[Supplementary information]

Mobile elites at Frattesina: flows of people in a Late Bronze Age 'port of trade' in northern Italy

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Strontium isotope analysis: general principles and application to Northern Italy

Strontium isotope ratios in odontoskeletal remains are regularly employed to assess the provenance and trace the mobility of individuals in different phases of their lives. These are determined by comparing the ratio between strontium-87 (^{87}Sr) and strontium-86 (^{86}Sr) in bones/teeth, with the local baseline values measured in faunal/vegetal samples (modern and/or ancient) from the archaeological site or its geologically coherent immediate hinterland. The technique has been in use for more than 30 years in bioarchaeological research and is described in detail in a number of publications (e.g. Grupe *et al.* 1997; Montgomery *et al.* 2000; Bentley & Knipper 2005; Douglas Price *et al.* 2012; Giblin *et al.* 2013; Scheeres *et al.* 2013; Harvig *et al.* 2014; Sjögren *et al.* 2016).

As radiogenic strontium-87 (^{87}Sr) originates over time from the radioactive decay of rubidium-87 (^{87}Rb; half-life of 48.8 Ma), the ratio ^{87}Sr/^{86}Sr depends on the age of a given bedrock, but also on its geochemical nature. Older geological units (>100Ma), such as Palaeozoic metamorphic and Mesozoic igneous rocks in the Alps, generally display higher ^{87}Sr/^{86}Sr values ($\geq$0.71), while younger materials, such as Cenozoic marine carbonates and chalks in the Apennines, show lower ratios ($\leq$0.709). Sediments in alluvial plains reflect the ratio of their parent material, or an admixture of the ratios that characterise the different geological units affected by the erosive activity of the rivers in the uplands.

Frattesina is located on the right bank of the Po di Adria palaeoriver, and therefore the local soils are composed of an admixture of the alluvial sediments collected from both the right (Apennine) and left (Alpine) tributaries. The River Adige runs not far north of the site, carrying exclusively sediments of Alpine origin. Other alluvial basins characterise the area within 50 km: the Brenta river valley in the north (Alpine origin) and the Reno and Panaro river valleys in the south (Apennine origin). Hence, $^{87}Sr/^{86}Sr$ values are anticipated to vary significantly within a relatively small radius.

Bioavailable strontium baselines have been mapped using an open-source geolithological map of Northern Italy (see http://sgi.isprambiente.it/GMV2/index.html), through Quantum GIS software (Figure 4). Ten different "geolithological zones" have been identified, where strontium isotope ratios are available and a framework of northeastern Italy has been summarised in Table S1 and Figure 4.

Thirty-five new baseline values have been produced within the present study, analysing animal tooth enamel from Bronze Age sites (Sant'Eurosia, Casinalbo, Fondo Paviani) or modern snails found on targeted geolithological units at different distances. Ancient faunal remains have been considered to represent an average bioavailable Sr isotope composition over their feeding area (Price *et al.* 2002; Bentley 2006). However, it is very unlikely that humans and domestic animals ate food from distinct locations, marked by different isotope compositions.

Tafuri *et al.*'s (2018) recent work has indeed demonstrated for the *terramara* at Fondo Paviani (as well as for other Terramare sites) that cattle, sheep/goats and domestic pigs were fed with C_4 plants, presumably millet, which was also identified in the pollen series and phytolith record from the site (Dal Corso *et al.* 2017). This means that, during the Terramare period and also presumably at Frattesina, animals were almost certainly fed with fodder cultivated in the surrounding fields, and for this reason their strontium isotope composition most likely reflects the local baseline. Obviously, animals could also be part of gifts/exchanges with other distant communities and, therefore, this source has to be considered critically in comparison with other sources, but aids in validating the inferred bioavailable ranges. For our study, we have added snail shells, also used by several authors as an indicator of the locally bioavailable strontium source (Bentley *et al.* 2002; Wright 2005; Evans *et al.* 2010; Nafplioti 2011; Frei & Price 2012; Laffoon *et al.* 2012; Shishlina *et al.* 2016; Emery *et al.* 2018; Panagiotopoulou *et al.* 2018). Some authors have pointed out that land snail shell $^{87}Sr/^{86}Sr$ can be biased towards values for soil carbonates; nonetheless their values are usually close to those of ground vegetation (Maurer *et al.* 2012).

