Transhumance pastoralism of Roccapelago (Modena, Italy) early-modern individuals: Inferences from Sr isotopes of hair strands

# Transhumance pastoralism of Roccapelago (Modena, Italy) Early-Modern individuals: inferences from Sr isotopes of hair strands

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# Abstract

**Objectives**

In this work, we use Sr isotopes to analyze human hair and determine short-term movements of a contemporary human traveler and of Early-Modern individuals from an archaeological site (Roccapelago, Modena, Italy, $16^{th}$-$18^{th}$ century).

**Materials and Methods**

Analyses were performed using a Neptune MC-ICP-MS. We first set up and tested the procedure on scalp hair of a contemporary human, who spent some time between Brazil and Italy. We then analyzed the $^{87}Sr/^{86}Sr$ ratios of 8 exceptionally well-preserved archaeological hair specimens associated with human mummies from Roccapelago. Trace elements were analyzed by LA-ICP-MS on single-hair specimens to check the preservation of the archaeological samples.

**Results**

The strontium isotope composition of modern human hair varies from 0.7087 to 0.7093. The $^{87}Sr/^{86}Sr$ ratios of the Roccapelago hair strands range from 0.7082 to 0.7137, with an average of 0.7093 ± 0.0031 (2σ), revealing also intra-individual differences in isotopic composition along the length of the hair shaft. Data were compared with local archaeological rodent bones and teeth and with published isotopic values of water and outcropping rocks. Trace element abundances of archaeological and modern human hair are similarly low in terms of Rare Earth Element (REE) and metal contents, in particular after $HNO_3$ leaching.

**Discussion**

The variable modern human hair $^{87}Sr/^{86}Sr$ ratios show that high-resolution hair sampling tracks the movements of this individual between the two continents. The Sr isotope composition of the mummy hair is consistent with sub-annual human movements from Roccapelago to an area with different $^{87}Sr/^{86}Sr$ ratios. Historical sources indicate that individuals from Roccapelago travelled with their herds to Tuscany for transhumance pastoralism practices. The high radiogenic $^{87}Sr/^{86}Sr$ ratios (> 0.71) found in some of the hair are possibly consistent with the Tuscan Magmatic Province and the Tuscan Metamorphic Complex isotope signature. To our knowledge, this is the first study in an archaeological context where the Sr isotope evidence of mobility is corroborated by historical documents.

## 1. Introduction

Over the last few decades, strontium isotopes have been established as a powerful tool to investigate the provenance of ancient and modern materials. In particular, their application in archaeology and paleontology has helped decipher how artefacts have moved between communities (e.g. Freestone et al. 2003; Degryse et al. 2010) and track patterns of mobility in humans and animals (e.g. Ericson 1985; Grupe et al. 1997; Bentley et al. 2002; Price et al. 2002; Evans et al. 2006; Bentley et al. 2012; Sorrentino et al. 2018;e.g. Pellegrini et al., 2008; Montgomery et al. 2010; Britton et al. 2011; Lugli et al. 2017a). More recently, in forensic contexts, strontium isotopes of hair strands have documented the movements of unknown bodies and possible immigrants (Font et al. 2012; Tipple et al. 2013; Vautour et al. 2015). The study of Sr isotopes from archaeological human hair is instead very scarce (Font et al. 2012; Frei et al. 2015; Frei et al., 2017), mainly because of the rarity of hair remains in archaeological contexts and the analytical difficulties related to the low-Sr concentration of human hair (from ~0.5 to ~10 ppm).

Mammal hair is formed of a scleroprotein, namely keratin, composed of several major elements (C, N, H, O, S), but also including trace elements such as Sr, Pb, Fe, K, Na and Ca (Font et al. 2012). Trace elements are fixed within the hair through diet and exposure to exogenous sources, mainly water and air. Given that the average human scalp hair growth is ~1 cm/month (0.35 mm/day on average; ranging between ~0.3 and ~0.5 mm/day; Krause and Foitzik 2006), time-resolved hair sampling can provide temporal information, mainly reflecting the elemental and isotopic composition of the blood at a certain time, given that blood vessels feed the roots in the scalp. In this sense, hair samples are a powerful biological archive of information about short-term human movements.

Strontium isotope ratios ($^{87}Sr/^{86}Sr$) of mammal tissues are generally directly correlated with the Sr isotope composition of the biologically available Sr (Bentley 2006). Since the bioavailable Sr is incorporated in hair mainly through diet and drinking water, the Sr isotopic composition of hair is linked to the geographical provenance of food sources eaten by the individual. However, one of the main problems with hair is contamination by Sr from external sources. These include both pre-burial (e.g. environmental exposure and cosmetic treatments; Chau et al. 2017) and post-burial sources (e.g. diagenetic contamination; von Holstein et al. 2015). To date, it is not possible to discriminate the endogenous from the exogenous Sr, and measured $^{87}Sr/^{86}Sr$ ratios result from a mix of both. Several hair-washing methods have been tested in the literature to try to remove the exogenous Sr portion from the keratin samples, but without a clear and unequivocal result. However, previous studies have demonstrated that even combining both Sr sources (endogenous plus exogenous), isotopic analyses of hair samples can be helpful to define geographical movements of individuals

(Vautour et al. 2015; Frei et al. 2015). In this view, modern forensic cases can be difficult to interpret given that the globalization of the economy has made food and water produced in certain regions available even on different continents and they contribute to the Sr budget of human body (Vautour et al. 2015). Contrariwise, in most of the archeological contexts, given the null or lower effect of globalization in terms of food consumption, the relation between Sr isotope and human provenance and mobility is more direct. However, such ancient contexts still present many issues in terms of data interpretation, mainly because of the possible post-burial contaminations and the common lack of direct comparisons to check the reliability of the interpretation itself.

A unique opportunity to test the application of Sr isotopes to human hair to distinguish mobility patterns has been given to us by the discovery of a church crypt in the village of Roccapelago (Modena, Italy) with partially mummified bodies of early-modern individuals with well-preserved hair samples (Lugli et al. 2017b). Individuals from Roccapelago, as known from historical sources, practiced transhumance pastoralism to Tuscany (Fig. 1), a form of pastoralism and social organization, where herds, driven by shepherds, move long distances generally twice per year in search of new fertile pastures (Chang 1993; Cazzola 1993; Traversari 2016). More than 60 partially mummified individuals where discovered in 2011 and dated between the 16$^{th}$ and the 18$^{th}$ century. The study of *liber mortis* (16$^{th}$-20$^{th}$ cent.) from Roccapelago revealed a high frequency of human seasonal travels to Tuscany, up to the Grosseto area, linked to the exploitation of transhumance pastoralism (Traversari 2016). Further evidence of the transhumance practice can be found in the seasonality of the deaths, with the highest number of deceased between April and October. In fact, during summer months, the population settled in Roccapelago increased after the return of shepherds from their travels. Similarly, also the frequency of conceptions, calculated using regression equations, increased during the same period of the year, when the number of people in Roccapelago was high (Traversari 2016). During the first decades of the 17$^{th}$ century, movement restrictions were applied to prevent the spread of the plague. Only after 1631, shepherds were able to travel again towards Tuscany, with more than 40,000 ovicaprids moving from the Roccapelago area (Traversari 2016).

Therefore, the Roccapelago site represents a rare opportunity to reconstruct the life and the traditions of a secluded farming community, which practiced animal husbandry and seasonally moved with their flocks towards Tuscany as highlighted in historical documents. In this scenario, the combination of high-resolution human hair sampling of Roccapelago individuals and strontium isotope analyses by dissolution MC-ICP-MS can help us to decrypt travel patterns in these humans and whether the individuals buried within the crypt where in fact shepherds practicing transhumance. This study is also possible because the geology of the surroundings of Roccapelago and of Tuscany are very different, and, consequently, such is also the expected isotopic composition of the bioavailable Sr.

### *1.1. Sr in the human body*

$Sr^{2+}$ enters the organism mainly through diet as $Ca^{2+}$ substitute, due to their similar ionic radii (1.13 Å vs. 0.99 Å respectively; Faure and Mensing 2005). Ca is preferentially assimilated with respect to Sr during biological activities (Burton et al. 1999), because the shift of Sr from the digestive system to the bloodstream is less efficient than that of Ca; thus, only part of the ingested Sr is absorbed (Lugli and Cipriani 2017). Most of the ingested Sr with an average diet (1.9 mg/day) is excreted through urine (0.34 mg/day) and feces (1.5 mg/day) and lost with sweat (0.02 mg); 99% of the gut-absorbed Sr is thus fixed in bones through surface exchanges and ionic substitution (Pors Nielsen 2004). The remaining small portion is fixed within hair keratin and other tissues (Font et al. 2012). Ingested Sr not only derives from solid food but also from drinking water. Depending on the Sr concentration of the water (ranging from several ppb to several ppm) and on the daily intake of water, the ingested portion of Sr can vary a lot. The Agency for Toxic Substances and Disease Registry indicates that, in the United States, on average ~2 mg/day of Sr are ingested through water (ATSDR 2004; Vautour et al. 2015). Considering the Sr concentration of the waters from this study (see next sections) and an average consumption of 2 L per day (World Health Organization standard quantity; Säve-Söderbergh et al. 2018), we can estimate a Sr water intake ranging from 0.04 to 0.5 mg/day.

