationships confirm the dominance of age and lithology of rock units in controlling bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ at the global scale (Bataille et al., 2014; Bataille and Bowen, 2012). However, we also observe a lack of association between geological variables and bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ for older rock units (Fig. 8). This observation confirms that the current set of geological predictors (i.e., bedrock model products and GLiM products) are inadequate for explaining the large variability of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ for rock units with higher $^{87}\text{Sr}/^{86}\text{Sr}$.

A few additional geological predictors influence bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ in the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset, including surficial deposit types (r.GUM), elevation (r.elevation), and soil proprieties (r.ph and r.clay). For surficial deposits, higher bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is found in regions dominated by siliciclastic surficial sedimentary units, including unconsolidated alluvial, fluvial, glacial and aeolian sediments (Fig. 8), whereas lower $^{87}\text{Sr}/^{86}\text{Sr}$ is observed for marine sediments (evaporites and carbonates), and pyroclastic units (Fig. 8). While the global unconsolidated sediment map (GUM) is a significant predictor of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$, its predictive potential could be improved in future modeling efforts by characterizing the parent rock of each sedimentary unit using detrital zircon databases. Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data also show a positive relationship with elevation, probably due to the preferential uplift and exposure of older radiogenic units during orogenies. Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ decreases with soil pH and soil clay content, likely underlining the dominance of carbonate weathering in more basic soils. We did not find any significant relationship between N and P fertilization inputs and bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. However, this does not rule out the potential impact of liming on bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ in some settings (Thomsen and Andreasen, 2019).

Multiple climate variables also strongly influence bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ shows an exponential increase with both mean annual temperature (r.mat) and potential evapotranspiration (r.pet). These relationships are likely coincidental and reflect the strong sampling bias towards hot regions located on Precambrian cratons (e.g., Madagascar, South Africa, and Tanzania; Fig. 2). The relationship between dust deposition (r.dust) and bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is complex. Bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ decreases for moderate dust deposition but increases at higher deposition rates. This relationship is probably associated with the different isotopic signatures of dust sources: Dust with elevated $^{87}\text{Sr}/^{86}\text{Sr}$ dominates in regions with the highest deposition rate (e.g., Sahara Desert) while lower $^{87}\text{Sr}/^{86}\text{Sr}$ and deposition rates are observed in other arid regions (e.g., Southwestern USA, South America). Last, bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ converges towards 0.71 with increasing sea salt aerosol deposition, which is consistent with inputs of marine-derived Sr in coastal ecosystems.