The analysis of vine branches for wine ‘authentication’ or geographic traceability both north and south of the River Po represents another source of biologically available strontium baselines (Aviani 2013; Trincherini *et al.* 2014; Durante *et al.* 2015, 2016).

We have also taken into account chemical analyses of natural mineral waters (Voerkelius *et al.* 2010). The work by Voerkelius *et al.* is relevant for comparison with the nearest baselines, but strontium isotope ratios from spring waters can only be used with caution, as they represent a very locally-specific kind of evidence, while an individual’s diet is an admixture of different sources from a specific, but wider, area.

The Po Plain is one of the most intensely exploited regions of Europe, with extremely few uncultivated, non-urbanised areas. A very recent detailed Sr isotope survey in Poland (Zieliński *et al.* 2016, 2018) showed that the modern biosphere (animals) and hydrosphere (surface waters) can be contaminated by anthropogenic strontium derived from agriculture, industrial and municipal sources. For that reason, comparison of multiple sample types is necessary to achieve a robust isoscape. Following Emery *et al.*’s (2018) ‘first map’, inspired by a number of examples, all of them interpolating a variety of strontium sources (Evans *et al.* 2010; Nafplioti 2011; Maurer *et al.* 2012; Hartman & Richards 2014; Willmes *et al.* 2014; Laffoon *et al.* 2017), we have considered previous studies, in order to make a comparison between three different sources, namely ancient animals, modern snail shells and modern plants. However, compared to other ‘isoscapes’, the strontium isotope map of Italy still lacks in spatial resolution and critical assessment of baselines, which need to be enhanced. The variation in the currently available strontium isotope ratios for each of the ten geolithological zones is shown in Table S1.

Concerning the different sources of strontium used for baselines, the $^{87}Sr/^{86}Sr$ obtained from different sources at Frattesina appear rather homogenous (0.70853, 0.708639 and 0.70898 for modern snails, 0.70892 for archaeological fauna). We can also compare the values obtained for Emilian Pliocene/Pleistocene limestone: the bedrock yielded a mean $^{87}Sr/^{86}Sr$ of 0.7087, soils 0.7087, snail shell 0.7085, springwater 0.7088, and wine 0.7090. Similarly geolithological zones 1, 2, 7-9 all display narrow ranges from a variety of samples and lithologies. We can therefore conclude that even if there is a slight variation of the isotopic composition, these are nonetheless relatively small, and the eventual impact of anthropogenic strontium (fertiliser/pollution) is negligible. Additional sources for local baselines are nonetheless necessary to refine the preliminary framework presented here.

Buffer zones were drawn around Frattesina at three different radii: 5km (site catchment area, direct control), 20km (immediate hinterland), 50km (broader hinterland), in order to model individual mobility in the territory. Since $^{87}Sr/^{86}Sr$ values within the 5 and 20km radii are rather uniform in this area, the two buffer zones were unified in a larger 0–20 km zone.

Table S1. The 10 identified geolithological zones, $^{87}Sr/^{86}Sr$ baselines (minimum, maximum, mean values), and related references.

Zone number	Zone name	Geolithology	$^{87}Sr/^{86}Sr$ min	$^{87}Sr/^{86}Sr$ max	$^{87}Sr/^{86}Sr$ mean	References
1	Emilian plain	Holocene alluvial sediments (derived from zone 2 or 3)	0.7084	0.7090	0.7087	Trincherini *et al.* 2014; Durante *et al.* 2015; present study
2	Emilian Apennines	Cenozoic marine sediments (sandstones, limestones, marls, turbidites, flysches, sands, clays, chalks)	0.7085	0.7090	0.7088	Vaiani 2000; Scheeres *et al.* 2013; Durante *et al.* 2015; Argentino *et al.* 2017; present study
3	Upper Taro River valley	Mesozoic ophiolites/green stones and Cenozoic marine sediments	0.7092	0.7109	0.7101	Voerkelius *et al.* 2010
4	Garda's moraine amphitheatre	Pleistocene moraine deposits (from zones 6 and 10)	0.7079	0.7080	0.7080	Present study
5	Mantova or Verona plain	Pleistocene alluvial sediments (from zones 6 and 10)	0.7088	0.7089	0.7089	Francisci *et al.* 2017; present study
6	Lower Adige and Lower Brenta valleys	Pleistocene/Holocene alluvial sediments (from zones 6, 9, 10)	0.7089	0.7107	0.7097	Aviani 2013