During life, Sr from bone is continuously removed and transferred to the blood through three main mechanisms: resorption during bone remodeling; surface rapid ionic exchange at the whole bone surface and slow diffuse exchange during which elements penetrate the entire bone volume (Dahl et al. 2001). Thus, Sr in the bloodstream results from a mix of the recently ingested Sr and the old bone Sr reservoir. This is the main reason why the equilibration of the Sr isotope signature in hair may take more than 18 months to reach a new Sr end-member (Font et al. 2012). However, the shift in the isotopic signal within the hair starts almost immediately after the residential change on a monthly scale, as well documented by Font et al. (2012) and Vautour et al. (2015).

### *1.2. Archaeological background*

Roccapelago is a small village located near Pievepelago (Frignano area, Modena Apennines) on the banks of the Scoltenna River. An important building for the Roccapelago community was the church of the Conversion of St Paul, erected originally by Obizzo di Montegarullo at the end of the 14$^{th}$ century as a military fortification. This structure is located on a high peak that dominates the surroundings. In 1585, it became a Christian church,

losing all its military purposes. In 2011, a forgotten crypt was discovered under the church's floor containing a large mass of bodies. These corpses were deposited there between the end of 16$^{th}$ century and 1786, when the current cemetery was built outside the church. The lower bodies were buried in the natural recesses of the floor as they were found covered with a landfill layer few centimeters thick. From this layer, only poorly preserved skeletonized remains were recovered with no preservation of clothes or soft tissues. The upper layers are composed of several well-preserved skeletonized individuals piled one on top of the other, and covered by a layer of rock chunks. The top of the upper layers is made of a pyramidal cumulus of human bodies. After death, individuals were likely dropped through a manhole from the church floor, naturally forming this cumulus (Figus et al. 2017). All bodies from this layer were naturally mummified due to the peculiar environmental condition of the crypt, characterized by low humidity and high aeration (Gruppioni et al. 2010; Lugli et al. 2017b). These individuals were clothed in tunics, hats and heavy socks, some of them wrapped in shrouds and/or bags. Considering the elevated number of skeletonized remains found at the base of the crypt and the partially mummified individuals (ca. 60), we inferred that ca. 400 individuals were buried/deposited in this crypt.

### *1.3. Geological background*

The incorporation of Sr into the body of humans and animals begins with the weathering of the underlying bedrock and continues with the formations of soils and the release of Sr into water (Bentley 2006). Therefore, the local geology defines the Sr isotopic composition that individuals and animals adsorb in their hair during growth. Given that from historical sources we know that Roccapelago individuals moved from their village to Tuscany, we briefly review the geology of the two localities.

Roccapelago is located in the northern Apennine chain, a complex tectonic structure related to the collision of the African with the European plate and the opening of the Western Mediterranean. The rock outcroppings in the Roccapelago area are all of sedimentary origin and range in age from the Oligocene to the Miocene, with a limited area of Cretaceous clays. These piles of sediments are deposited directly from seawater (carbonates) or are sediments derived from the weathering of continental rocks. We can predict the isotopic composition of marine sediments from the seawater curve of McArthur et al. (2001). For the Roccapelago area, we expect therefore the local bioavailable Sr to range between 0.7078 (Oligocene lower limit) and 0.7090 (Miocene upper limit).

The geology of Tuscany is characterized by marine sedimentary sequences that mostly consist of carbonates, clays and sandstones, ranging in age from the Triassic to the Pliocene (Fig. 2). However, a striking difference

with the geology of the surroundings of Roccapelago is the large presence of young volcanic rocks (Tuscany Magmatic Province) and an outcropping of metamorphic crystalline basement (Tuscan Metamorphic Complex). Both volcanic and metamorphic rocks have very distinctively high Sr isotopic compositions with $^{87}Sr/^{86}Sr$ ratios ranging from 0.710 to 0.725 and from 0.714 to 0.753, respectively (Hawkesworth and Vollmer 1979; Ferrara and Tonarini 1985; Nisi et al. 2008;

### *1.4. The local bioavailable Sr*

In provenance studies of an archaeological sample, Sr isotope data need to be compared with those of the local bio-available Sr. The identification of the local isotopic signature is not always an easy task. In fact, several sources of Sr (e.g. weathered rocks, rainwater and atmospheric deposition, rivers, underground streams) can contribute to the isotope composition of water and soil, which are the main reservoirs of the local bioavailable strontium (Bentley 2006).

In this paper, we use literature Sr isotope ratios of rocks and waters from Roccapelago and Tuscany for comparison with our archaeological samples (Holm and Munksgaard 1982; Hawkesworth and Vollmer 1979; Ferrara and Tonarini 1985; Dinelli et al. 1999; Peccerillo and Donati 2003; Boschetti et al. 2005; Nisi et al. 2008; Pennisi et al. 2008; see Supplementary Table 1). Generally, the Sr isotopic ratio of the geological substratum is correlated to that of the plants thriving in the area (Hartman and Richards 2014; Marchionni et al. 2013). For instance, wines from Central and Southern Italy present $^{87}Sr/^{86}Sr$ ratios that correlate well with the geological substratum signature of the relative vines (Marchionni et al. 2013). In addition, waters can be used to determine the local $^{87}Sr/^{86}Sr$ ratio and are generally good predictors of the bioavailable Sr isotope pool (Maurer et al. 2012), being a mix of Sr from atmospheric deposition and mineral weathering processes.

However, even if a local bioavailable isotopic baseline is identified, Sr isotope provenancing is limited by the fact that spatially distant places can present a similar, if not identical, local isotopic composition. This results in a high degree of uncertainty in the identification of a possible non-local isotope signal. Thus, some inferences can be made by integrating geochemical studies with historical information.

### *1.5. Hair diagenesis*

In order for the elemental and isotopic data to be meaningful, they need to be measured in uncontaminated hair specimens. High metal concentrations in hair have been interpreted as due to diagenetic enrichment in wet

environments (see e.g. Kempson et al. 2010). Since metal ions are positively charged, they have a high affinity for negatively charged melanin polymers (Morton et al. 2002; Frei et al. 2009). In fact, it has been shown that buried wool textile may present exceptionally high Sr concentration for this reason (von Holstein et al. 2015). In archaeological and paleontological studies of bones, Rare Earth Elements (REEs) are commonly used as a proxy for elemental uptake during diagenetic processes of human and animal remains, being usually present at very low concentration within the living organism and thus mostly coming from post-burial uptake (see e.g. Reynard and Balter 2014). However, REE behavior in terms of hair preservation is not well known. In vivo, REE content of hair has been investigated mainly in relation to environmental exposure in areas characterized by peculiar geological backgrounds (i.e. China mining areas described in Wei et al. 2013), showing that REEs can accumulate in human hair during *in vivo* processes (Rodushkin and Axelsson 2000; Tong et al. 2004). This is especially true for light REE that can reach concentrations of several ppm (Wei et al. 2013). Currently, we have little or no information about the behavior of REE during the post-burial history of hair. However, considering the abovementioned affinity of hair with metal cations absorbed from the environment (Kempson et al. 2010), we could expect a similar affinity for REE trivalent cations.

## 2. Materials and Methods

### *2.1. Modern hair samples*

Sr analytical procedures were tested on modern hair samples before the application on archaeological ones. Hair strands were thus sampled from an anonymous donor, known to be regularly travelling between Brazil (São Paulo) and Italy (Modena), and, therefore, suitable for the time-resolved analysis of hair samples. The donor is an Italian male omnivore and smoker; he used to drink tap and bottled water in Brazil, but mainly bottled water in Italy. He is a heavy coffee drinker. Before sampling, the donor spent five months in Brazil and three in Italy without cutting the hair. Hair was sampled in several portions of different lengths following two different protocols. In the first one (Method 1), several thin hair strands were sampled from different positions of the nape (TGH-S) and cut in sub-samples of 1 cm. Corresponding samples, related by the distance from the scalp, were put together to obtain enough material for the analysis. This was necessary due to the low-Sr content of human hair (~0.5 to ~10 ppm). In the second protocol (Method 2), a single thick and long hair strand (10 cm) was cut from the upper portion of the head and sub-sampled in 2-cm-long portions (TGH-L).

All resulting samples (n = 12) weighed approximately 20 to 50 mg.

### *2.2. Archaeological hair samples*

Archaeological hair specimens were collected from combs and clothes within the crypt in Roccapelago. All available archaeological hairs were collected and analyzed for this work, for a total of 8 individuals. None of the hair samples was covered with soil, in fact they were collected from the uppermost layers nearby the mummified bodies. No hair was preserved on the scalps of the mummies, thus it was not possible to collect samples directly from the head. Therefore, the distance from the hair tip to the scalp itself is unknown, and we were not able to determine the root or the tip of the strand, e.g. the younger and the older portion of the strand. However, even though we do not know the temporal direction of the hair archive, we still can collect useful time-resolved information. Specimens ROC-US23-1 and ROC-US26-2 were sampled, respectively, from a braid with a feminine metallic hair clip and from a hair strand within a bonnet. These are the only two samples that we could attribute the sex to with certainty.