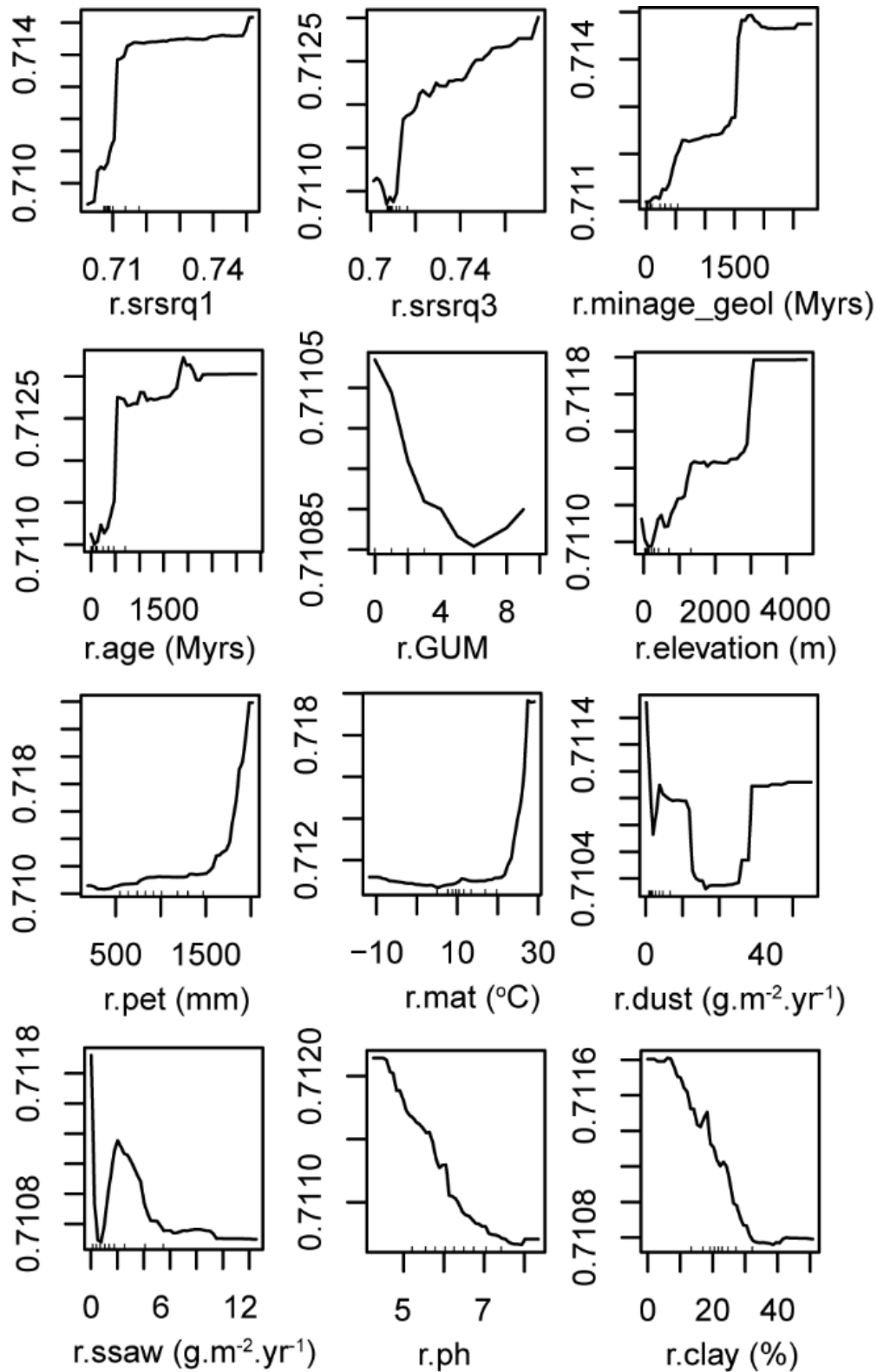


Figure 8: Partial dependence plots between predictors (x-axis) and predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ (y-axis) from random forest regressions using the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset. Refer to Tables 1 for description and sources of each covariate. Hash marks along the x axis show covariate sample decile values. For $r.GUM$, the x-axis represents unconsolidated sediment categories with 0 = No surficial sediment reported; 1 = Colluvial; 2

= Eolian; 3 = Glacial; 4 = Lacustrine; 5 = Marine; 6 = Organic; 7 = Evaporite; 8 = Pyroclastics; 9 = Coastal.

4.4.4 Patterns in bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ variability

We used the random forest regression results from the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ at the global scale (Fig. 9). The spatial uncertainty map associated with these predictions is calculated using the relationship between absolute residual value and bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ predictions (Fig. 7D). The median bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ predictions show similar spatial patterns to predictions by the mechanistic global bedrock model (Fig. 5). High bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is predicted for cratonic and mountainous regions dominated by older, felsic bedrock units, as well as arid regions across the Sahara Desert and the Middle East, where dust with elevated $^{87}\text{Sr}/^{86}\text{Sr}$ substantially contributes to the bioavailable Sr pool. Low $^{87}\text{Sr}/^{86}\text{Sr}$ is found in arc settings and carbonate-dominated regions. However, the overall variability in predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is lower than the bedrock model. This buffering is consistent with our knowledge of Sr isotope cycling from rocks to ecosystems. The majority of predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ falls within a tight range from 0.7085 to 0.711, and converges towards 0.710 (Fig. 3).

This convergence towards 0.710 likely reflects the mixing of Sr from two main sources: 1) Siliciclastic Sr and 2) marine Sr. Siliciclastic sediments are volumetrically the dominant parent material to most ecosystems (Hartmann and Moosdorf, 2012). As described earlier, silicates have a broad range of $^{87}\text{Sr}/^{86}\text{Sr}$ ranging from 0.703 in mafic environments to more than 0.720 in older felsic units with an average upper crust value of 0.716. However, while silicates constitute the main parent material to most soils, they contain little Sr, weather slowly, and do not represent the dominant source of Sr to most ecosystems. Conversely, carbonates and evaporites cover only a small portion of the Earth surface (Hartmann and Moosdorf, 2012), but they contain more Sr and weather faster than other rock types (Palmer and Edmond, 1992). Consequently, marine Sr, with its comparatively tight isotopic range (0.707-0.709) tends to contribute to most ecosystems across the globe through direct weathering of carbonate units, weathering of trace carbonates in shales, deposition of carbonate dust, and/or addition of sea salt aerosols.

