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Title: Strontium Isotope Investigation of Human Mobility Based on Teeth from Cave T1

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Seven adult human teeth from Cave T1 were analysed for trace element concentrations using Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICPMS) and strontium isotope compositions using Laser Ablation Multi-Collector Inductively Coupled Plasma Mass Spectrometry (LA-MC-ICPMS). The aim of this study was to examine whether the people buried in Cave T1 were of local or foreign origin, given the known connection of Philistine Tell es-Safi/ Gath with the Aegean, Cyprus and Anatolia (e.g., Maeir 2012). The results for the burial cave specimens were compared to bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values obtained from soil and rock samples from key geological units throughout Israel. This comparison shows that at least four of the individuals have $^{87}\text{Sr}/^{86}\text{Sr}$ values that correspond to the area immediately surrounding the site. The remaining samples analysed in this study have $^{87}\text{Sr}/^{86}\text{Sr}$ values that do not correspond to any mapped samples from Israel. This result is explained not by mobility, but as an artifact of the very low strontium concentration in these teeth, which precludes an effective correction for isobaric interferences during laser ablation analysis.

Strontium Isotopes and Archaeology

Strontium isotope measurements of biological materials from archaeological sites have become a commonly applied technique to discern provenance and to document the distance and vector of mobility during the formation of biogenic carbonates (reviewed by Bentley 2006; Budd et al. 2004; Montgomery 2010; Price et al. 2002). To investigate mobility, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of biominerals from fossil samples are compared to regional values obtained from local faunal material (Price et al. 2002) or from the analyses of the bioavailable component of strontium from plants, regolith or bedrock (Montgomery et al. 2007). The most suitable biomineral is tooth enamel, due to its mineralization during early life and resistance to post-burial diagenesis (Trickett et al. 2003, Austin et al. 2013), although, while being more challenging, bone and dentine can also be used.

The question of mobility can be addressed in a number of ways using $^{87}\text{Sr}/^{86}\text{Sr}$ isotopes. On the most basic level, the method can simply be used to determine if individuals are local or non-local with reference to a background value obtained from the archaeological site or the surrounding area (Bentley et al. 2007; Conlee et al. 2009; Schweissing and Grupe 2000). A more robust approach is to map key geological units from the surrounding area using plant, regolith, bedrock or faunal bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values (Evans 2010; Hodel 2004; Montgomery, Evans and Cooper 2007; Sillen 1998). In the case of bedrock and regolith, this necessitates leaching the samples in a way that mimics the bioavailable component of their isotopic composition (eg. Capo et al. 1998).

Strontium Isotope Mapping in Israel

A comprehensive map of the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values of soil and rock from the principal geological units of Israel has been created (Moffat 2013). The results show that the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values of Israel cluster into four broad domains: the carbonate and siliciclastic province covering most of the country has a $^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.7073–0.7101, basalts and other basic volcanic rocks predominately exposed in northern Israel have a $^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.7052–0.7072, the metamorphic and igneous lithologies in the Eliat region to the south range in $^{87}\text{Sr}/^{86}\text{Sr}$ values from 0.7084–0.7407 and areas with no bedrock (particularly on the coastal plain) have a $^{87}\text{Sr}/^{86}\text{Sr}$ range of 0.7078–0.7147.

A number of other studies have mapped bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ in Israel, including Hartman and Richards (2014), Herut, Starinsky and Katz (1993), Perry et al. (2008; 2009), Rosenthal, Katz and Tchernov (1989), Shewan (2004) and Spiro et al. (2011), which show broadly similar results to those described above.

The Samples

Seven human teeth, which are a subset of those described in Faerman and Smith (this volume), were used in this study. All teeth were from adult individuals with fully formed crowns. The samples included one incisor (M23), four canines (M24, M25, M26, M28) and two molars (M27 and M32). The teeth generally appear heavily weathered.

Methods

LA-ICPMS data were collected using an ESI NW213 laser ablation system coupled to an Agilent quadrupole 7700. Two sets of ablation tracks were collected on each sample after a preablation run to remove potential surface contamination. Dentine and enamel were analysed in the same run when possible. Measurements were undertaken using a spot size of 100 μm at 80% intensity and using a 20 $\mu\text{m/s}$ stage rate movement under a mix He and Ar gas flow. The standards NIST 611 and NIST 613 were analysed at the beginning and end of each analysis to allow drift correction and calculation of baseline and concentration values. Dentine from a fossil hippo calibrated by MC-ICPMS measurements was used to estimate the matrix effect.