Eight hair strands were cut into several sections (n = 14), to obtain the minimum sample weight required to run the analysis (~20 mg). The maximum time resolution achieved for a single hair strand (ROC-US26-1) subdivided in four portions is of 3 months (= 3 cm samples), thus representing one year of life of the individual. Because of the low amount of material, three hair strands (ROC-T9, ROC-US23-1 and ROC-US23-2) were not split and thus analyzed in bulk. As previously suggested by others, hair samples from archaeological contexts may retain a primary isotopic signature (Font et al. 2012; Frei et al. 2015). Moreover, as reported by Lugli et al. (2017b), human osteological samples from the Roccapelago crypt show little diagenetic alterations having been piled up in a crypt and not in the soil, and protected from rainwater. However, to check for possible post-depositional contaminations, we analyzed leachates from the cleaning protocol of the hair samples. Sequential leaching should result in a progressive cleaning of the hair from macro and microscopic particles of soil or dust and from the exogenous portion of Sr exchanged by those sites, within the hair chemical structure, more susceptible to alteration.

### *2.3. Landscape Sr isotopic composition*

To understand if an individual has moved through the landscape it is necessary to compare the hair Sr isotope composition to that of one of the possible localities visited by the individual. We can infer the local isotope signature by measuring the $^{87}Sr/^{86}Sr$ ratios in animals with a small home range (e.g. are considered to have always lived locally) and/or in the local drinking water. The former approach is generally used in archaeological contexts, the latter in modern case studies (Bentley 2006). Therefore, tap and bottled waters from Brazil (n = 3)

and from Italy (n = 3) were collected to compare them to our modern hair donor Sr data. Waters were selected based on the individual dietary habits. Sr in waters was concentrated evaporating ca. 50 mL of the sample. For the archeological case study of Roccapelago we determined the isotopic baseline by measuring the Sr isotopic composition in rodent bones and teeth (n = 10) sampled directly from the Roccapelago crypt.

### *2.4. Sample cleaning and digestion*

Ultrapure MilliQ® water (~18.2 MΩ resistivity) from a Millipore (Merck) system was used during the cleaning treatment and to dilute the concentrated acids. All acids employed in this study were of Suprapur® grade. Concentrated $HNO_3$, HCl, chloroform (HPLC grade) and methanol (HPLC grade) were bought from Sigma-Aldrich (St. Louis, Missouri, US). The entire analytical procedure was conducted in a class 1000 clean lab, under a laminar flow hood with Sr blanks typically lower than 100 pg.

Modern and archaeological hair samples were treated and dissolved following the protocols of Font et al. (2012). Each modern hair sample was sonicated for 10 minutes in a 2:1 chloroform:methanol mixture and rinsed with MilliQ® water. This step was repeated three times. Samples were then left to dry at room temperature. Archaeological hair samples were first treated to remove the dust and dirt fraction mixed with the hair strands and sonicated for 5 minutes with MilliQ® water. This step was repeated until the water was completely clear. For our samples, more than five washes were generally required. Then, samples were leached with 2 M $HNO_3$ for 30 minutes. Finally, samples were rinsed three times with MilliQ® water. Both archaeological and modern hair samples were dissolved in a Teflon beaker at 110°C for 24 h with 2 mL of *aqua regia* (1.5 mL of 14 M $HNO_3$ plus 0.5 mL of 8 M HCl) and left to evaporate. The residue was taken up in 1 mL of 14 M $HNO_3$, left on the hot plate at 110°C for 24 h, and finally dried down. At the end, to oxidize the remaining organic matter, the sample was fluxed with 100 µL of 14 M $HNO_3$ and 100 µL of ultrapure $H_20_2$, on a hot plate at 105 °C, with the beaker open. This process was repeated until all organic matter was digested.

Leachates from the MilliQ® leaching step were treated with HF to digest all the sediment washed from the hair specimens, dried down and then adjusted to 3 M $HNO_3$. Leachates from the 2M $HNO_3$ water were instead directly adjusted to 3 M $HNO_3$. Archaeological rodent samples were treated following the protocol described in Lugli et al. (2017a). Samples were sonicated 3 times with MilliQ® water (20 min per wash) and leached with 1 mL of 1.5 M $HNO_3$ for 30 min to remove surface contaminants. Dissolution was performed using 2 mL of 6 M $HNO_3$.

All dissolved samples were then taken up in 3 mL of 3 M $HNO_3$ for column chromatography.

### *2.5. Column chromatography*

Ion-exchange chromatography was performed on hair samples following standard procedures (Weber et al. 2018). The Sr separation uses columns with a 300 µl volume filled with Eichrom Sr spec–resin (100-150 µm bead size). Resin was first cleaned with MilliQ® water and floating particles were pipetted out after settling. After filling, the columns were preflushed with 1 ml of 3 M $HNO_3$, washed three times with MilliQ® water (1 mL each) and conditioned with 1 ml of 3 M $HNO_3$. Samples (3 mL) were then loaded into the columns. Matrix ions were removed with the stepwise addition of 3 M $HNO_3$ (3 mL overall). Sr was then eluted with MilliQ® water (5 steps, 0.5 mL per step) and collected in clean Teflon beakers. Each solution was then adjusted to 4% w/w $HNO_3$ for MC-ICP-MS analysis.

### *2.6. MC-ICP-MS analysis*

The $^{87}Sr/^{86}Sr$ ratio of the samples was determined using a double focusing MC–ICP–MS with a forward Nier–Johnson geometry (Thermo Fisher Scientific, Neptune™) housed at the Centro Interdipartimentale Grandi Strumenti (CIGS) of the University of Modena and Reggio Emilia (Modena, Italy). Seven Faraday detectors were used to collect signals of the following masses: $^{82}Kr$, $^{83}Kr$, $^{84}Sr$, $^{85}Rb$, $^{86}Sr$, $^{87}Sr$, $^{88}Sr$. The $10^{12}$ Ω resistors were employed for the two Kr masses and for $^{84}Sr$ and the $10^{11}$ Ω resistors for the remaining masses. For low-Sr concentration specimens, solutions were introduced into the Neptune via a high-sensitivity desolvating inlet system (ESI-Apex IR) and a 100 µl/min nebulizer. For higher concentration samples, a quartz spray chamber was employed. Samples, standards and blanks were analysed in a static multi–collection mode and single blocks of 100 cycles, with an integration time of 8.4 s per cycle. To monitor and correct possible drifts of the instrument we employed a standard-sample-standard bracketing sequence.

The presence of Kr in the argon was monitored collecting masses 82 and 83. Data were corrected using a $^{86}Kr/^{83}Kr$ ratio of 1.505657. Mass 85 was used to correct the signal on mass 87 for the presence of isobaric Rb, using a $^{87}Rb/^{85}Rb$ ratio of 0.3856656.

Mass bias normalization was performed through exponential law according to Eq. 1 and 2:

$$(^{87}Sr/^{86}Sr)_{norm} = (^{87}Sr/^{86}Sr)_{meas} / [(^{88}Sr/^{86}Sr)_{meas} / (^{88}Sr/^{86}Sr)_{true}]^a \quad (1)$$

$$a = \ln[M_{87Sr}/M_{86Sr}] / \ln[M_{88Sr}/M_{86Sr}] \quad (2)$$

where 'meas' refers to the measured un-normalized ratio; $(^{88}Sr/^{86}Sr)_{true}$ is the IUPAC $^{88}Sr/^{86}Sr$ ratio (8.375209; Berglund and Wieser 2011) and 'M' are the isotope atomic masses.

Mass fractionation for both Kr and Rb has been assumed equal to that one of Sr. The $^{87}Sr/^{86}Sr$ ratios were corrected for instrumental bias to an NIST SRM 987 value of 0.71026 ± 0.00002 reported by Ehrlich et al. (2004). Repeated analyses of the NIST-SRM-987 diluted to 30 ppb yielded a $^{87}Sr/^{86}Sr$ ratio of 0.71024 ± 0.00003 (2σ).

Following Copeland et al. (2010) and Lugli et al. (2017), we obtained the Sr concentration of our samples by interpolating the solution Sr concentration (ppm) from the signal intensities (V), using materials with known Sr content. This method yielded results that are a reliable approximation, in terms of accuracy, of the true Sr concentration, as observed for our known water samples.

### *2.7. LA-ICP-MS analysis*

Three archaeological hair samples (ROC-T9, ROC-US26-2; ROC-US28-2) were analyzed by LA-ICP-MS (Sforna & Lugli 2017) to check for possible post-depositional uptake of trace elements. Samples were selected based on Sr isotope results, covering the broadest possible interval of both Sr contents and isotopic ratios. One of the contemporary hair specimens from the TGH-L strand (here simply renamed TGH) was analyzed as a baseline for the elemental content of hair not affected by post-burial diagenesis. This sample was washed with MilliQ® water and the 2:1 chloroform:methanol mixture. Moreover, to check whether our cleaning protocol had an effect on the hair elemental content, we analyzed two sets of samples: one after a simple MilliQ® ultrasonic cleaning (samples labelled as 'W') and a second one after the 2M $HNO_3$ leaching step (samples labelled as 'Ac'). Then, a single hair per strand was glued on a Teflon disk using high-purity carbon tape. In situ analyses were run on a quadrupole ICP-MS (X-Series$^{II}$, Thermo Fisher Scientific) coupled to a 213 nm Nd:YAG laser ablation system (New Wave Up) housed at the CIGS UNIMORE (see Giovanardi et al., 2018), employing 1500 µm-long linear scans with a spot size of 30 µm, a scan speed of 10 µm/s and a laser energy output of ~8 J/cm$^2$. Elemental intensities were normalized to $^{34}S$. We quantified the absolute elemental concentrations (ppm) of the hair specimens using a pressed pellet of CAAS Sulphide-Ore 1 as reference material (Sine et al. 1969). The sulfide was selected because of its S content (~12%), similar to that of human hair (~5%). However, given the non-matrix matched nature of this standard, we build our interpretation upon the elemental normalized intensities and not the absolute values. Nonetheless, trace element concentrations are reported within the Supporting Information.

