Problems with Strontium Isotopic Proveniencing in Denmark?

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ABSTRACT

A recent study by Thomsen and Andreasen (2019) has induced a negative reaction to the utility of strontium isotope proveniencing in Denmark. Although there are higher strontium isotope values in the landscape, Thomsen and Andreasen are not correct about the impact of their finding on studies of prehistoric mobility. Several case studies identify such "hotspots" in the landscape and help evaluate their consequences for identifying non-local individuals. In sum, (1) there are small areas of higher strontium isotope values in Denmark, (2) surface water is not a reliable proxy for baseline information on local strontium isotope sources, and (3) strontium isotope proveniencing remains a very useful method for identifying non-local individuals.

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Introduction

There seems to be some confusion and consternation at the present time with regard to the utility of strontium isotopic proveniencing in the archaeology of southern Scandinavia. Strontium isotopic proveniencing is a method for determining if an individual was local or non-local to the place of burial. The principles of the method are straightforward and based upon the premise that ratios of strontium isotopes ($^{87}Sr/^{86}Sr$) vary geographically due to underlying differences in local geologies (Burton et al., 2003; Price et al., 1994, 2002, 2008, 2010). Strontium isotopes enter the human skeleton via the food chain, from rock to sediment, to soil nutrients, to plants, to animals such as humans. Because strontium has a heavy mass, there is no fractionation or change along this path. ^{86}Sr is stable and approximately 10 % in nature; ^{87}Sr is radiogenic, created by the decay of rubidium 87 with a half-life of almost 50 billion years; ^{87}Sr is variable and around 7 % in nature. Thus, older rocks and rocks with more rubidium have higher $^{87}Sr/^{86}Sr$ values and younger rocks generally have lower values. This premise has been demonstrated through several decades of geological research.

These isotopes are deposited in the skeleton and teeth. Most tooth enamel forms in the period before birth until about six years of age. Tooth enamel is one of the most durable tissues in the skeleton, both before and after death and shows very little indication of diagenesis – post-mortem chemical change due to burial. $^{87}Sr/^{86}Sr$ values in human tooth enamel vary from approximately 0.705 to 0.735 and are measured on mass spectrometers accurate to the fourth or fifth decimal pace. If the strontium isotope ratio of tooth enamel differs from that of the local region, the individual likely was not born locally.

In actual fact, levels of strontium isotopes may vary from local geology for several reasons (Maurer et al. 2012; Price et al. 2002), including differing surface deposits introduced by glacial, aeolian, or fluvial processes or in coastal areas, sea spray and rainfall may introduce a marine signal with a value ca. 0.7092. For these reasons it is necessary to measure *bioavailable* levels of $^{87}Sr/^{86}Sr$ to characterize local strontium isotope ratios (Price et al. 2002; Sillen et al. 1998). Local strontium isotope baseline values can be determined by measuring archaeological fauna or even modern specimens of plants or animals in areas where preservation is poor. This information is compared to the isotope ratios in the tooth enamel or bone. If the ratios are different then in all probability the individual in question was not born locally. The specific place of origin can be difficult to determine since different places can have the same strontium isotope ratio, but it is sometimes possible to constrain a potential homeland using a combination of isotopic and archaeological evidence.

I have been working with strontium isotope proveniencing for almost 30 years. Our Laboratory for Archaeological Chemistry at the University of Wisconsin-Madison processed more than 10,000 samples from five continents in that time. I have published or co-authored a number of papers on isotopic proveniencing in Scandinavia over the last decade or so (Price and Gestsdóttir 2006; Sjögren and Price 2006, 2013; Sjögren et al. 2008, 2013, 2016, 2017; Price et al. 2007, 2008, 2010, 2015, 2012, 2013, 2016, 2017, 2018; Price 2008, 2013, 2011, 2018; Frei and Price 2012; Dobat et al. 2014; Frei et al. 2015, 2019; Arcini et al. 2015; Bäckström and Price 2016; Wilhelmson and Price 2017; Bergerbrant et al. 2017; Naumann et al. 2014; Naumann and Price 2020; Kjällqvist and Price 2019).