LA-MC-ICPMS data were collected with a Finnigan Neptune with acquisition parameters optimised using a piece of modern *Tridacna* shell. Sampling was undertaken in discrete spots of 233 μm to increase the spatial accuracy of the results, despite the possibility that the creation of pits can lead to decreased intensity and increased fractionation (Ramos et al. 2004). The analysis spots were collected in transects which crossed enamel and dentine on each tooth, as shown in Fig. 4.25. LA-MC-ICPMS data were corrected offline by applying a 'gas blank' Kr correction, a mass bias correction, a rare earth element correction and a rubidium correction (in that order). This analysis used relative element abundances based on Berglund and Wieser (2011) and relative atomic masses based on Audi, Wapstra and Thibault (2003).

An additional correction was used to deal with an offset thought to exist between solution and laser values from the same sample due to the effect of a Ca+P+O or Ar+P+O (Willmes et al. 2016) polyatomic interference on mass 87. The effect of Ar+P+O (Willmes et al. 2016) of this offset is thought to increase, apparently making laser values progressively more

radiogenic, with decreasing strontium concentration (Copeland et al. 2008; Horstwood, Evans and Montgomery 2008). To correct for this effect, a number of human and faunal tooth samples were examined by both solution and laser analysis at the Australian National University by Boel (2011), Kelly (2007) and Lees (2010). These values were used to define a relationship between solution/ laser offset and strontium concentration, shown in Fig. 4.26. The average strontium concentration values obtained using LA-ICPMS for dentine and enamel tissues in this study were used to correct the strontium isotope values.

The accuracy of LA-MC-ICPMS results were evaluated by comparison to values from a *Tridacna* clam shell, which is a calcite with a homogenous $^{87}\text{Sr}/^{86}\text{Sr}$ composition that corresponds to modern seawater at 0.70918. LA-MCICPMS data were also evaluated with reference to the $^{84}\text{Sr}/^{86}\text{Sr}$ values from each sample, which should correspond to the value of 0.0565 if $^{87}\text{Sr}/^{86}\text{Sr}$ values are robust.

Fig. 4.25: Teeth following LA-MC-ICPMS analysis showing analysis tracks: a) M23; b) M24 c) M25 d) M26 e) M27 f) M28 g) M32.

Results (Table 4.8, Table 4.9)

The concentration of strontium in enamel in this study is within the normal range suggested by Evans et al. (2012: 756). The values for M23, M24 and M27 are quite low and suggest that LA-MCICPMS analysis for $^{87}\text{Sr}/^{86}\text{Sr}$ values may be problematic. The concentration of strontium in dentine in these teeth is generally high, suggesting that the LA-MC-ICPMS values should be robust. The low concentration of uranium and thorium measured in all of the teeth in this study suggests that they have not been subject to post-burial diagenesis (e.g., Eggins et al. 2005).

The $^{87}\text{Sr}/^{86}\text{Sr}$ results of dentine (with the exception of M25) are within 2σ error of each other and have a weighted average of 0.70782 ± 0.00007 which corresponds to bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ mapping results from a number of Israeli carbonate limestone units. The host geology of the site is somewhat unclear as it is reported by Ackermann and Bruins (2012:123) as belonging to the Zor'a Formation from the Avedat Group, while Sneh, Bartov and Rosenshaft (1998) regard this location as belonging to the Maresha Formation. Regardless of which unit actually outcrops at the site, both are Lower-Middle Eocene chalk or limestone units and so should have a similar strontium isotope value. A $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.70784 ± 0.0001 , which was obtained from a rock sample from the Avedat group (Moffat 2013), corresponds well with the dentine value obtained from the teeth in this study. The correspondence of the dentine to the geological unit containing this site suggests either that the dentine has been subject to post-burial diagenesis or that these individuals are local to the area. As discussed above, the generally low levels of uranium and thorium measured in these samples supports the hypothesis that post-burial diagenesis may not have been extensive.