## 3. Results

Modern hair samples $^{87}Sr/^{86}Sr$ ratios range from 0.7087 to 0.7093 (Table 1), with an average of 0.7089 ± 0.0004 (2σ). In Figure 5, we plot the relative temporal variation of the $^{87}Sr/^{86}Sr$ ratios of the two strands and observe that the roots of both strands have a common root value of 0.7087 and increase to higher ratios towards the tip. The temporal variations in the two strands deviate at about 4-5 cm from the root and while the longer strand reaches values of 0.7093 at the tip, the shorter strand plateaus at about 0.7089.

In terms of Sr concentration, modern hair samples range from 2 to 11 ppm. The TGH-L long hair strand shows a decrease in Sr content from the tip (11 ppm) to the root (2 ppm). In addition, the Sr content of the TGH-S short hair strand tends to decrease from the tip (8 ppm) to the root (4 ppm), when excluding sample C with a low Sr concentration (2 ppm). Moreover, Sr isotope ratios of the TGH-L strand (Fig. 6) are negatively correlated with the inverse of the Sr concentration ($^{87}Sr/^{86}Sr$ vs. 1/Sr; $r^2 = 0.79$; $n = 5$; $p < 0.05$) in contrast to what we observe for our sampled Brazilian and Italian waters that are positively correlated ($^{87}Sr/^{86}Sr$ vs. 1/Sr; $r^2 = 0.69$; $n = 6$; $p < 0.05$).

LA-ICP-MS elemental results of human hair are shown in Figure 7 and reported within the Supporting Information (Supplementary Tables 2, 3 and 4). Rare earth elements (REE) show very low intensities in both contemporary and archaeological samples, generally near or below the instrumental detection limit. After the nitric acid leaching, total average REE loss is ~69% (see Table 4 and Supplementary Table 5). Sr and Pb before the nitric acid step are higher in archaeological samples than in the contemporary hair specimen. In particular, Sr is up to 4 times higher, while Pb up to 16 (see Supplementary Table 4). After the nitric acid leaching, average Sr loss is ~89% and average Pb loss is ~96% (see Table 4), resulting in Sr and Pb contents lower or equal to the contemporary hair. A similar behavior is observed also for other elements, such as Ca, Cu, Zn and Ba, reported in the literature as possible post-burial contaminants of hair (e.g. Kempson et al. 2010; Trunova et al. 2015). Remarkably, the Rb concentration of all the archaeological samples does not change significantly after the $HNO_3$ leaching, always showing a content comparable to the contemporary hair. Instead, the Hg content is lower than in the contemporary hair both before and after the $HNO_3$ leaching (see Supplementary Figure 4). Sample ROC-US26-2 shows a very high increase of Ce and Ta contents after leaching (~1000%; Supplementary Table 5): however, given that both elements have very low intensities, close to background levels, such increase can easily result from a mathematical artefact during the normalization.

Archaeological hair $^{87}Sr/^{86}Sr$ ratios of leachates are reported in Table 2 and range between 0.7085 and 0.7089, with an average of 0.7086 ± 0.0002 (2σ). The local bio-available Sr for Roccapelago was obtained by analyzing rodent bones and teeth from the crypt (Table 3), yielding a $^{87}Sr/^{86}Sr$ ratio of 0.7086 ± 0.0001 (2σ). The Sr isotope

ratios of the leachates are statistically indistinguishable from the local rodent $^{87}Sr/^{86}Sr$ ratios (two-tailed Mann-Whitney *U*-test; $p = 0.80$; Supplementary Figure 2).

The Roccapelago archaeological hair samples $^{87}Sr/^{86}Sr$ ratios are presented in Table 1 and range from 0.7082 to 0.7137, with an average of 0.7093 ± 0.0031 (2σ). Four samples, ROC-US26-1A and B and ROC-US28-2A and B, are characterized by very radiogenic $^{87}Sr/^{86}Sr$ ratios, quite different from the other hair values and from the local-rodent values. For three hair strands (ROC-US26-2; ROC-US28-1 and ROC-US28-4), the intra-individual isotopic variability is very limited, with the highest difference between the most and least radiogenic ratio of 0.0003 for specimen ROC-US28-1 (from 0.7086 to 0.7083). The highest intra-individual isotopic variability is observed in samples ROC-US28-2A (0.7137) and ROC-US28-2B (0.7107), with a difference of 0.0030. In addition, the hair strand labeled as ROC-US26-1 is characterized by a large isotopic variability, with a maximum intra-difference of 0.0023.

## 4. Discussion

We successfully measured the Sr concentration and Sr isotopic composition of both archaeological and modern human hair samples. We first discuss the results of the modern hair in the context of the water supply available to our human case study and the travels accomplished. We then evaluate the diagenetic status of our archaeological samples based on leachate Sr isotope analyses and trace element hair analyses. Finally, the archaeological Roccapelago hair data and the historical sources associated with this case study are also discussed.

### *4.1. Modern human hair*

In figure 5, we present the Sr isotope composition of two modern human hair strands plotted with time. The $^{87}Sr/^{86}Sr$ ratios show some differences in their absolute values, especially in the older portion, likely related to the sampling method. In fact, while Method 1 (short strand) samples yielded $^{87}Sr/^{86}Sr$ ratios between 0.7087 and 0.7090, Method 2 (long strand) samples range from 0.7087 to 0.7093. The more averaged values observed in the tip portions of Method 1, could arise from the fact that several short hair strands were put together to form these samples. Even if corresponding sub-specimens of the different hair strands were used, the error in the manual split of the hair can easily bias the resulting $^{87}Sr/^{86}Sr$ ratio from the 'real' month-value to an average

value. Given that the hair sub-sampling started from the hair root to the tip, more averaged values are expected for the older portions, according to what we observe in our data.

Nevertheless, both hair strands show a similar profile (Fig. 5), with values varying between the high $^{87}Sr/^{86}Sr$ ratios of the São Paulo area, also observed in the Brazilian waters (0.7147 - 0.7278), and the low Sr isotope ratios of the Modena and Reggio Emilia areas (water ranging from 0.7083 to 0.7086; Table 3). Notably, the highest Sr isotope ratios observed in the hair do not reach isotopic equilibrium with the Brazilian end-member. This is likely due to the fact that hair tissue can take more than 18 months to reach the new isotopic end-member because of the buffer of old Sr from the bone reservoir (Font et al. 2012; Vautour et al. 2015). Being the donor of Italian origin, the bone should retain a low Sr isotope signal, similar to the Italian end-member derived from drinking water (~0.708). Moreover, the $^{87}Sr/^{86}Sr$ ratio of hair tissue is the result of several inputs (e.g. diet, atmospheric pollution and modern contaminants), thus, it is difficult to detect the exact Sr isotope end-member. Very little data are reported for Brazilian environmental samples (Rummel et al. 2010). Rummel et al. (2010) report $^{87}Sr/^{86}Sr$ ratios of Brazil orange juices, ranging between 0.7075 and 0.7188 because of the highly heterogeneous lithology of the bedrock (from basalts to old metamorphic rocks) and thus attesting for a great variability of the Brazilian environmental Sr isotope composition.

Considering an average human hair growth rate of ~0.35 mm/day, the TGH results do not exactly agree with the movements of the individual. In fact, the decrease of the $^{87}Sr/^{86}Sr$ ratio to the less radiogenic end-member seems to anticipate the travel to Italy (Supplementary Figure 1). On the contrary, assuming a slightly faster (~+30%) hair growth of ~0.5 mm/day (Fig. 5), common in caffeine users (Fischer et al., 2007), the trend of the $^{87}Sr/^{86}Sr$ ratio better fits the human movements, with the start of the decrease almost coinciding with the travel time to Italy (around June/July 2016). Another possible explanation for this discrepancy can be found in growth cycle errors, due to the presence of inactive hair within the sampled hair strand (telogen phase; Schwertl et al. 2003). In fact, an average hair strand contains about 85-90% of active hair and 10-15% of inactive hair, formed at different times (Schwertl et al. 2003; Williams et al. 2011). A telogen hair does not contain recently formed hair tissue, thus missing recent isotopic information (Schwertl et al. 2003). As showed by Schwertl et al. (2003) and Williams et al. (2011), mixed hair specimens, with both telogen (inactive) and anagen (active) hair, present a “delay” in terms of stable isotope signal with the respect to anagen-only samples. Conversely, our contemporary case study seems to anticipate the predicted isotopic signal, presenting “younger” ratios than what we should expect, and thus suggesting that the issue is more likely related to an incorrectly assumed hair growth.