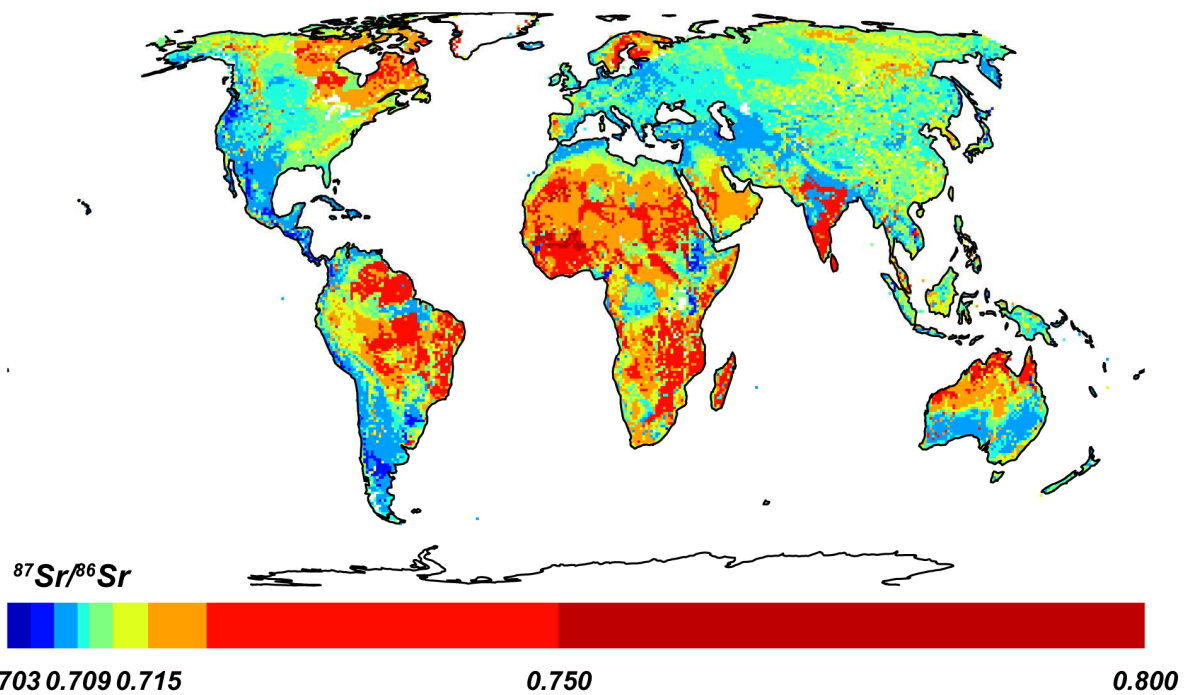


Figure 9: Global map of predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ from random forest regression.

5. Testing the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape

5.1 Regional dataset

We tested the performance of the model in two regions with different geological settings and sampling density to provide guidance on how to use this global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape. First, we used data collected through the GEMAS project (Hoogewerff et al., 2019) to test the performance of the global model in a data-rich region. To date, GEMAS is the most systematic and comprehensive continental-scale dataset of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. The dataset includes close to 1,200 soil samples from a large collection of grazing (Gr) and agricultural (Ap) soils in Europe. It also covers a broad geographic range, including Eastern and Northern Europe with diverse geology, climate and environmental conditions (e.g., Baltic Shield), making the dataset ideal for testing the performance of the global model in a data-rich region. Second, we examined bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ for the island of Madagascar. The dataset includes published data (Burney et al., 2020; Crowley et al., 2018, 2017b; Crowley and Godfrey, 2019) as well as previously unpublished data from plants, modern rodents and subfossil local animals ($n = 279$) at 54 individual locations (See supplementary material Appendix B). Madagascar, and Africa as a whole, is a data-poor region for bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$. The closest bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset in our global database is from South Africa (Copeland et al., 2016). Additionally, Madagascar is geologically heterogeneous and complex. Its bedrock geology spans Earth's history, from the Archean to the Quaternary (reviewed in Crowley and Sparks, 2019). Most of the island is dominated by Precambrian units with varied lithologies. Variable climate and rugged topography further complicate Sr isotope cycling from rocks to ecosystems. Ultimately, Madagascar is one of the regions of the world where bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ is likely to be highly heterogeneous, which will likely affect the predictive accuracy of our global model.

5.2 Model comparison approach

We compared the performance of four modeling approaches to predict bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ for the GEMAS dataset and the newly amassed Madagascar dataset. First, we compared each bioavailable dataset to bedrock model predictions. Second, we compared the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ datasets to predictions from our non-locally calibrated global random forest regressions. For these models, we applied random forest regression using the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ compilation (i.e., combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset) but selectively removed bioavailable data from GEMAS or from Madagascar depending on the location tested. The goal of this step was to assess the possibility of extrapolating global model predictions for an area with little or no bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data. Third, we compared the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ datasets to the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ model calibrated using the global compilation including $^{87}\text{Sr}/^{86}\text{Sr}$ data from GEMAS and Madagascar. Lastly, we compared the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ datasets to locally calibrated models. For these models, we applied random forest regression using only the GEMAS or newly amassed Madagascar $^{87}\text{Sr}/^{86}\text{Sr}$ dataset, respectively (Fig. 9).

5.3 Results of model comparisons

5.3.1 Europe

In Europe, we found that the bedrock model alone explains 30% of the variance in the GEMAS $^{87}\text{Sr}/^{86}\text{Sr}$ dataset (Fig. 10A), confirming that bedrock $^{87}\text{Sr}/^{86}\text{Sr}$ is an important driver of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ (Hoogewerff et al., 2019). The good performance of the bedrock model also reflects the high precision of geological maps used in the GLiM database for Europe (Hartmann and Moosdorf, 2012). Random forest regression using exclusively local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data is the model that best predicts bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ from the GEMAS dataset (Fig. 10D). However, the performance of this locally calibrated model is comparable to the globally calibrated models (Fig. 10B and 10C).

