Human enamel $^{87}\text{Sr}/^{86}\text{Sr}$ evidence of migration and land use patterns at Tell Brak, a Late Chalcolithic urban centre in NE Syria

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Abstract
During the Late Chalcolithic 3/4 (c. 3900–3200 BCE), Tell Brak in NE Syria became a large urban centre with central administration, craft specialization and a settlement size of at least 130 ha. $^{87}\text{Sr}/^{86}\text{Sr}$ values in enamel of 34 human individuals from Tell Brak representing four temporal subsets were measured and compared against the local background to understand the pattern of migrations and origin of food consumed by inhabitants of the city. The rate of migration from areas with different $^{87}\text{Sr}/^{86}\text{Sr}$ background values was constantly low, and there is no evidence of long-distance mobility. Decrease of average $^{87}\text{Sr}/^{86}\text{Sr}$ values with time was paralleled by their higher dispersal. Finally, in the last subset representing the end of first urbanization period data are again less variable. All this indicates a rise in spatial heterogeneity of resources to feed the growing population of the expanding city, which was likely caused by a gradual extension of land used for food production.

Keywords Diet · Subsistence · Agriculture · Urbanization · Human enamel · Mobility

Introduction
One of stable strontium isotopes ($^{87}\text{Sr}$) is radiogenic, and its proportion, expressed as $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, differs between rocks of different age and origin (Price et al. 2002; Veizer et al. 1999). As strontium isotopes are not significantly fractionated by biological processes, local geological $^{87}\text{Sr}/^{86}\text{Sr}$ signature is transferred through food to human tissues, which allows for the identification of lifetime migration or non-local diet of humans by comparing $^{87}\text{Sr}/^{86}\text{Sr}$ values in their tissues (as, e.g. enamel) and in local environment (Bentley 2006; Slovak and Paytan 2012), with spatial resolution depending on variability of $^{87}\text{Sr}/^{86}\text{Sr}$ values in local environments.

This tool has been successfully used in several bioarchaeological studies, including research on migration pattern in the southern Levant during the Iron Age (Beherce et al. 2016), Roman and Byzantine periods (Al-Shorman and El-Khouri 2011; Perry et al. 2008; Perry et al. 2011; Perry et al. 2017; Sheridan and Gregoricka 2015). Other parts of the Near East and earlier periods, however, have not been so intensively investigated using this method. For ancient Mesopotamia, there is only one limited comparison of two individuals from the Royal Cemetery at Ur and three sheep from nearby Mashkan-shapir with human remains from Harappa in the Indus valley (Kenoyer et al. 2013). Strontium isotope approach has been adopted in the wider scope only for studies on glass provenance and circulation in northern Mesopotamia (Degryse et al. 2009; Henderson et al. 2009a; Henderson et al. 2010). Any in-depth analyses of migration patterns in Mesopotamia using strontium isotopes are lacking.

Human mobility is an important topic in studies of prehistory and early history of Mesopotamia. Discussion and queries in research have been raised regarding the origin of the Sumerians who inhabited South Mesopotamia in the Late Chalcolithic and Early Bronze Age (Sołtysiak 2006), the relations between farmers and herders (Porter 2012), population movements inferred from settlement patterns (Altaweel et al. 2015) as well as how Iranian and Mesopotamian peoples interacted with each other (Carter and Stolper 1984).

Ongoing discussion about the human mobility during the formation time of first urban centres in the Late Chalcolithic, i.e. 4th millennium BCE (Ur et al. 2007; Algaze 2008; Algaze 2018), is also at the forefront of research.