The root to tip Sr concentration profiles of the modern hair strand do not follow the Sr concentration of the waters consumed by the donor. The Sr isotope ratios of the TGH-L strand are negatively correlated with the inverse of the Sr concentration while the sampled Brazilian and Italian waters are positively correlated. In fact, Italian waters seem to be characterized by higher Sr content (0.07 – 0.23 ppm) when compared to Brazilian waters (0.02 – 0.06 ppm). This evidence agrees with the conclusions drawn by Vautour et al. (2015), suggesting that hair strands become more concentrated in Sr (and Pb) from the root (younger) to the tip (older), with no relation to the Sr-content of the water end-member (Fig. 6). This typical root-tip/low-high Sr concentration profile in hair is due to both exogenous and endogenous sources, where Sr tends to accumulate from ambient exposure with time. Moreover, given the enrichment of the hair tip in terms of exogenous Sr, it is still not clear if this hair portion is indicative of diet and movements or whether it solely represents the isotope composition of the exogenous sources (Chau et al. 2017). The Sr content of human tissues is also complicated by many variables that affect the Sr bioavailability. In particular, given that Sr mimics Ca during physiological processes, but it is discriminated in favor of Ca along the food chain, Sr concentration within a tissue is highly sensitive to the total amount of Ca in the diet (Montgomery 2010).

### *4.2. Diagenetic conditions of archaeological hair specimens*

The diagenetic conditions of our archaeological hair samples is discussed based on LA-ICP-MS analysis of hair and Sr isotope leachate data. All analyzed leachates show $^{87}Sr/^{86}Sr$ ratios indistinguishable from the local bioavailable Sr of the rodent bone/tooth specimens. . This is true also for the Nitric acid leachates from the hair specimens with the highest Sr isotope ratios (ROC-US26-1 A and B and ROC-US28-1 A and B). We cannot undoubtedly determine whether the exogenous Sr is removed during the leaching steps, and if the remaining signal derives from the sole endogenous Sr. However, we can certainly state that the radiogenic isotopic signal of the hair samples (>0.710) does not likely originate from post-burial contaminants (i.e. the crypt environment), because such signature has not been detected within the respective leachates (~0.709). This suggests the possibility of a biogenic and/or a pre-burial exogenous origin of the high radiogenic Sr isotope signals observed during this study.

Most trace elements in the archaeological hair specimens present intensities similar to those observed in the contemporary sample. In particular, metals (e.g. Cu, Fe and Zn) and REEs show values comparable or even lower than the contemporary TGH specimen (see Supplementary Table 2). We note here that, although the contemporary individual has experienced a different pre-burial exogenous exposure, this is the only possible term of comparison we have for our archaeological specimens. REEs have been long used to assess diagenesis

in human tissues such as bones and teeth (Reynard and Balter 2014), but also in hair studies (Kempson et al. 2010; Wei et al. 2013; Rodushkin and Axelsson 2000; Tong et al. 2004). In this sense, the low REE content we observe for both the archaeological and the contemporary hair may be explained in three different ways: 1) the contemporary individual and the Roccapelago individuals were both not exposed to REEs in their environment; 2) REEs are not incorporated in hair during diagenesis and thus they do not represent an appropriate diagenetic proxy for hair tissue; 3) in contrast, assuming that REEs are a suitable diagenetic proxy, archaeological samples are little or not altered, having a REEs content similar to the contemporary specimen. Compared to the experimentally diagenized hair of Kempson et al. (2010), metals (Mg, Fe, Cu and Zn) in Roccapelago hair seem far less concentrated, resembling the elemental content of the not- diagenized modern hair (Kempson et al. 2010). The only element showing slightly higher concentrations in Roccapelago than in diagenized hair from Kempson et al. (2010) is Mn, but this is mostly lost after the nitric acid leaching. In fact, our elemental data also indicate that after the 2M nitric acid leaching most of the metal content is lost (Table 4). It is not clear if the totality of the exogenous metals (e.g. Ca, Sr and Pb) is removed with this leaching (Font et al. 2012). However, the fact that after the acid wash the metal content drops drastically in archaeological hair samples, occasionally reaching levels below the contemporary hair threshold, may suggest that part of the contaminants is removed.

### *4.3. Archaeological human hair*

Among the archaeological human hair of Roccapelago, some samples (ROC-US26-2A; ROC-US28-1B and ROC-US23-2) are consistent with the local Roccapelago range, as measured through the Sr isotope signature of local rodents, likely demonstrating a local isotopic signature (Fig. 8). Other samples (ROC-US26-1C and D; ROC-T9; ROC-US26-2B; ROC-US28-1A; ROC-US28-4A and B; ROC-T9) fall slightly outside (~0.0001) the local range. The more interesting samples are ROC-US26-1A and B and ROC-US28-2A and B, showing radiogenic $^{87}Sr/^{86}Sr$ ratios quite different from the Roccapelago local values (Fig. 8).

The four sub-specimens of individual ROC-US26-1, (ROC-US26-1A, ROC-US26-1B, ROC-US26-1C and ROC-US26-1D) represent the highest-resolution sampling of this study. Each one of these sub-samples should reflect the average $^{87}Sr/^{86}Sr$ ratio of a ca. 3 month period. For this rough estimation, we did not take into account possible growth cycle errors or different growth rates of the hair (contemporary individual of this study; Williams et al. 2011). In fact, we are not interested in trying to precisely decipher the timing of the movements, but rather simply to understand whether the travels were on a sub-annual scale. If we wanted to correct for growth errors, we should analyze a sub-sample containing anagen hair only (Williams et al. 2011): this is quite difficult, if not impossible, for our case study, where a very limited amount of specimen is at our disposal. Moreover, given the

nature of the samples, it was not even possible to accurately decipher the root-tip direction. However, considering an average human hair growth rate of 0.35 mm/day, ROC-US26-1 hair strand preserves information of about 1-year-of-life of a Roccapelago individual. To be conservative, even including a likely error of ~30%, the sample should represent a time span between ~8 and ~16 months. Within this strand, we observed a large isotopic variability, from high Sr isotope ratios of two sub-samples (both 0.7105) to lower ratios (0.7084 and 0.7082) close to the local Roccapelago $^{87}Sr/^{86}Sr$ values.

The large variability, from lower to higher radiogenic $^{87}Sr/^{86}Sr$ ratios, observed in the Roccapelago human hair seems to suggest the presence of at least two main destinations for the travels of these individuals. The least radiogenic locality (~0.708) is likely recognizable as the Roccapelago area. Although the local rodents show a quite narrow isotopic interval (0.7086 ± 0.0001), we believe that even those samples falling slightly outside this range can be considered as local. As a matter of fact, many thermal and fresh water samples collected in Tuscan localities nearby (< 20 km) Roccapelago show Sr isotopic ratios around 0.708 (0.7079 – 0.7085; Boschetti et al. 2005), within the Roccapelago local Sr isotope signature.

Conversely, the observed highly radiogenic Sr isotope ratios (>0.71) are not commonly found in Roccapelago or in the Modena Apennines region. Even if most of the Italian geology is dominated by sedimentary rocks dated between the Cretaceous and the Holocene (Lugli et al. 2017c), some places with a high radiogenic Sr isotope signature can be found within the Italian landscape (Voerkelius et al. 2010). Alpine and Southern Italy areas (Calabria and Sicily; Rummel et al. 2010) present elevated Sr isotope ratios (>0.71), mainly related to the presence of old metamorphic rocks (Voerkelius et al. 2010). Moreover, Latium and some Southern Italian volcanoes (e.g. Albani Hills and the Middle Pleistocene lavas of Roccamonfina; Lugli et al. 2017c) present high radiogenic Sr ratios, between 0.709 and 0.710 (Marchionni et al., 2013). However, none of these places seem the best suited travelling destination for our case study. First of all, the Roccapelago community was poor and secluded, and it is unlikely that its inhabitants were used to very long travels (up to the Alps or to Southern Italy), especially on a sub-annual scale. Then, one of the nearest areas within the abovementioned places, namely the Latium volcanoes area (~300 km far), shows Sr isotope ratios up to 0.710, averagely less radiogenic than some of our Roccapelago hair. Therefore, our main explanation for the high Sr isotope signature in some individuals of Roccapelago, based on historical sources, is that the Roccapelago individuals seasonally traveled to Tuscany with their herds. In fact, the close Tuscan Magmatic Province (TMP), which consists of a series of mafic to acid intrusive and extrusive centers, shows high radiogenic Sr isotope ratios (Peccerillo and Donati 2003), similar to those observed in our human hair samples. The TMP $^{87}Sr/^{86}Sr$ ratios are on average higher than 0.710, with the highest observed values of 0.725 (Hawkesworth and Vollmer 1979). In a similar way, also the Tuscan Metamorphic Complex (TMC), a metamorphic Paleozoic crystalline basement, is characterized by Sr

isotope ratios similar to the TMP ratios, reaching even more radiogenic end-members (0.753; Ferrara and Tonarini 1985). However, given that hair takes several months, if not years, to reach isotopic equilibrium with the new Sr isotope end-member (Font et al. 2012), we certainly expect human $^{87}Sr/^{86}Sr$ ratios lower than the highest ratios observed for Tuscany, but still within or nearby the likely environmental range.