When calibrating a random forest using only GEMAS soil data, nitrogen fertilization rate becomes a significant predictor in the regression (Fig. 10D). As GEMAS is exclusively focused on agricultural soils, it is expected that fertilization practices (e.g., liming) impact $^{87}\text{Sr}/^{86}\text{Sr}$ in the exchangeable soil fraction (Frei et al., 2020; Hoogewerff et al., 2019; Thomsen and Andreassen, 2019). This predictor was not selected when calibrating the model using the combined global and local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset, which suggests that local calibration might be more appropriate in certain cases when trying to predict one specific substrate (e.g., soil). However, use of local calibrations that include more regionally- or system-specific model relationships carries the potential risk of producing errant predictions if the model is applied to areas where these relationships are irrelevant or inconsistent with the calibration data. A firm understanding of the underlying mechanisms and drivers of $^{87}\text{Sr}/^{86}\text{Sr}$ variation is crucial in developing and using such models.

We further demonstrate that the global random forest regression excluding GEMAS data (Fig. 10B) performs nearly as well as the random forest that includes GEMAS data (Fig. 10C). This observation highlights the potential of extrapolating predictions in data-rich regions. Importantly, we underline that removing the GEMAS dataset from the training set does not remove all the European data. Many studies have collected bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data in Western Europe (see compilation in Bataille et al., 2018). While those datasets do not cover parts of eastern and northern Europe included in the GEMAS dataset, they do provide a strong basis for calibrating the relationships between bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and the covariates in unsampled regions across the entire European continent. The success of this extrapolation

likely depends on the similarity of geological and environmental conditions found in the under-sampled areas with those of the training set. This observation also indicates that the sampling density in Europe is probably sufficient to train accurate $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes with the current set of predictors. Additional sampling will only improve this performance marginally.

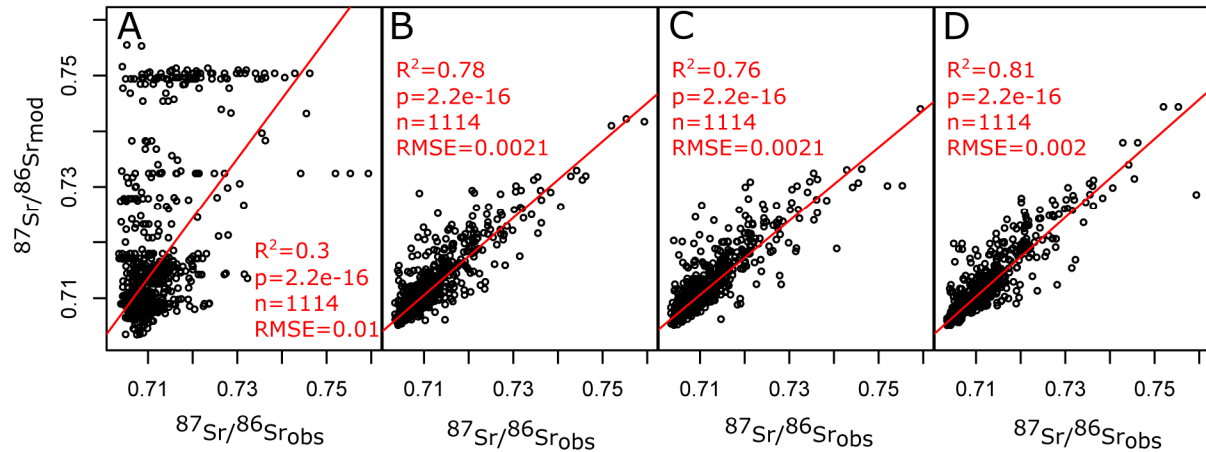


Figure 10: Cross-validation of different models for the GEMAS dataset: A) Bedrock model; B) Random forest regression calibrated using the global bioavailable data without data from GEMAS, C) Random forest regression calibrated using global bioavailable data including bioavailable data from GEMAS, and D) Random forest regression calibrated using bioavailable data from GEMAS only. Red lines are best-fit linear models.

5.3.2 Madagascar

In Madagascar, we found that the bedrock model does not perform as well as in Europe (Fig. 11A). This was not surprising as the map used in the GLiM for Madagascar is outdated and has a low resolution (Besairie, 1964). As mentioned earlier, Madagascar is a very geologically complex region, and the lack of detailed geological maps strongly limits the ability of the bedrock model to predict $^{87}\text{Sr}/^{86}\text{Sr}$ in the geosphere or the biosphere. A much more detailed geologic map of Madagascar does exist (Roig et al. 2012); integrating updated products like this into the GLiM will help improve global $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape models.