The earliest known fully developed city in Northern Mesopotamia was Tell Brak (36°40’00"N, 41°03’30"E), with
an administrative centre and craft specialization already present there during the Late Chalcolithic 2 (LC 2, c. 4200–3900 BCE) (Oates et al. 2007). Its settlement size dramatically increased from c. 55 to 130 ha in the Late Chalcolithic 3 (LC 3, c. 3900–3600 BCE) and 4 (c. 3600–3200 BCE) (Ur et al. 2011). During archaeological excavations at the site, a large midden also was found, located on the north-western outskirts of the LC 3 city, at a small satellite mound called Tell Majnuna (36°40′28″N, 41°03′15″E) (Lawler 2007). Accumulated in a relatively short time (3786–3521 cal. BC, four radiocarbon dates based on charred grains retrieved from several strata), the midden contained not only large amounts of ashes, sherds and kitchen waste but also several clusters of disarticulated or partially articulated human remains (McMahon et al. 2007) that represented at least three episodes of catastrophic mortality, with their relative dating based on stratigraphy. The top of the midden was used as a regular cemetery during the LC 4 (Soltysiak and Chilińska-Drapella 2009). After LC 4, Tell Majnuna was no longer used as a midden, and the settlement size at Tell Brak dramatically decreased (Ur et al. 2011). The oldest assemblage of human remains at Tell Majnuna (in terms of stratigraphy) has been found in Area MTW south to the midden, and perhaps it preceded the formation of the mound itself. Partially articulated skeletons of at least 62 individuals were disposed of in a random pattern in a large area. Both males and females were present, but the number of infants was much lower than expected at an atritional cemetery. Pattern of disarticulation and abundant evidence of scavenging suggest that this assemblage was the result of a catastrophic event of mass mortality, unburied bodies were exposed for some time on the ground and finally gathered and disposed of in a large pit (Soltysiak 2010). At the midden itself, in its southern part (Area EM), a dense linear cluster of almost completely disarticulated bones was found (EM loc. 6), containing remains of at least 84 individuals, mainly females, older children and adolescents, and almost no infants. Evidence of scavenging by carnivorous mammals was also abundant. Although this cluster was stratigraphically later than the assemblage of skeletons in Area MTW, complete disarticulation and the pattern of post-mortem damage suggest that these bones belonged to individuals who died in another catastrophic event before that witnessed by the cluster MTW (Soltysiak 2010). Above EM loc. 6 another small cluster has been found (MNI = 8), containing mainly crania (EM loc. 25). Both stratigraphy and the pattern of articulation suggest that it was later than two previous clusters (Soltysiak 2010). Finally, 35 articulated skeletons have been found at the top of the midden (Area EME) (Soltysiak and Chilińska-Drapella 2009), and it was the youngest assemblage of human remains at the site, dated to the end of the LC 4 (3370–3099 cal. BC, two dates based on bone carbonate from EME loc. 8).

Assemblages of human remains from Tell Majnuna make it possible to research not only the patterns of human migrations during the time when Tell Brak was developing into a large urban centre, from the beginning of the LC 3 to the end of the LC 4, but also on possible changes in areas, from which food was acquired for a growing urban population. Upper Miocene and Pliocene sediments around Tell Brak (Ponikarov and Mikhailov 1964) have expected 86Sr/87Sr value of 0.7088–0.7090 (McArthur et al. 2012), but there are also several volcanic basalt fields in the area, with 86Sr/87Sr between 0.7031 and 0.7035 (Lease and Abdel-Rahman 2008), including Kaukab volcano just c. 20 km south-west to Tell Brak. Due to exposure of Upper Miocene sediment surface to contamination by basalt dust, expected 86Sr/87Sr values are slightly lower in the areas SW to Tell Brak and slightly higher NE of the city, where Pliocene sediments are more common and surface contamination by basalt dust is less likely. Therefore, food (cereals and dairy products) obtained at various spots around Tell Brak may have slightly different 86Sr/87Sr signatures, providing some insight into patterns of land use.

86Sr/87Sr values in enamel of early developing permanent human teeth (mainly first incisors and first molars) representing four consecutive assemblages of human remains from Tell Majnuna are compared in this study with expected local 86Sr/87Sr signatures. These signatures are corroborated by available plant and soil samples, in order to identify people that migrated to Tell Brak from elsewhere and to see how expansion and then reduction of the settlement size impacted the diversity of 86Sr/87Sr values in humans, which would reflect possible changes in land use patterns around the city.

Material and methods

Background 86Sr/87Sr data are scarce for Northern Mesopotamia, but fortunately two 86Sr/87Sr values of plants growing at Tell Brak are available (Henderson et al. 2009b). In addition, soil samples have been gathered at Tell Majnuna and three other archaeological sites in NE Syria: Tell Barri located c. 10 km north-east of Tell Brak (36°44′20″N, 41°07′37″E), as well as Tell Ashara (34°55′19″N, 40°34′06″E) and Tell Masaikh (34°58′24″N, 40°33′23″E) in the Euphrates valley (Fig. 1, Table 1). The expected 86Sr/87Sr values at the latter sites are lower due to the presence of basalts in the upstream parts of the valley (Soltysiak 2019).