7	Colli Euganei	Palaeogene-Miocene volcanics, carbonates, dolomites, marls,	0.7077	0.7088	0.7081	Aviani 2013; present study
8	Colli Berici	Palaeogene-Miocene volcanics, carbonates, dolomites, marls,	0.7072	0.7082	0.7077	Present study
9	Monti Lessini	Mesozoic carbonates and dolomites; Cenozoic basalts	0.7076	0.7084	0.7079	Present study
10	Alps (upper Adige/Isarco river valleys)	Palaeozoic metamorphics and volcanics	0.7132	0.7236	0.7202	Müller *et al.* 2003

Methods

Cremated bone samples were drilled using the method reported by Harvig *et al.* (2014) and pre-treated following Snoeck *et al.* (2016: 401). In addition to bioavailable strontium isotope values from the literature (Table S1), baseline samples were taken from pig tooth enamel from the Frattesina settlement and snails from different locations within 2 km of the site. The demineralization of the samples was performed by acid decomposition: a portion of about 50mg of samples was dissolved in 10ml of NHO3 UP 4M.

Ultrapure HNO3 obtained from a sub-boiling system (DuoPUR, Milestone, Bergamo, Italia) and ultrapure 18.2 MΩ water from a Milli-Q (Millipore, USA) system were used for the sample dissolution. HCl of hyperpure grade (Panreac, Barcelona, Spain) was used for sample treatment. SRM-987 isotopic standard from the National Standards and Technology (NIST, Gaithersburg, MD, USA) was used for external precision measurement and method validation. The certified NIST value for the isotopic ratio is $^{87}Sr/^{86}Sr = 0.71034 \pm 0.00026$, which corresponds to an internal precision equal to 0.037%.

The sample solution was loaded into a chromatographic extraction column packed with Sr-resin (Triskem, Bruz, France) where Sr and also Na, K and Ca are retained. A Sr-resin specific method was used (Trincherini *et al.* 2014; Brescia *et al.* 2005) for the elution of the elements and was performed in three steps, using respectively: 5mL 2M HNO3 (fraction 1), 5mL 8M HNO3 (fraction 2) and 5mL of ultrapure Milli-Q for the elution of Sr (fraction 3).

The content of Sr, Rb, Na, K and Ca was measured in the solution obtained after mineralization of the samples (a small aliquot of 100µL was collected just after mineralization) and in each of the three solutions eluted from the chromatographic column. The measurements were performed using the Agilent 7500a ICP mass spectrometer. The solution obtained from the third step of the elution (fraction 3) was then evaporated to dryness and the residue was dissolved in about 50µL of 1% nitric acid solution, in order to ensure a concentration of Sr suitable for TIMS analysis (≈ 200µg g-1).
A Thermal Ionization Mass Spectrometer model MAT 262 VMC from Finnigan (Bremen, Germany), located at the Laboratory of Isotopic Mass Spectrometry (LIMS) of Laboratori Nazionali del Gran Sasso (LNGS) was used for isotope analysis. The instrument is equipped with 5 Faraday cups placed in a variable multicollector, with extensive optical geometry, but corresponding to a system that has a conventional geometry, with a 64cm deflection radius. A characteristic of the thermal ionization source is the stability of the signal, which guarantees a high precision of the measurement. "Zone refined" rhenium filaments were used for sample loading. The double filament technique was adopted. The software Spectromat (Bremen, Germany) was used for data acquisition and analysis; mass calibration and gain calibration were performed daily (Wieser & Schwieters 2005). Six blocks of ten replicates were acquired for each measurement reaching an associated average internal precision ≤ 0.003%.

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