In Madagascar, the performance of the locally calibrated model (Fig. 11D) is significantly improved in comparison with the globally calibrated models (Fig. 11B and 11C). Additionally, the globally calibrated model excluding Madagascar data performs poorly (Fig. 11B). While most of the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data fall on a strong correlation line, several large residuals limit the model accuracy. These large residuals are from bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data collected on old metamorphic and sedimentary rock units that represent geological and/or environmental conditions that were not encountered in the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ training dataset. Under such conditions, extrapolation of global $^{87}\text{Sr}/^{86}\text{Sr}$ predictions becomes invalid. Even when including the Madagascar data in the global compilation, these data are not sufficient to fully overcome the strong predictive bias towards data-rich regions. This result underlines the need for local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data in regions that are geologically complex and under-sampled. With the current set of covariates, the global isoscape is well-calibrated for Europe and North America where most bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data have been sampled but poorly calibrated in other regions. To solve this issue, more bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data are required across Madagascar and Africa, particularly from

older radiogenic units, in order to calibrate the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ model with geological and environmental predictors in this region.

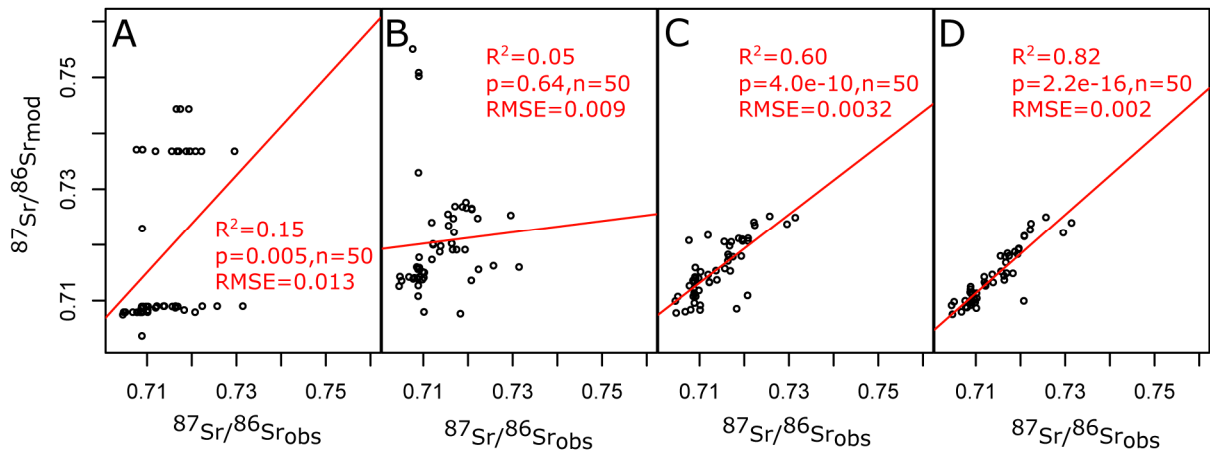


Figure 11: n-fold cross-validation of different models on the Madagascar dataset: A) Bedrock model; B) Random forest regression calibrated using the global bioavailable data without data from Madagascar, C) Random forest regression calibrated using global bioavailable data including bioavailable data from Madagascar, and D) Random forest regression calibrated using bioavailable data from Madagascar only. Red lines are best-fit linear models.

6. Guidelines, knowledge gaps and new research avenues

6.1 Guidelines for using the global isoscape

Provenance studies have underlying assumptions specific to the sample type and the question being addressed. The predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape presented here (Fig. 9) is best suited as a broad scale approach for 1) excluding provenance areas and 2) informing where targeted sampling for a specific research question should occur. When samples in question exhibit a limited range in bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$, as is the case for plants, soils, and animals with small feeding ranges (e.g., non-migratory rodents), the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape can also be used to predict areas of natal origin. However, the current bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape can be considered robust only in data-rich areas, and extrapolations to other regions should be approached cautiously, particularly where geological and environmental conditions differ from those represented in the training set. In data-poor regions, the accuracy and resolution of the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape should be tested by collecting additional data. We encourage other researchers to test, and if required recalibrate, their own $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes using the global framework presented here (see supplementary material script S1), and also to add new local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data to the global compilation (more details provided below). The number of additional data required to calibrate the model to a specific study area is challenging to determine. A cost-effective strategy would be to collect an initial small dataset to test the accuracy of the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ model to help verify its performance. When performance is poor (e.g., Madagascar), collecting more data should be considered depending on the scale of the study, the complexity of the geology and the existing distribution of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data. However, as additional data are included in the modeling, model predictions will probably degrade in some data-rich regions as the global model looks for the best prediction compromise given available data and covariates. The number of predictors will need to be increased to avoid this biasing. As demonstrated in this review, another possible solution for local and regional studies is to calibrate a model using only local to regional bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data.