As shown in Figure 8, TMP ratios are compatible with the $^{87}Sr/^{86}Sr$ of samples ROC-US28-2A, ROC-US28-2B, ROC-US26-1A, ROC-US26-1B, ROC-US26-1C and ROC-US26-1D, matching the Southern Tuscan area as a possible travel destination for Roccapelago inhabitants. Moreover, as clearly highlighted by hair strand ROC-US26-1, at least one individual from Roccapelago spent 6 months in a high radiogenic region, as the TMP, and the following/previous 6 months in a less radiogenic site, isotopically similar to Roccapelago. In addition, also hair strand ROC-US28-2 shows a large intra-variability. This evidence suggests that travels were probably on a sub-annual scale, according to what we expect for transhumance pastoralism, where shepherds spend half of the year away from home with their flocks (Traversari 2016). Historical sources indicate that women were also migrating away from Roccapelago seasonally in search of employment, but only as far south as Northern Tuscany (mainly in the town of Lucca). Aside from the presence of a metamorphic outcrop south of Lucca, the town and its surroundings are characterized by sedimentary rocks with $^{87}Sr/^{86}Sr$ ratios similar to Roccapelago. This could explain why the two “female” hair strand specimens (ROC-US23-1 and ROC-US26-2) retain a Sr isotopic signal similar to the local Roccapelago signature.

## 5. Conclusions

In this work, we successfully analyzed the Sr isotopic composition of human hair to determine the movements of contemporary and Early-Modern individuals.

We show that hair strands of an individual travelling between Brazil and Italy retained different Sr isotope ratios, likely related to the living locality and the sub-annual movements. The Sr isotope composition of hair never reaches isotopic equilibrium with the drinking water end-members suggesting that: 1) other end-members need to be considered (e.g. environment deposition and food; Vautour et al. 2015) and/or 2) a longer time-period is required for the equilibration process because of the old bone buffer (e.g. Font et al. 2012).

The case study of the Roccapelago individuals demonstrated the transhumance practices exploited by some of these shepherds, involving sub-annual travels to a geologically different locality, likely Tuscany, as testified by historical sources. As observed by Font et al. (2015), we suggest that, under peculiar conditions of preservation, archaeological hair may retain their primary Sr isotope signals, correlated with the movement of the individual

through the landscape. However, more studies about the chemical preservation of archaeological hair are needed to check for possible diagenetic alteration of these unusual, but strongly informative, remains. In this sense, archaeological hair samples can be investigated by single-hair LA-ICP-MS analysis to check for potential post-burial contaminations.

We stress that a successful high-resolution Sr isotope investigation of hair strands to monitor human migration is feasible and requires careful sampling to pinpoint the position of an individual in space and time.

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**Table 1.** Contemporary and archaeological human hair specimens data.

| Hair strand ID | Sub-sample | $^{87}Sr/^{86}Sr$ | 2se | Sr (ppm) | Sample length (cm) | Type |
|---|---|---|---|---|---|---|
| TGH-S | A | 0.70890 | 0.00001 | 8.00 | 1 | Modern |
| | B | 0.70892 | 0.00001 | 6.07 | 1 | Modern |
| | C | 0.70897 | 0.00001 | 2.20 | 1 | Modern |
| | D | 0.70891 | 0.00001 | 6.18 | 1 | Modern |
| | E | 0.70880 | 0.00001 | 6.37 | 1 | Modern |
| | F | 0.70872 | 0.00001 | 5.49 | 1 | Modern |
| | G | 0.70876 | 0.00001 | 4.49 | 1 | Modern |
| TGH-L | A | 0.70927 | 0.00001 | 10.55 | 2 | Modern |
| | B | 0.70919 | 0.00001 | 9.65 | 2 | Modern |
| | C | 0.70900 | 0.00001 | 4.38 | 2 | Modern |
| | D | 0.70878 | 0.00001 | 4.14 | 2 | Modern |
| | E | 0.70870 | 0.00002 | 2.09 | 2 | Modern |
| ROC-US26-1 | A | 0.71052 | 0.00003 | 0.61 | 3 | Archaeological |
| | B | 0.71052 | 0.00003 | 0.57 | 3 | Archaeological |
| | C | 0.70838 | 0.00003 | 0.42 | 3 | Archaeological |
| | D | 0.70823 | 0.00003 | 0.67 | 3 | Archaeological |
| ROC-US26-2 | A | 0.70871 | 0.00001 | 2.30 | 8 | Archaeological |
| | B | 0.70878 | 0.00001 | 2.48 | 8 | Archaeological |
| ROC-US28-1 | A | 0.70831 | 0.00003 | 1.08 | 7 | Archaeological |
| | B | 0.70864 | 0.00003 | 0.70 | 7 | Archaeological |
| ROC-US28-2 | A | 0.71374 | 0.00002 | 0.74 | 4.5 | Archaeological |
| | B | 0.71071 | 0.00002 | 0.68 | 4.5 | Archaeological |
| ROC-US28-4 | A | 0.70838 | 0.00003 | 0.93 | 7 | Archaeological |
| | B | 0.70825 | 0.00003 | 1.28 | 7 | Archaeological |
| ROC-T9 | - | 0.70822 | 0.00003 | 0.39 | Bulk | Archaeological |
| ROC-US23-1 | - | 0.70878 | 0.00002 | 0.90 | Bulk | Archaeological |
| ROC-US23-2 | - | 0.70848 | 0.00004 | 0.65 | Bulk | Archaeological |

**Table 2.** Water and archaeological rodent samples data.

| Sample | $^{87}Sr/^{86}Sr$ | 2se | Type | Sr (ppm) |
|---|---|---|---|---|
| W1 | 0.71474 | 0.00001 | Tap water - USP São Paulo fountain | 0.06 |
| W2 | 0.72818 | 0.00001 | Bottled water - Brazil | 0.04 |
| W3 | 0.72777 | 0.00002 | Bottled water - Brazil | 0.02 |
| W4 | 0.70855 | 0.00001 | Bottled water - Italy | 0.23 |
| W5 | 0.70828 | 0.00001 | Tap water - Reggio Emilia | 0.08 |
| W6 | 0.70841 | 0.00001 | Tap water - Reggio Emilia | 0.07 |
| R-ROC-1 | 0.70855 | 0.00001 | Archaeological rodent bone | n.d. |
| R-ROC-2 | 0.70861 | 0.00001 | Archaeological rodent tooth | n.d. |
| R-ROC-3 | 0.70847 | 0.00001 | Archaeological rodent tooth + bone | n.d. |
| R-ROC-4 | 0.70856 | 0.00001 | Archaeological rodent tooth | n.d. |
| R-ROC-5 | 0.70873 | 0.00001 | Archaeological rodent bone | n.d. |
| R-ROC-6 | 0.70853 | 0.00001 | Archaeological rodent tooth | n.d. |
| R-ROC-7 | 0.70866 | 0.00001 | Archaeological rodent bone | n.d. |
| R-ROC-8 | 0.70860 | 0.00001 | Archaeological rodent tooth + bone | n.d. |
| R-ROC-9 | 0.70856 | 0.00001 | Archaeological rodent tooth | n.d. |
| R-ROC-10 | 0.70855 | 0.00001 | Archaeological rodent tooth + bone | n.d. |

**Table 3.** $^{87}Sr/^{86}Sr$ ratios of archaeological hair leachates.

| Sample | $^{87}Sr/^{86}Sr$ | 2se |
|---|---|---|
| L-ROC-US26-1 W* | 0.70863 | 0.00001 |
| L-ROC-US28-1 W* | 0.70890 | 0.00001 |
| L-ROC-US28-2 W* | 0.70861 | 0.00001 |
| L-ROC-US26-1 A | 0.70855 | 0.00001 |
| L-ROC-US26-1 B | 0.70855 | 0.00001 |
| L-ROC-US26-1 C | 0.70861 | 0.00001 |
| L-ROC-US26-1-D | 0.70855 | 0.00001 |
| L-ROC-US28-1 A | 0.70848 | 0.00001 |
| L-ROC-US28-1 B | 0.70849 | 0.00001 |
| L-ROC-US28-2 A | 0.70877 | 0.00001 |
| L-ROC-US28-2 B | 0.70855 | 0.00001 |

*leachate from MilliQ washes.

Other samples are 2M $HNO_3$ step leachates.

**Table 4.** Average losses (%) of some possible contaminants (REE, Sr and Pb) after the 2M $HNO_3$ leaching step for all the studied samples.

| **Contaminant** | **Loss (%)*** |
|---|---|
| REE** | 30.7 |
| $^{88}Sr$ | 88.7 |
| Pb*** | 96.3 |

*Loss is calculated as the difference between the elemental intensities before and after the nitric acid leaching. **140Ce from sample ROC-US26-2 excluded. ***Pb includes $^{206}Pb$, $^{207}Pb$ and $^{208}Pb$.


**Figure 1.** (a,b) Annotations from the Roccapelago parish archive. (a) Italian transcription: «*Gli abitanti della Parrocchia di Rocca Pelago sono soliti andare fuori di Patria l'Inverno a guadagnarsi il vitto e vestito*»; English translation: «*The inhabitants of the Parish of Rocca Pelago commonly move away from their homeland in winter to earn food-and-board and clothing*» (Stato delle Anime, v. 46, A.P.Ro). (b) Italian transcription: «*Biagio* [figlio] *di Domenico Ugolini morse e fu seppellito nell'Hospitale di Grosseto secondo tutti li sacramenti della Chiesa*»; English translation: «*Biagio* [son] *of Domenico Ugolini died and was buried at the Hospital of Grosseto following the* [Catholic] *Church rituals*» (Libro dei Morti dal 1599 al 1738, p. 47, nota 3, A.P.Ro). (c) One of the partially mummified individuals recovered from the Roccapelago crypt. (d) Map of Italy with the location of the Roccapelago site; other areas are also reported (see text).