From four assemblages of human remains retrieved at Tell Majnuna, 34 early developing permanent teeth of adult individuals were selected for 86Sr/87Sr measurement (Table 2), mainly first molars and first incisors. Small fragments of enamel (up to 20 mg) were collected using a Dremel tool from the lower part of the crown, near the cemento-enamel junction. In the case of first molars, it represents 2nd–3rd year of
life, in case of upper first incisors—3rd–4th year of life (AlQahtani et al. 2010), i.e. weaning and early post-weaning time. As most skeletons were disarticulated or only partially articulated, age-at-death and sex were assessed using cranial characteristics such as dental wear degree and overall robustness (Buikstra and Ubelaker 1994), which may be however not well adjusted for a local population (Soltysiak 2010).

Isolation of tooth enamel, homogenization of sediment samples, chemical separation of Sr and measurements of Sr isotope ratios were carried out in the Isotope Laboratory of the Adam Mickiewicz University in Poznań. Tooth enamel was cleaned in an ultrasonic bath in ultrapure water in order to remove the sediment particles. Afterwards, 11–13 mg of powdered enamel was treated sequentially according to the procedure described by Dufour et al. (2007) with 0.1 ultrapure acetic acid (5 times) to eliminate the diagenetic Sr contamination. Subsequently the samples were dissolved on a hot plate (~100 °C, overnight) in closed PFA vials using 1 N HNO 3. The powdered sediment samples (~90–100 mg) were dissolved on a hot plate (~100 °C, 3 days) in closed PFA vials using a mixture of concentrated hydrofluoric and nitric acid (4:1).

The miniaturized chromatographic technique described by Pin et al. (1994) was applied for Sr separation, with some modifications in the column size and concentration of reagents (Dopieralska 2003). Strontium was loaded with a TaCl 5 activator on a single Re filament and analysed in dynamic collection mode on a Finnigan MAT 261 mass spectrometer. Total procedure blanks were less than 80 pg. The 87Sr/86Sr values were corrected to 86Sr/88Sr = 0.1194. The Sr results were normalized to certified the value of NBS-987 = 0.710240. Measured NBS-987 were 0.710228 ± 0.000011 (2σ for a series of ten analyses).

### Results

Available 87Sr/86Sr values for soil and plant samples from NE Syria are shown in Table 1. All differences between samples taken from one location are relatively low, and there is no overlap between sites. Most human tooth 87Sr/86Sr values

| Site         | Source | 86Sr/87Sr | Error  |
|--------------|--------|----------|--------|
| Tell Ashara  | Soil   | 0.70787  | 0.000011 |
| Tell Ashara  | Soil   | 0.70777  | 0.000010 |
| Tell Masaikh | Soil   | 0.70799  | 0.000013 |
| Tell Masaikh | Soil   | 0.70805  | 0.000010 |
| Tell Brak    | Plant  | 0.70815  |        |
| Tell Brak    | Plant  | 0.70823  |        |
| Tell Majnuna | Soil   | 0.70842  | 0.000015 |
| Tell Majnuna | Soil   | 0.70852  | 0.000018 |
| Tell Majnuna | Soil   | 0.70843  | 0.000020 |
| Tell Barri   | Soil   | 0.70876  | 0.000010 |
| Tell Barri   | Soil   | 0.70885  | 0.000010 |

Plant data from Henderson et al. (2009a)
are scattered in a relatively narrow range of 0.7080–0.7083 (Table 2), which is consistent with the local environmental background (Fig. 2). There are only four clear outliers (c. 12% of whole sample), two with lower \(^{87}\text{Sr}^{86}\text{Sr}\) values in EM loc. 6 (0.7078) and two with higher \(^{87}\text{Sr}^{86}\text{Sr}\) values in MTW (0.7086) and EME (0.7084).

In spite of relative homogeneity of the sample, an interesting temporal trend is observed if the outliers are omitted. In the first three chronological subsets, \(^{87}\text{Sr}^{86}\text{Sr}\) values become gradually more dispersed, and simultaneously the average drifts towards slightly lower values (Table 3). Finally, the sample from the top cemetery (EME) is the most homogeneous, and its average moves towards higher values in comparison to previous subset, although here sample size is very low. One-way analysis of variance (ANOVA) shows that differences between subsets are significant \(F = 4.20, p = 0.015\), with the post hoc LSD test indicating that this significance is due to EM loc. 6 distinguished from EM loc. 25 and MTW in pairwise comparison (Table 4). Observed temporal difference in dispersal is not statistically significant (Levene’s test of homogeneity of variances, \(F = 2.33, p = 0.097\)), but it may be the effect of small sample size, especially in the two last subsets. Although sex assessment was available for many individuals, there is under-representation of males in the two earliest temporal subsets, and no clear differences between sexes may be seen in the two later subsets.