Perhaps I should also point out my preference for the term "provenience", rather than "provenance". To my mind, provenance has subjective connotations from art history that refer more to context and artist, while provenience is a more objective term from geology that refers to the study of sources and measurement of composition.

This essay is organized as follows to discuss specific aspects of isotopic proveniencing. I initially consider the study by Thomsen and Andreasen. I offer some examples that may explain why there are variations in the $^{87}Sr/^{86}Sr$ baseline in southern Scandinavia. I then turn to a broader discussion of the utility of isotopic proveniencing and why it is important to be cautious in its application. This essay is not intended as an overview of strontium isotope analysis or a critique of Frei's study of the Egtved Girl.

Thomsen and Andreasen (2019)

Many of the concerns with this method in Denmark grew out of a paper by Thomsen and Andreasen (2019) that pointed to the modern practice of liming agricultural fields in Western Jutland and the effect on $^{87}Sr/^{86}Sr$ levels in surface water. This essay is intended to address those concerns and discuss some of the weaknesses and strengths of the method of strontium isotope analysis.

The study by Thomsen and Andreasen was inspired by doubts raised by an article by Frei et al. (2015) that argued on the basis of strontium isotopic proveniencing of her teeth, hair, nails, and bone and some information from an artifact that the Egtved Girl, an iconic burial from the Danish Bronze Age, was not originally from Denmark, but had traveled several times between her homeland in or near the Black Forest in Germany to her residence and place of burial in Denmark, a distance of more than 800 km. The local baseline level of $^{87}Sr/^{86}Sr$ for comparison was determined from measurements of surface water (Frei and Frei 2011). For an archaeologist's perspective, see comments by Hvass (2019) who questions the feasibility of travel over such long distances and argues for investigation of the origin of the artifacts that were included with the burial.

The Thomsen and Andreasen study was an interesting and potentially damaging critique of the Frei et al. (2017a) article and of strontium isotopic mobility studies in general. In essence the authors argued that there were higher $^{87}Sr/^{86}Sr$ values in the landscape, especially in western Jutland, and that modern agricultural applications of lime in areas with non-calcareous soils reduced the strontium isotope ratio of surface water and thus made calculation of local bioavailable strontium from water unreliable. Specifically, the projections of Thomsen and Andreasen meant that the Egtved Girl could have been from Denmark all along. If these conditions applied, then basic reference data for strontium mobility studies would be unreliable.

Fortunately, there are some limitations with the Thomsen and Andreasen study that restrict its conclusions. Frei et al. (2019) argue that the introduction of lime onto fields affects only the upper 60 cm of soil and does not change strontium levels in water below that depth. There are examples where surface water seems to provide a good indicator of baseline values (Maurer et al. 2012; Blank et al. 2018), but many studies (probably most) of bioavailable strontium do not focus on surface water with good reason. One simply does not know the sources of strontium in water. Although it is easy to collect, surface water is not necessarily representative of a particular location as water moves, often long distances, and may incorporate the strontium signal of the different places through which it passes. This caveat involves both depth and distance in water movement. Moreover, water because of its normally low concentration of strontium, does not contribute much to the human consumption of strontium (Bryant et al. 1958, Comar et al. 1957; Elias et al.1957; Lewis et al. 2017) and plays a relatively minor role in body levels of $^{87}Sr/^{86}Sr$. Soil is also not a good proxy for bioavailable strontium as differential weathering of the various minerals in soil can produce very different $^{87}Sr/^{86}Sr$ values (e.g., Maurer et al. 2012; Frei et al. 2019).

There are several other kinds of material more appropriate for measuring strontium isotope baselines than surface water or soil (Bentley et al. 2004; Price et al. 2002; Sillen et al. 1998). These include modern fauna, modern vegetation, archaeological fauna, and/or archaeological human bone. Because there is no fractionation of strontium isotopes due to the heavy mass of the element, almost any organic material in the environment can be measured to obtain the local $^{87}Sr/^{86}Sr$ value. We have for some time (since 2002) advocated the use of archaeological fauna (especially small wild mammals) for the determination of strontium baselines. Grimstead et al. (2017) also have some suggestions for the standardization of strontium isotope baseline environmental data.