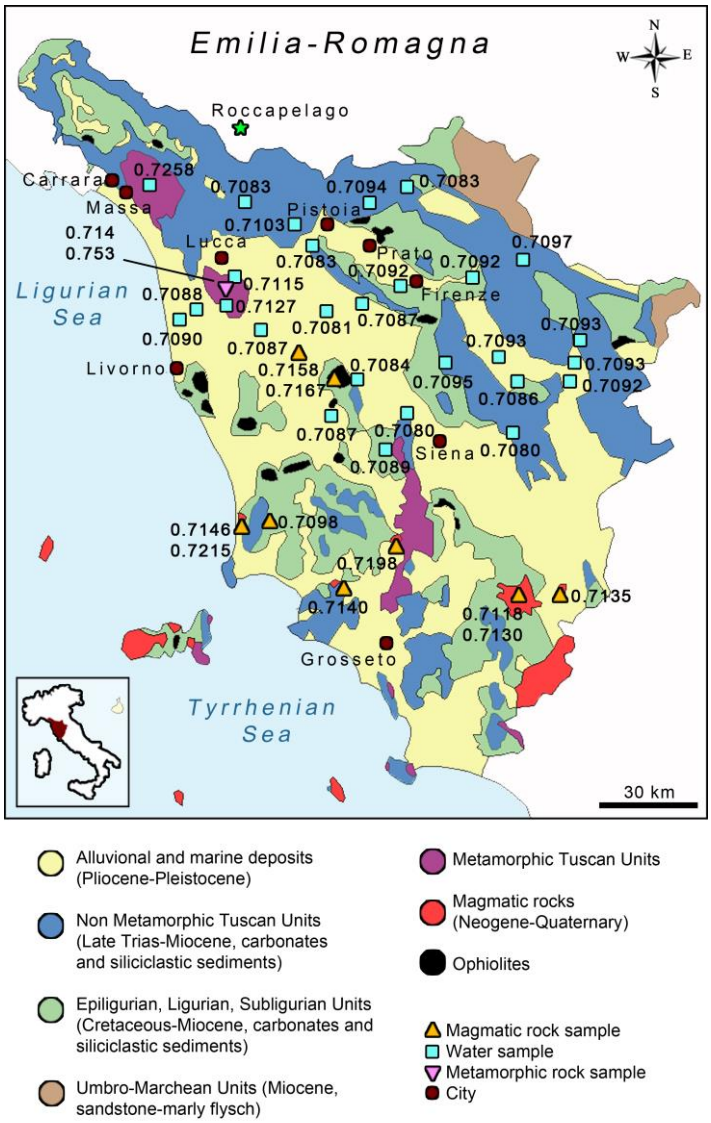


**Figure 2.** Simplified geological map of Tuscany (modified from Perrone et al., 2006). Sr isotope ratios of several samples or rocks and waters from literature are reported. Waters are from Nisi et al. 2008; Boschetti et al. 2005. Magmatic rocks are from Peccerillo and Donati 2003; Ferrara and Tonarini 1985; Hawkesworth and Vollmer 1979. Metamorphic rocks are from Ferrara and Tonarini 1985.

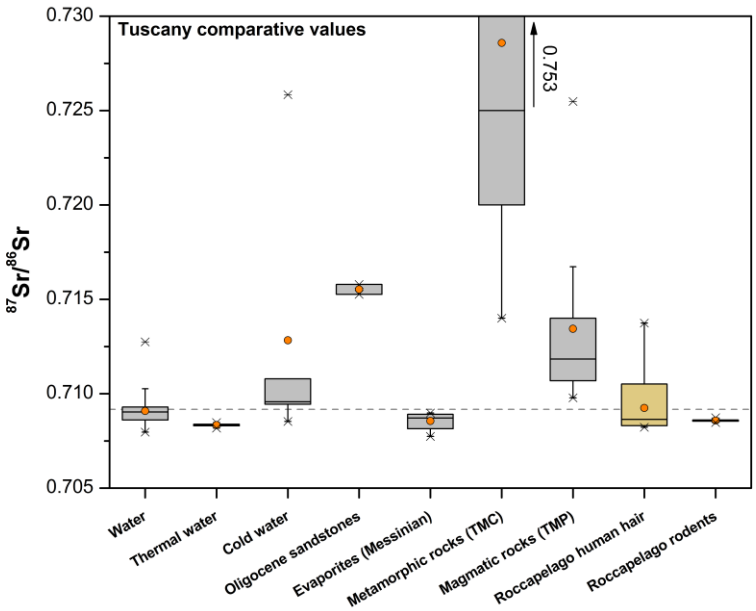


**Figure 3.** Tuscan Sr isotope ratios of rocks and waters from literature in comparison to Roccapelago inhabitants human hair and local rodents. Waters are from Nisi et al. 2008. Thermal waters from Boschetti et al. 2005. Cold waters from Boschetti et al. 2005. Oligocene sandstones from Boschetti et al. 2005. Evaporites from Boschetti et al. 2005; Dinelli et al. 1999. Metamorphic rocks (Tuscan Metamorphic Complex) are from Ferrara and Tonarini 1985. Magmatic rocks (Tuscan Magmatic Province) are from Peccerillo and Donati 2003; Ferrara and Tonarini 1985; Holm and Munksgaard 1982; Hawkesworth and Vollmer 1979. The dashed line is modern seawater (~0.7092).

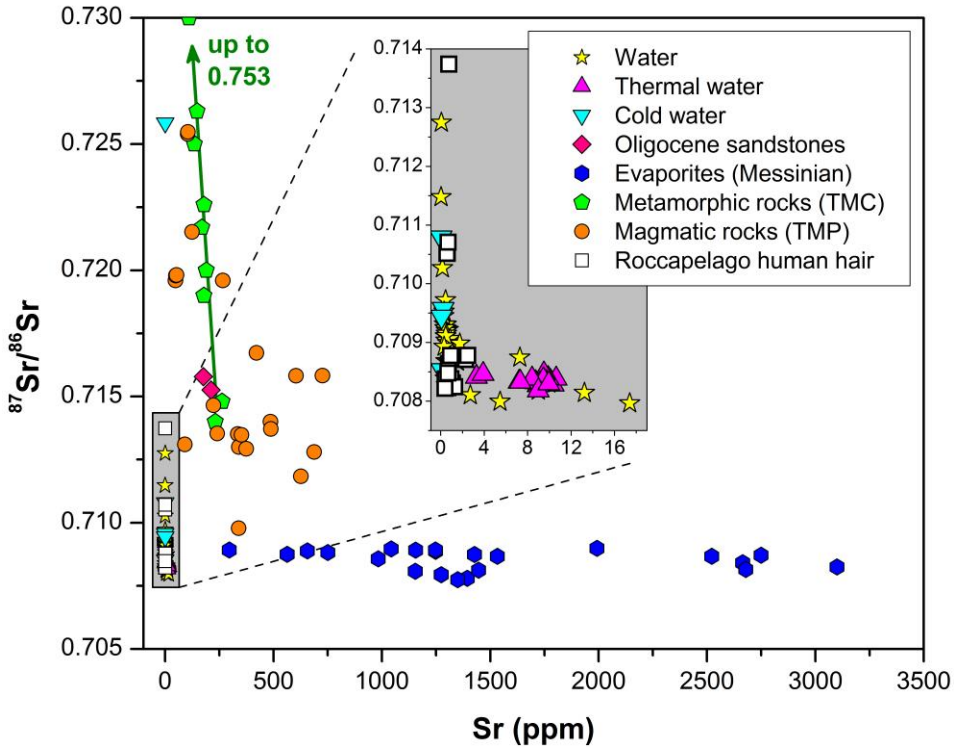


**Figure 4.** $^{87}Sr/^{86}Sr$ versus Sr (ppm) of Tuscan water and rock samples from literature [references as in Fig. 3].