### Table 2

Strontium isotope values in human skeletal remains from Tell Majnuna

| No | Tag | Sex | Tooth | Subset | \(^{87}\text{Sr}^{86}\text{Sr}\) | Error |
|----|-----|-----|-------|--------|-----------------|-------|
| 1  | EM 6#149 | ?   | ULI1  | EME    | 0.70822         | 0.000012 |
| 2  | EM 6#208 | F*  | URM1  | EME    | 0.70783         | 0.000010 |
| 3  | EM 6#21C | ?   | LLM1  | EME    | 0.70781         | 0.000010 |
| 4  | EM 53.1132.2.18 | ? | LRI1 | EME    | 0.70822         | 0.000009 |
| 5  | EM 53.L/4.54 | F* | URM1 | EME    | 0.70822         | 0.000009 |
| 6  | EM 6.L/50.1#1 | F* | ULM1 | EME    | 0.70820         | 0.000010 |
| 7  | EM 53.L/5.32 | F | ULM1 | EME    | 0.70817         | 0.000009 |
| 8  | EM 6#140B | ? | ULM1 | EME    | 0.70820         | 0.000010 |
| 9  | EM 53.L/6.16 | ? | URI1 | EME    | 0.70817         | 0.000019 |
| 10 | EM 29.1.4  | F*  | LRM1  | EME    | 0.70828         | 0.000010 |
| 11 | EM 53.L/5.11 | F | ULM1 | EME    | 0.70812         | 0.000010 |
| 12 | EM 53.2143.3 | F* | ULM1 | EME    | 0.70812         | 0.000010 |
| 13 | MTW 66.1.A.77 | M* | LRI2 | MTW    | 0.70811         | 0.000010 |
| 14 | MTW4 65.5.52 | F* | ULI1 | MTW    | 0.70813         | 0.000009 |
| 15 | MTW4 65.2.69 | F* | ULI1 | MTW    | 0.70803         | 0.000010 |
| 16 | MTW4 65.4.60 | F* | LLM1 | MTW    | 0.70813         | 0.000014 |
| 17 | MTW4 65.75  | F*  | ULM1 | MTW    | 0.70805         | 0.000010 |
| 18 | MTW 59.2810 | F  | URI1 | MTW    | 0.70819         | 0.000010 |
| 19 | MTW 66.1.A.72 | F* | LRM1 | MTW    | 0.70822         | 0.000010 |
| 20 | MTW4 63.21  | F*  | LLM1 | MTW    | 0.70817         | 0.000009 |
| 21 | MTW 66.1.A.76 | F | LRM1 | MTW    | 0.70813         | 0.000010 |
| 22 | MTW 66.1.A.99 | F* | ULI2 | MTW    | 0.70863         | 0.000010 |
| 23 | MTW4 65.2.30 | M  | LLM1 | MTW    | 0.70812         | 0.000010 |
| 24 | MTW4 65.89  | F   | LRM1 | MTW    | 0.70816         | 0.000010 |
| 25 | EM 25.HR1.max | ? | URM1 | EM loc. 25 | 0.70803 | 0.000010 |
| 26 | EM 25.HR1.6/7 | ? | ULI1 | EM loc. 25 | 0.70797 | 0.000031 |
| 27 | EM 25.HR.2.1 | F* | ULM1 | EM loc. 25 | 0.70820 | 0.000010 |
| 28 | EM 25.HR.1.10 | F* | URM1 | EM loc. 25 | 0.70809 | 0.000010 |
| 29 | EM 25.HR.2.11 | M* | URM1 | EM loc. 25 | 0.70815 | 0.000010 |
| 30 | EM 25.HR.1.8 | M* | LLM1 | EM loc. 25 | 0.70804 | 0.000015 |
| 31 | EME 2.21.Z | M*  | URI1 | EME    | 0.70843         | 0.000009 |
| 32 | EME #811 | ? | URM1 | EME    | 0.70813         | 0.000011 |
| 33 | EME 8D   | ?   | ULI1 | EME    | 0.70814         | 0.000009 |
| 34 | EME 8E   | M*  | LRM1 | EME    | 0.70816         | 0.000010 |

Four outliers are marked with bold face. Sex assessment based on criteria that have not been tested for Mesopotamian populations is marked with an asterisk.