6.2 Knowledge gaps

6.2.1 Improving the global database, analytical methods, sampling strategy and centralized repository

Although the $^{87}\text{Sr}/^{86}\text{Sr}$ data used in the combined local bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ training dataset appear to be quite dense (Fig. 1), there are still huge data gaps in many environmental, geographic and geological settings that limit the accuracy of the global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape. The current global compilation is heavily biased towards agricultural and densely populated regions of Europe and North America (Fig. 2). This sampling bias is propagated to the global $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape, leading to degradation of predictions in under-sampled parts of the world (e.g., Madagascar). High latitudes, desert, semi-arid regions, tropics, wetlands, and mountainous areas are largely under-represented. Most cratonic regions and their associated sedimentary basins are also largely under-represented (e.g., Africa, Australia, Canada, or Brazil). Gaps from many remote regions could be filled through targeted Sr isotope analysis of samples from museum collections (e.g., rodents; <https://arctosdb.org/>; Fig. 12). These samples could be used to facilitate the development of $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes in high-latitude regions with applications for migratory birds, megafauna or early human mobility. As demonstrated in this review, regional accuracy of the global model could be significantly improved by adding only tens to hundreds of points in under-sampled areas (Fig. 11). Conversely doubling the number of points in already well-sampled regions will only bring minor improvement (e.g., Europe). As bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data are positively skewed, sampling needs to account for the higher variability in older and more complex geological settings. Sampling those regions at high density might help capture some of the high variance observed in these regions. This type of sampling rationale can be systematized using available statistical algorithms (e.g., Latin Hypercube) that use the distribution of existing covariates (e.g., geology, climate) to optimize the sampling strategy at a given location or globally (Minasny and McBratney, 2006).

Filling these data gaps will require $^{87}\text{Sr}/^{86}\text{Sr}$ analysis of thousands of samples. Despite significant analytical improvements in the last decades, analyzing $^{87}\text{Sr}/^{86}\text{Sr}$ is currently expensive and slow in comparison with other isotopic systems analyzed using continuous flow stable isotope ratio mass-spectrometry or cavity ring down spectroscopy. In the last few years, new methods have emerged that make $^{87}\text{Sr}/^{86}\text{Sr}$ analysis faster and more affordable, increasing the possibilities of generating high-density datasets. For example, the use of Laser Ablation (LA)-MC-ICPMS instead of solution methods for analyzing solid samples with high Sr content (e.g., animal teeth) allows very high throughput, limited sample preparation and sufficient analytical precision for most provenance studies (± 0.0001). This method could help develop large datasets from museum specimens (e.g., rodents) and improve the accuracy of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes in remote regions (Fig. 12). More recently, the use of ICP-MS/MS with in-line Rb separation has been proposed to increase throughput, decrease cost and limit sample amount for solution methods (Murphy et al., 2020). This method provides fast and relatively inexpensive analysis of small biological samples (e.g., insect tissues) with similar analytical precision to LA-MC-ICPMS (± 0.0001). Lastly, the addition of an autosampler with syringe injection on MC-ICP-MS instruments (e.g., microFAST-MC) has contributed to increased throughput and reduced mass requirements without compromising analytical precision. To increase throughput, MC-ICP-MS users could further reduce the integration time and number of ratios analyzed. This would decrease analytical precision, but, as mentioned above, precision of ± 0.0001 is usually sufficient in provenance studies. We encourage researchers to further develop and adopt these analytical methods to continue decreasing the price and time required for $^{87}\text{Sr}/^{86}\text{Sr}$ analysis.

One final critical issue when generating a bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ dataset is the lack of guidance on the metadata required when collecting bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data (Grimstead et

al., 2017). Most bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ datasets include some fields representing location and isotopic data. The remaining metadata provided by authors vary as no metadata template exists in the community. This is problematic because different fields or substrates require different types of metadata. Moreover, many metadata fields are often required to better screen and use bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data for provenance applications. In compiling the dataset for this study, geographic coordinates, substrate type (e.g., plant, soil, water, animal tissue), sample details (e.g., plant species, soil depth), tissue sampled (e.g., enamel versus dentine, whole plant versus leaves), analytical method, and analytical precision needed to be mined from the main manuscript or directly from the authors in many cases. This is time-consuming, not always successful, and could be entirely avoided if appropriate data and metadata templates were provided to the community. These challenges are being addressed by the development of IsoBank, a centralized repository for isotope data (Pauli et al., 2017). This repository will contain data templates specific for bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data that will facilitate the integration of data from multiple sources and fields (planned launch date is 2021).