Fig. 4.26: Relationship between Strontium concentration and $^{87}\text{Sr}/^{86}\text{Sr}$ laser/solution offset used for correction of LA-MC-ICPMS Data in this study.

Fig. 4.27: LA-MC-ICPMS $^{87}\text{Sr}/^{86}\text{Sr}$ Results from Human Teeth from Tell es-Safi/Gath. Mapping values from the local geology are indicated by the horizontal line on the plot.

Table 4.8: LA-ICPMS Results from Human Teeth from Tell es-Safi/Gath.

Sample	Material	Sr (ppm)	Ba (ppm)	Th (ppm)	U (ppm)
M23	Enamel	58.2	3.3	<0.1	<0.1
M23	Dentine	318.2	83.5	<0.1	<0.1
M24	Enamel	43.9	1.7	<0.1	<0.1
M24	Dentine	269.1	61.5	<0.1	<0.1
M25	Enamel	145.6	6.3	<0.1	<0.1
M25	Dentine	413.8	55.5	<0.1	0.3
M26	Enamel	91.4	3.9	<0.1	<0.1
M26	Dentine	308.9	76.6	<0.1	0.8
M27	Enamel	62.1	1.7	<0.1	<0.1
M27	Dentine	248.4	21.8	<0.1	<0.1
M28	Enamel	229.2	8.1	<0.1	<0.1
M28	Dentine	352.5	83.3	<0.1	0.7
M32	Enamel	146.1	22.4	<0.1	<0.1
M32	Dentine	324.4	137.4	<0.1	0.8

Table 4.9: LA-MC-ICPMS $^{87}\text{Sr}/^{86}\text{Sr}$ Results from Human Teeth from Tell es-Safi/Gath.

Sample	Enamel $^{87}\text{Sr}/^{86}\text{Sr}$	2 σ	Dentine $^{87}\text{Sr}/^{86}\text{Sr}$	2 σ
M23	0.7016	0.0007	0.7077	0.0001
M24	0.7037	0.0063	0.70778	0.00002
M25	0.7078	0.0001	0.70642	0.00005
M26	0.7081	0.0032	0.7079	0.0001
M27	0.7007	0.0016	0.7079	0.00008
M28	0.7080	0.0008	0.70786	0.00003
M32	0.7079	0.0004	0.7080	0.0001

The $^{87}\text{Sr}/^{86}\text{Sr}$ results from the enamel of the Tell es-Safi/Gath samples cluster in two domains (as shown in Fig. 4.27): specimens M23 and M27 have results within 2σ error of each other with a weighted average of 0.7015 ± 0.0013 while specimens M25, M26, M28 and M32 also have results within 2σ error of each other with a weighted average of $0.7078 \pm$

0.0003. M24 has a very large 2σ error, which crosses both domains, making it impossible to determine the provenance of this sample. The $^{87}\text{Sr}/^{86}\text{Sr}$ value of the domain containing M23 and M27 is significantly less radiogenic than any value obtained from geological units in Israel and thus cannot be correlated to any location. This, as well as the very high 2σ error values for M24, can be explained by the low strontium concentration values obtained for these sample (43.9–62.1 ppm). At these concentrations, the correction applied to the $^{87}\text{Sr}/^{86}\text{Sr}$ results to correct for the Ca+P+O interference is extreme, thus the results should be treated with caution. In contrast, the results from M25, M26, M28 and M32, which have higher strontium concentrations, can be considered more robust. All of these samples have results, which correlate with the bioavailable $^{87}\text{Sr}/^{86}\text{Sr}$ values obtained for the Lower-Middle Eocene unit, in which Tell es- Safi/Gath is located, suggesting that these individuals were local to the area when their tooth enamel formed.

Conclusions

Seven human teeth from Cave T1 were analysed for strontium isotopes to determine the extent of mobility of the people who were interred there. For four of these teeth, robust results were obtained, which showed an excellent correspondence between the $^{87}\text{Sr}/^{86}\text{Sr}$ results for the enamel, dentine and local geology, suggesting that these individuals inhabited the site and/or its immediate surroundings at an age when their teeth formed. For the remaining three teeth, no robust $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios could be obtained for the enamel, however, their dentine values, which correspond to the local site, might indicate that these were also local to the area.

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