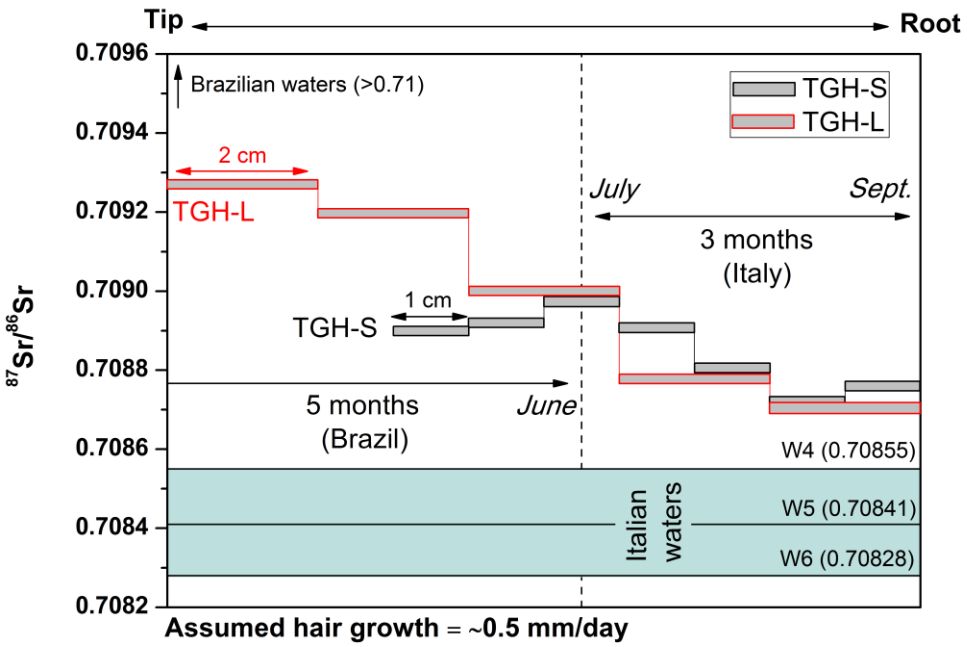


**Figure 5.** Temporal Sr isotope ratio profiles of the modern hair strands. Bottled/tap water samples from Italy and Brazil are reported for comparison (see Table 2). A hair growth of ~0.5 mm/day is assumed as the likely best-fit (see text for details). The width of the symbol represents the length of the subsample, the height of the symbols (y-axis) is the 2σ propagated error of the analysis.

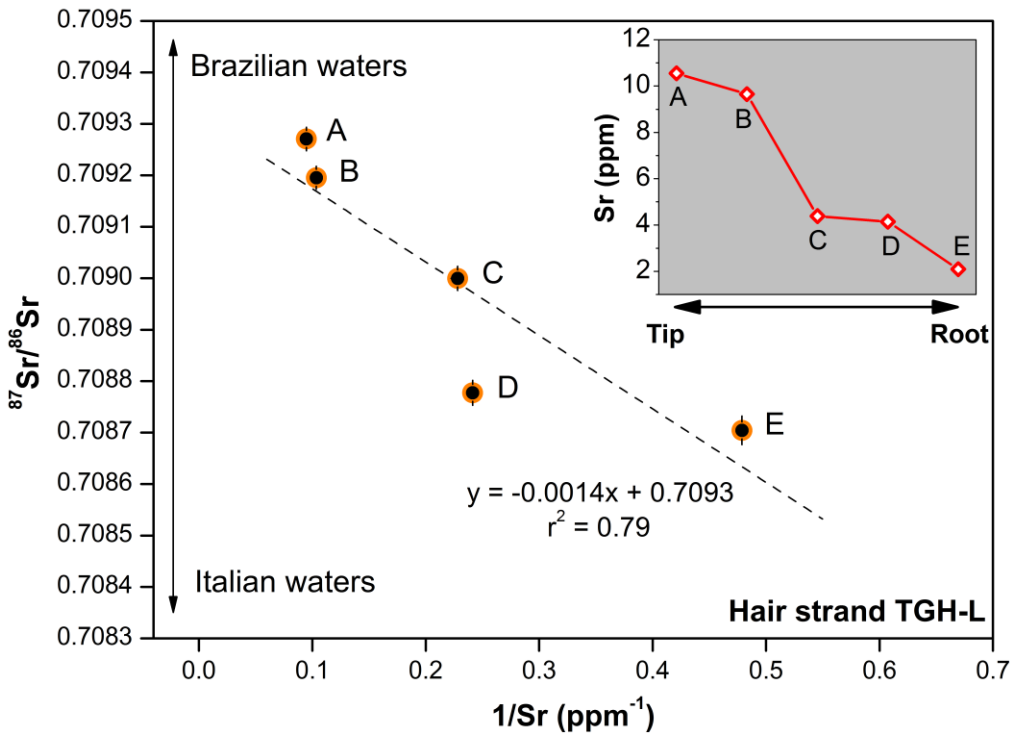


**Figure 6.** $^{87}Sr/^{86}Sr$ vs. 1/Sr ($ppm^{-1}$) of modern hair strand TGH-L. In the inset, the Sr compositional profile of the TGH-L hair strand, showing the highest Sr concentration at the hair tip. Y-axis error bars are 2σ of the Sr isotope analysis. The arrows along the Y-axis indicate the direction of the Sr isotopic composition of the water end-members of Italy and Brazil used in this study.

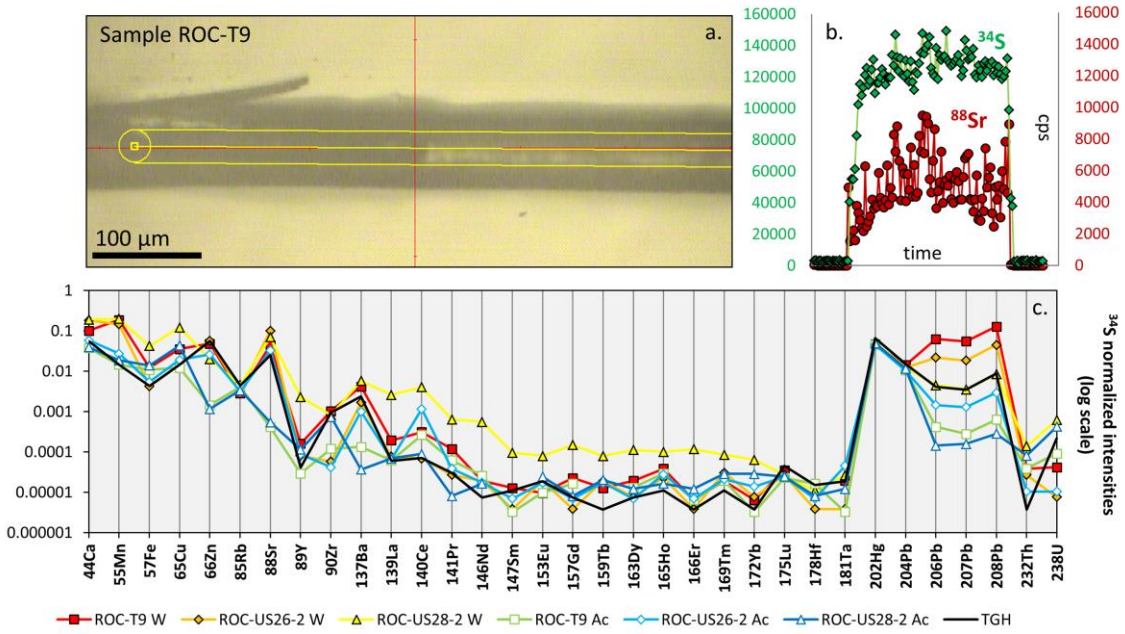


**Figure 7.** LA-ICP-MS trace element analysis of human hair specimens. (a) Microphotograph of sample ROC-T9 during laser ablation analysis. (b) Typical $^{34}S$ and $^{88}Sr$ elemental profiles of hair in counts per second (cps). (c) $^{34}S$ normalized elemental intensities of contemporary (TGH) and archaeological hair samples; specimens labelled as 'W' were washed with MilliQ® water only; samples labelled as 'Ac' were also leached with 2M $HNO_3$. For cleaning protocol of modern hair see text.

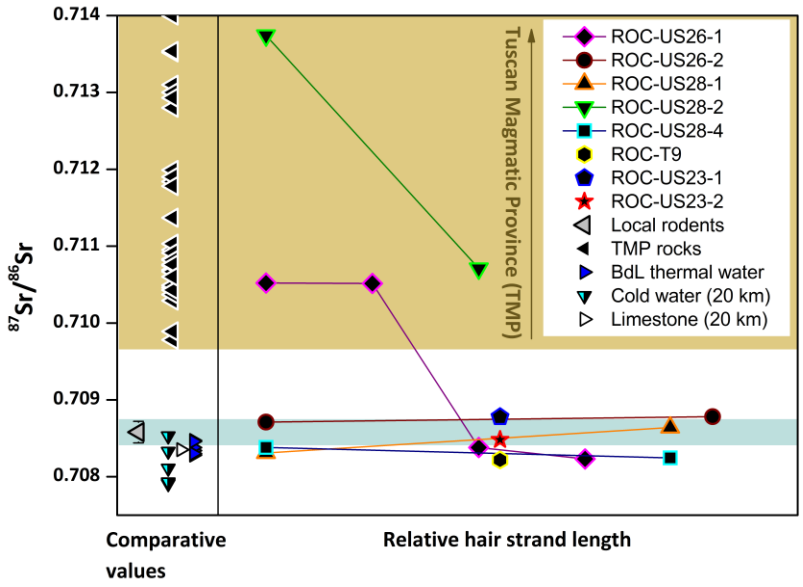


**Figure 8.** $^{87}Sr/^{86}Sr$ ratios of the Roccapelago hair specimens. For comparison, TMP rocks (Peccerillo and Donati 2003; Holm and Munksgaard 1982; Hawkesworth and Vollmer 1979); Bagni di Lucca (BdL) thermal water (Boschetti et al. 2005); 20km-far cold water (Boschetti et al. 2005) and 20km-far limestone (Boschetti et al. 2005) are reported. Archaeological rodent bones and teeth (0.7086 ± 0.0001; 2σ) are also reported as the most likely local reference range (light blue area). Hair strands ROC-US26-2 and ROC-US23-1 are attributed to female individuals. 'A' sub-samples are always on the left portion of the graph.