Unless the strontium from modern lime somehow contaminates archaeological fauna, this practice would seem to obviate the potential problems from lime application. Fertilizer does not appear to significantly alter strontium isotope ratios either, as most brands have low to intermediate levels of strontium isotope ratios (Frei and Frei 2011; Ria et al. 2004). Moreover, materials buried below 30 cm seem to avoid most contaminants (Bacon et al. 1996; Budd et al. 2000; Frei et al. 2019; Rasmussen et al. 2019).

It should also be noted that tooth enamel is unusually resistant to diagenesis and normally does not take on strontium from ground water after burial (Budd et al. 2000). It is also the case that we have measured archaeological fauna from throughout Denmark and obtained consistent results in the range of 0.709-0.711 (Figure 3A), with a few exceptions discussed below.

High Strontium Values in the Landscape

What is most important from the Thomsen and Andreasen study is the fact that there exist areas with higher strontium isotope values in the landscape that neither the broad sweep of surface water sampling (Frei and Frei 2011) nor the analysis of owl pellets and other faunal remains (Frei and Price 2012) identified. That low visibility suggests that these "hot spots" may be limited in number and small in size, at least outside of western Jutland. There are a few confounding cases of higher values elsewhere in southern Scandinavia as well as some higher strontium spots in the landscape of northern Germany. They are present and need to be identified. I will discuss two examples of such "hotspots" before examining their cause.

One example comes from the Iron Age site of Alken Enge where the human remains of battle victims were placed in lake and bog deposits (Holst et al. 2018; Løvschal and Holst 2018), not far

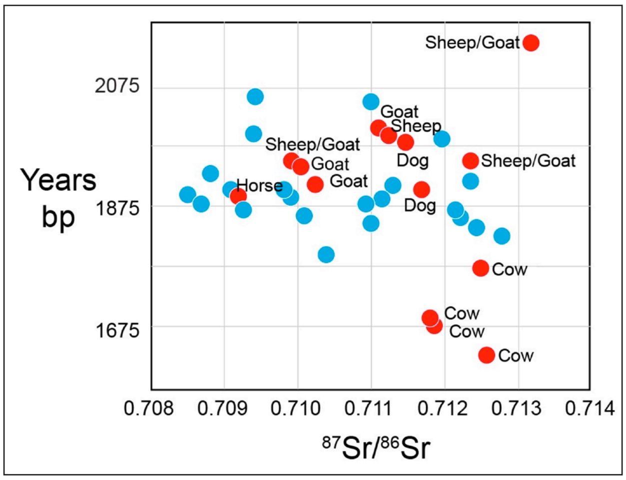
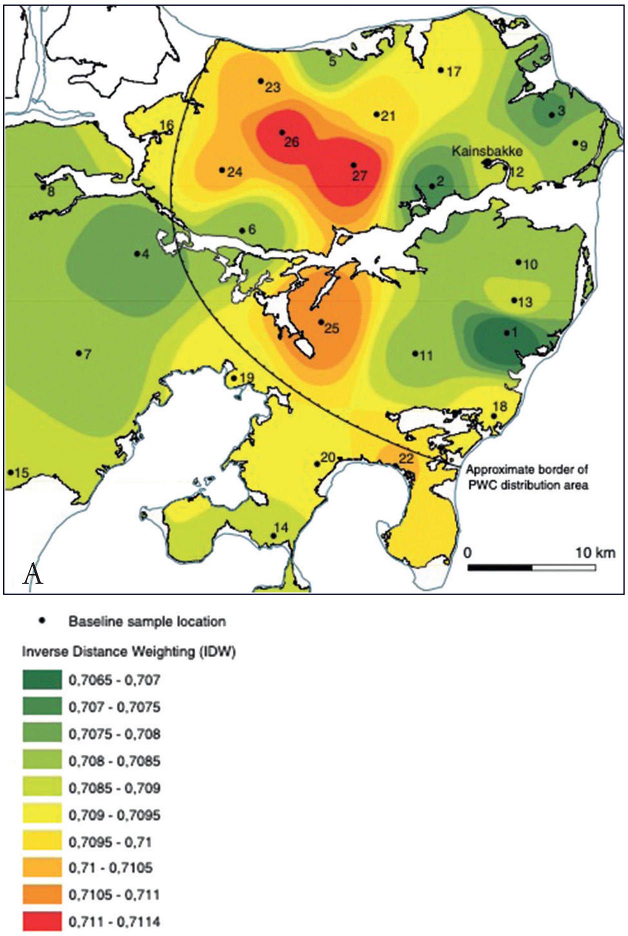
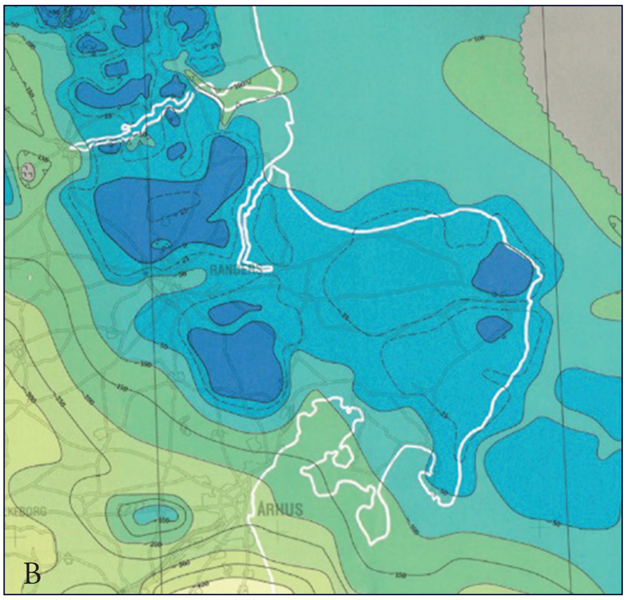