#### Discussion and conclusion

\(^{87}\text{Sr}^{86}\text{Sr}\) values in plant samples from Tell Brak (Table 1) were close to 0.7082 and lower than expected when taking into account the prevalence of Upper Miocene and Pliocene sediments in the area but explained by the presence of nearby basalt fields (Ponikarov and Mikhailov 1964). The effect of basalt dust seems to be confirmed also by higher soil \(^{87}\text{Sr}^{86}\text{Sr}\) values at Tell Barri, which is located farther away from Kaukab. \(^{87}\text{Sr}^{86}\text{Sr}\) values at Tell Majnuna are higher than at Tell Brak, which is most likely the result of high concentration of ashes in the midden, and they most likely represent fuel transported from various locations. Also strontium concentration at Tell Majnuna is much higher (527–882 μg/g, Sołtysiak 2010) than at Tell Brak (cf. Henderson et al. 2009b). For that reason, soil samples from Tell Majnuna are not used in further analysis. \(^{87}\text{Sr}^{86}\text{Sr}\) values at Tell Ashara and Tell Masaikh, two sites located in the middle Euphrates valley, are lower than at Tell Brak, most likely due to the presence of basalt in the river bed (Demir et al. 2007).

Taking into account the catastrophic character of three out of the four temporal subsets, it is hard to assume that the samples are representative for the whole population of the city. Most likely the bodies disposed at the midden were not transported for a long distance, and they rather represent individuals that died in the NW part of the city and its suburban area.

Among the 34 human samples, there were only four outliers in the \(^{87}\text{Sr}^{86}\text{Sr}\) values, although if regional background data are considered, all of these individuals may have migrated from relatively nearby areas, up to 10 km in SW direction (lower values) or in N/NE direction (higher values), depending on the distance from the basalt fields. Unfortunately, a more detailed regional map of strontium isotopic signatures...
is not available, and therefore, it is not possible to investigate the pattern of migrations with a greater depth. As a result, at this point, there is no evidence of massive constant immigration to the growing city, as suggested by Algaze (2018), and it seems rather that its expansion was due to the gradual concentration of local people, with mobility in most cases limited to nearby villages.

Comparison of data for three consecutive clusters, from EM loc. 6 through MTW to EM loc. 25, reveals a quite clear temporal trend, i.e. a small but statistically significant shift towards lower values accompanied by their higher dispersal. This may be the effect of two processes: either the land used for food production was expanded, specifically in SW/W direction where expected soil $^{87}$Sr/$^{86}$Sr values were lower, or there was a transition from a more centralized economy, with products from various fields around Tell Brak gathered in a central storage place and then re-distributed among final consumers, to more dispersed storage and distribution pattern, resulting in a higher heterogeneity of resources available for particular groups of consumers. Without a more fine-grained map of local strontium isotopic signatures, it is not possible to distinguish properly between these two scenarios, but the data are more consistent with the scenario assuming that development of the urban centre during the LC 3 was accompanied by increased heterogeneity of food sources and growing population size triggered a shift in land use around Tell Brak. In the last temporal subset, $^{87}$Sr/$^{86}$Sr values are again more homogeneous and relatively close to average of the first subset. This is consistent with estimated much lower population size at the LC 4/5 transition.

The observed trend of increasing and then decreasing heterogeneity of $^{87}$Sr/$^{86}$Sr values is consistent with paleoclimatic and archaeobotanical data. During the period of extensive urbanization (LC 3 and 4), climatic conditions were relatively humid and then worsened around 3200 BCE. In spite of this, the proportion of more drought-resistant barley to wheat was higher during LC 3/4, and also alternative crops (as grass pea) were more common in that time, suggesting broader use of lands that were less suitable for plant cultivation (Charles et al. 2010). Also the decrease of average $\delta^{15}$N values in plants suggests a shift from a more intensive to more extensive agriculture in that time (Styring et al. 2017). In the same time,
cross-sectional geometry of male femora at Tell Brak shifted indicating a higher level of physical activity, supposedly related to gathering resources from more distant areas (Sołtysik 2015). All this suggests that growing population size necessitated expansion of land used for agriculture and/or herding also to more dry and marginal areas around the city. Finally, after the collapse of the early city, fields in the closer neighbourhood of Tell Brak were again sufficient to feed much smaller population.

Although limited by available regional background data, the study on $^{87}\text{Sr}/^{86}\text{Sr}$ values in enamel of individuals that died at Tell Brak during the period of early urbanization has produced two important outputs. First, there is no evidence that the $^{87}\text{Sr}/^{86}\text{Sr}$ values are largely dispersed and/or distant from expected local values, which suggests lack of mass migration to the city from places with different $^{87}\text{Sr}/^{86}\text{Sr}$ signatures. All supposed shifts between childhood and perinatal/marginal areas to feed growing population of the rapidly expanding mega-city of that time.

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