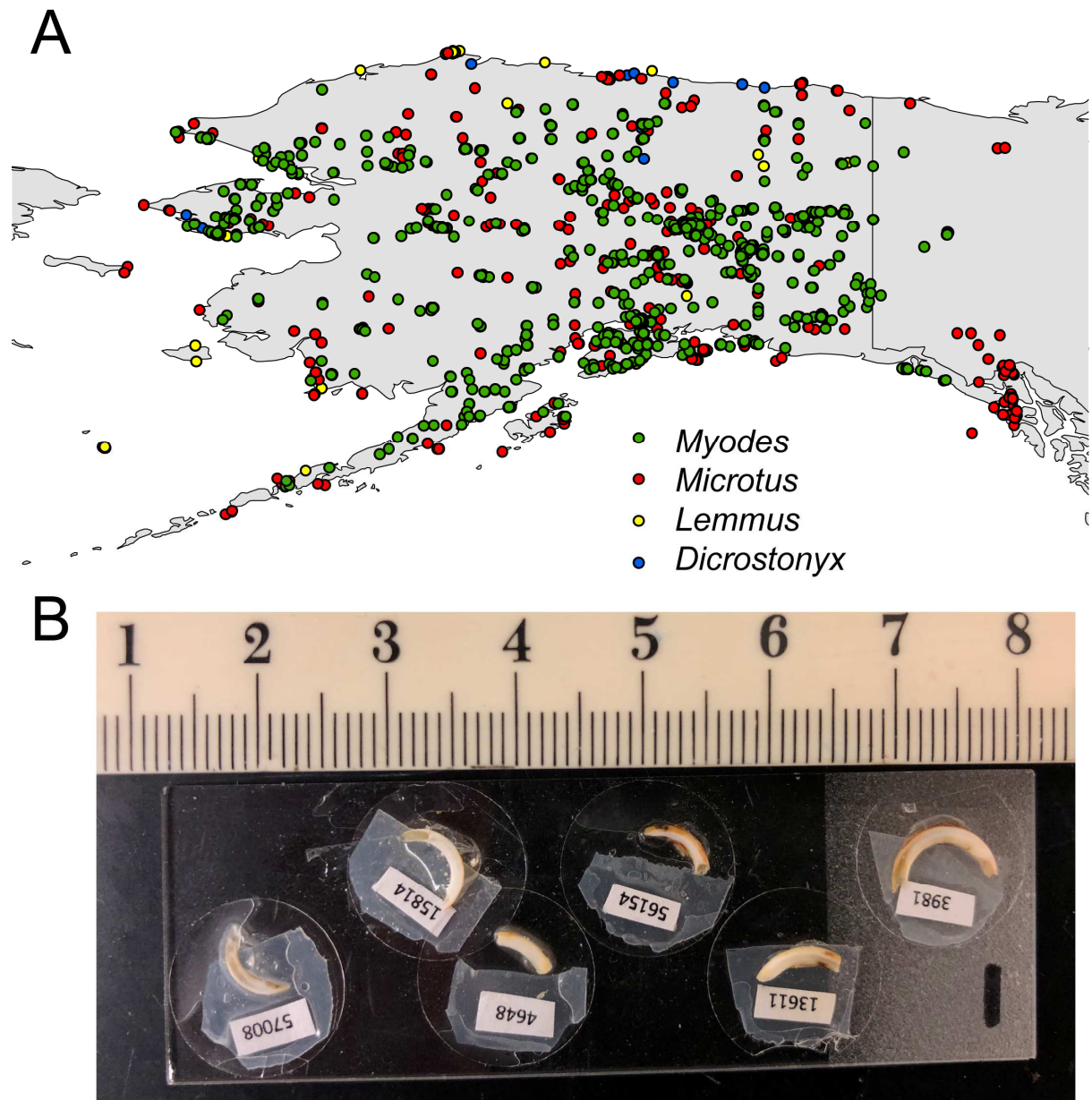


Figure 12: An example of filling $^{87}\text{Sr}/^{86}\text{Sr}$ data gaps in a remote region (Alaska): A) Results of a query on the Arctos database (<https://arctosdb.org/>; Accessed April 1, 2020) for teeth of selected non-migratory rodents (including *Lemmus*, *Microtus*, *Myodes* and *Discrostonyx* species) in the mammal collection at the University of Alaska Museum of the North. B) Mounting of rodent teeth prepared for cost-effective and rapid LA-ICP-MS $^{87}\text{Sr}/^{86}\text{Sr}$ analysis.

6.2.2 Improving modeling

It has already been demonstrated that random forest outperforms most linear and non-linear models by better representing complex non-linear relationships in bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data (Bataille et al., 2018). The current script (see supplementary material script S1) is designed to handle large datasets with dozens of covariates. This script uses parallelization to boost raster calculation and random forest prediction through the *caret* (Kuhn, 2008) and *doParallel* packages (Calaway et al., 2018). However, as the database of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ data grows and/or more covariates are accounted for, the regression matrices will exceed the capability of desktop computers. This high computational intensity is one of the drawbacks of using random forest regression as the computational loads grow exponentially with more data. Random forest is also sensitive to noise and errors in the data and requires a careful quality check, which is often an issue with large interdisciplinary compilations.