Figure 1. Scatterplot of radiocarbon age vs. $^{87}Sr/^{86}Sr$ value for humans (blue circles) and fauna (red circles) at Alken Enge. Animals with high strontium isotope values can be seen above and below the horizon with the human remains on the right side of the graph.

from the famous war weapons sacrifices at Illerup, roughly 20 km southwest of the modern city of Aarhus. This study was done in collaboration with Mads Holst and the Alken project, funded by the Carlsberg Foundation. We measured strontium isotope ratios on a number of human and animal remains in the bog deposits at Alken Enge. Radiocarbon dates on these samples allow us to see that animals in the younger deposits, which were presumably local, have high $^{87}Sr/^{86}Sr$ values between 0.7117 and 0.7125 (Figure 1). An older sheep or goat has a value greater than 0.713. The human values (blue dots) range from 0.7085 to 0.7127. All of the values greater than 0.712 are higher than expected from the baseline information for Denmark. Given the high values for what should be local animals, it appears that there is a "hotspot"– a higher strontium source at or near Alken Enge.

A second example comes from a region of Denmark known as Djursland and the CONTACT project funded by the VELUX Foundation (Klassen 2020), concerned with the movement of ani-





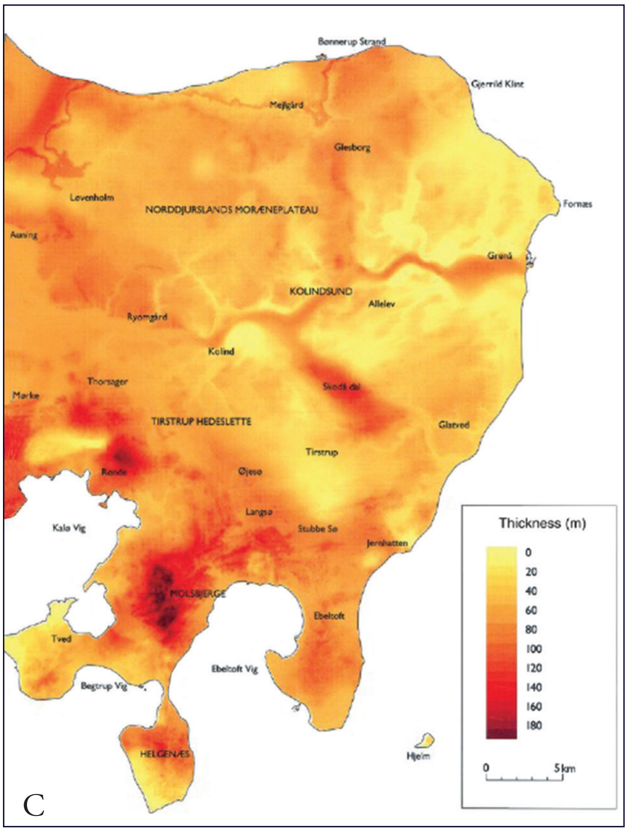


Figure 2. (A) sample locations and $^{87}Sr/^{86}Sr$ contours on Djursland. Coastline reconstructed for the time of occupation of the Kainsbakke site (early 3rd millennium BC) by Klassen (2014). Sample site numbers refer to Table 3. Graphics: Casper Skaaning Andersen, MOMU, with additions, (B) Subsurface bedrock on Djursland. Blue is limestone, green is calcareous clays, Structural map of the Top Chalk Group on Djursland (GEUS; Ter-Borch 1991).
(C) Thickness of glaciogenic sediments on Djursland (GEUS; Schack Pedersen/Strand Petersen 1997).

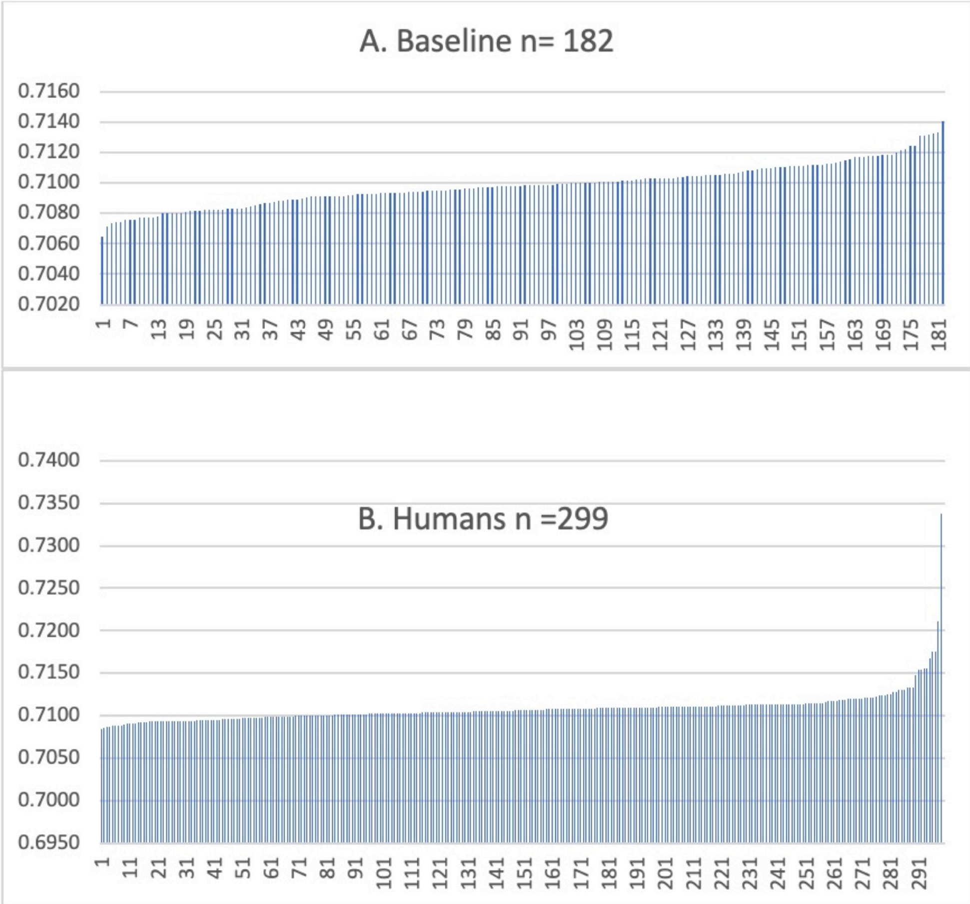


Figure 3. Ranked distribution of $^{87}Sr/^{86}Sr$ values for (A) 182 baseline values primarily from fauna, and (B) 299 values from human tooth enamel from Denmark, all time periods.

mals (associated with people) from western Sweden to eastern Jutland, Denmark, as a part of the Pitted Ware Culture (Klassen et al. 2020; Price et al. 2021). Baseline strontium isotope values were measured in modern mice and voles collected across the peninsula of Djursland. Klassen et al. (2020) note that these values corresponded well with the depth of glacial deposits on top of limestone and chalk in the subsurface deposits, i.e., 87Sr/86Sr values were higher in areas with thicker glacial deposits and deeper calcareous bedrock (Figure 2).

Frei and Frei (2011) noted that the variation in $^{87}Sr/^{86}Sr$ across Denmark could be explained by the variable mixing of the two major sources of Sr. These are (a) Sr derived from pre-Quarternary carbonaceous sediments ($^{87}Sr/^{86}Sr$ = 0.7078–0.7082) and (b) Sr derived from a radiogenic component in Pleistocene glaciogenic soils with Precambrian granitoid components ($^{87}Sr/^{86}Sr$> 0.712). Apparently, Thomsen et al. (2021) have also discovered this mixing of different sources of strontium isotopes. This seems to be exactly the case in Djursland and is likely the situation throughout most of Denmark. The moral of this story is that surface water and large-scale mapping of $^{87}Sr/^{86}Sr$ is unlikely to identify such "hotspots". National baseline reference maps (isoscapes) can provide some sense of regional variation, but it is necessary to develop detailed local baseline maps for every study.

There is good reason to suspect that these "hotspots" are generally small and do not have a major impact on human $^{87}Sr/^{86}Sr$ values in southern Scandinavia. Figure 3 shows the ranked distribution of $^{87}Sr/^{86}Sr$ values for (A) 182 baseline values primarily from fauna, and (B) 299 values from human tooth enamel from Denmark. In both cases the values fall predominantly between 0.708 and 0.712, the range of baseline values predicted for Denmark by surface water (Frei and Frei 2011) and fauna (Frei and Price 2012). The question is whether these areas of higher values within Denmark are large enough to contribute sufficiently higher $^{87}Sr/^{86}Sr$ to raise enamel values. Based on the fact that most of the measured human samples from Denmark fall within the estimated baseline and the fact that higher values are usually distinctly uncommon, the effect of strontium hotspots in Denmark appears to be negligible. It is essential to remember that humans average their intake of strontium isotopes over a period of months or years in building bone and teeth. Thus, if these "hotspots" are small, their contribution to the average ratio measured in enamel will also be small.

Conclusions

My remarks are not intended to answer questions of the origin and mobility of the Egtved Girl. There are several issues in that study that complicate a direct answer (von Holstein et al. 2015; Kootker et al. 2020; Toxvaerd 2020). I would reiterate that most of the information derived from mobility studies is quite informative and useful. I think it is important to note that strontium mobility studies generally work well to identify non-local individuals, but determining place of origin is a much more complicated and difficult undertaking. It is essential to be cautious in the interpretation of such data because it is easy to be mistaken. The existence of multiple areas with the same strontium isotope signature is a strong reason for not attempting to determine place of origin. The complex variation in strontium isotope baseline values in some areas is another reason to be cautious.

Isotopic proveniencing is a relatively new method in archaeology and as such is still under development. There are problems, often associated with establishing ancient baselines for various isotopes. Analysis for isotopic proveniencing is expensive. The method only works for first generation immigrants. It is also the case that determining the place of origin for non-local individuals is rarely possible. At the same time it is obvious that isotopic proveniencing has become an important tool for bioarchaeology. The ability to identify non-local burials or the movement of ancient animals or plants has revolutionized our understanding of the past and contributed to an understanding that movement was common in the past. Mobility and migration have always characterized the human condition.

Conflict of Interest

I have no conflict of interest associated with this essay.

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