As demonstrated in this review, the extrapolation of models fitted using random forest is suitable for data-rich regions but risky for data-poor regions with geologies that fall outside of the calibration dataset, or have outdated geology maps in the GLiM. Sampling biases are also propagated into the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ predictions. Once enough bioavailable data are available from all parts of the world, one solution to limit biasing and computing time will be to train a model on a geographically well-distributed subset of the database. Many of the relationships found by random forest regression are also non-deterministic and not stationary. A good example of this issue is represented by the relationship between bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and dust deposition (Fig. 8). In the global model, dust deposition combines information about dust flux and dust sources into one single variable (r.dust). If new bioavailable data are collected in regions with elevated dust flux with low $^{87}\text{Sr}/^{86}\text{Sr}$ (e.g., China), this will likely degrade the relationship between bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ and dust in the model. This type of issue underlines the need for additional and better covariates which would open-up more targeted modeling opportunities. A large part of the model uncertainty is due to the limitation of global geological maps. Including geological map products is currently essential, as they are the dominant predictors of bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ variability. However, their inaccuracies, lack of homogeneity in resolution and classification scheme, and boundary issues, are also transmitted to the predicted bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ variations. A continuous geospatial dataset that captures geological variability (e.g., radiometric data) or multispectral satellite data (e.g., WorldView-3) would considerably improve the situation. Lastly, while random forest can train accurate models, expert knowledge on geology, geochemistry and environmental science remain critical for interpreting model results. Calibrating a bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape requires carefully verifying the relationships between predictors and response variables. Ultimately random forest regression models should pave the way to improve mechanistic modeling approaches.

6.3 Conclusions and perspectives

Here we have presented the first validated, high-resolution $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape at the global scale, which should be useful to many researchers interested in provenance applications. These $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes provide powerful templates for extrapolating between and beyond

the bounds of existing databases. They can be used independently or coupled with other isotopic systems (e.g., hydrogen, oxygen, and carbon) to provide provenance assignments in a range of fields. To date, $^{87}\text{Sr}/^{86}\text{Sr}$ has been largely underused in ecological applications in comparison with other isotopic systems. However, with the rapid advances in the development of $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes, we anticipate that $^{87}\text{Sr}/^{86}\text{Sr}$ will become a tool of choice for investigating the mobility of migratory species at large spatial scales. $^{87}\text{Sr}/^{86}\text{Sr}$ exhibits limited temporal variability but high-resolution spatial patterns, and offers unique advantages relative to other isotopic systems. New applications of $^{87}\text{Sr}/^{86}\text{Sr}$ to assess the population dynamics and migratory pathways of bird and insect species are ongoing and will reveal the potential of $^{87}\text{Sr}/^{86}\text{Sr}$ in this type of ecological applications. The global bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape is also relevant to investigations of the ecology of extinct animal species. Global $^{87}\text{Sr}/^{86}\text{Sr}$ paleo-isoscapes will be key to resolving questions about megafaunal ecology, early human dispersals, or human societies. Advances in $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes should also make this geochemical tracer increasingly relevant in forensic and food sciences. Such applications will probably require calibrating substrate-specific global $^{87}\text{Sr}/^{86}\text{Sr}$ baselines (e.g., drugs, wine). The up-front cost might be challenging, but once developed, these calibrated isoscapes will be valid for the long term and readily applicable to other markers. While considerable gaps remain in the development of global $^{87}\text{Sr}/^{86}\text{Sr}$ isoscape, this study paves the way for rapid advances in the applications of this tracer in large-scale provenance applications.

For researchers interested in biogeochemical cycles, the development of global $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes and $^{87}\text{Sr}/^{86}\text{Sr}$ compilations offers a novel and exciting research avenue for improving global Earth systems models. We have already underlined that developing pure mechanistic isoscapes is beyond our current knowledge of Sr isotope cycling. However, these knowledge gaps point to a key opportunity for advancing our understanding of biogeochemical cycles through $^{87}\text{Sr}/^{86}\text{Sr}$ modeling. It has long been known that $^{87}\text{Sr}/^{86}\text{Sr}$ is a unique tracer of elemental cycling in rivers, aerosols, and ecosystems at the local scale. Global predictive $^{87}\text{Sr}/^{86}\text{Sr}$ modeling provides the opportunity to scale up this tracer from the local to the global scale. For example, bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ modeling could provide a novel method for understanding soil weathering processes, or estimating the elemental contribution of aerosol inputs to ecosystems, while constraints gained from regression models could help advance the quantitative theory describing the controls of elemental cycling in the hydrosphere, atmosphere and ecosphere. Similarly, developing global $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes in river water would be relevant to better partitioning solute sources in watersheds. At the global scale, $^{87}\text{Sr}/^{86}\text{Sr}$ isoscapes could help quantify the global elemental flux from continental surfaces refining thereby global elemental budget in seawater. With the advances of plate models and paleogeological reconstructions, it might even be possible in the future to reconstruct $^{87}\text{Sr}/^{86}\text{Sr}$ on the Earth's surface in deep time to provide new constraints on global biogeochemical cycles in specific time periods.

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