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DOI
10.1016/j.scitotenv.2020.138992

Publication date
2020

Document Version
Final published version

Published in
Science of the Total Environment

Science of the Total Environment 729 (2020) 138992

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Spatial patterns in $^{87}Sr/^{86}Sr$ ratios in modern human dental enamel and tap water from the Netherlands: Implications for forensic provenancing

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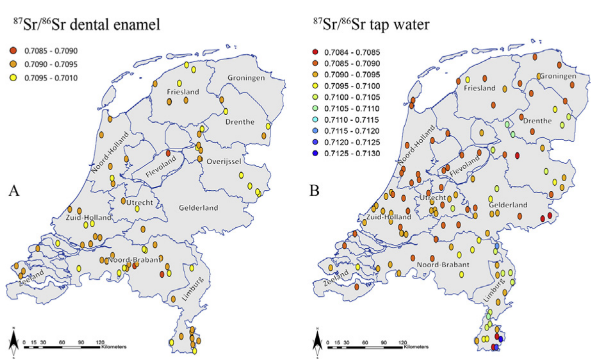
HIGHLIGHTS

- $^{87}Sr/^{86}Sr$ ratios of modern dental enamel and tap water were measured and compared.
- No correlation between $^{87}Sr/^{86}Sr$ ratios of tap water and human dental enamel.
- A model for the prediction of dental enamel $^{87}Sr/^{86}Sr$ data could not be established.
- 98% of Dutch inhabitants have $^{87}Sr/^{86}Sr$ ratios between 0.7088 and 0.7099.
- Sr isoscapes must be used with caution in forensic provenancing investigations.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 March 2020
Received in revised form 22 April 2020
Accepted 23 April 2020
Available online xxxx

Editor: Mae Sexauer Gustin

Keywords:
Strontium isotopes
Human enamel
Tap water
Forensic provenancing
The Netherlands

ABSTRACT

The analysis of strontium isotope ratios in human dental enamel has become important in the fields of archaeological and forensic science for determining provenance and hence mobility. The prerequisite for the approach relies on a correlation between dietary Sr intake and the underlying local geology. This premise is brought into question for anthropological forensic investigations by the increasing globalisation of food supply, the establishment of nation-wide or international supermarket chains, and increasing urbanisation. To better understand the processes that cause spatial variation of Sr isotope ratios in the modern environment, this study determines the range of $^{87}Sr/^{86}Sr$ ratios in the modern Dutch environment based on 296 modern human dental enamel and tap water samples. Tap water $^{87}Sr/^{86}Sr$ from the Netherlands range from 0.70837 to 0.71278 ($\Delta Sr_{max-min} = 0.0044$) and modern human enamel from 0.70847 to 0.70995 ($\Delta Sr_{max-min} = 0.0015$). The $^{87}Sr/^{86}Sr$ ratios of tap water are predominantly determined by the underlying bedrock geology at the sampling point. In contrast, the human enamel data record an insignificant, weak correlation with water supply or local geology. Hence, the main principle behind the application of $^{87}Sr/^{86}Sr$ as a proxy for mobility appears invalid in the modern globalised Dutch context. The range of $^{87}Sr/^{86}Sr$ in modern Dutch humans that can be used for anthropological forensic investigations is between 0.7085 and 0.7100 (n = 153), with 98.0% of individuals between 0.7088 and 0.7099.

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https://doi.org/10.1016/j.scitotenv.2020.138992
0048-9697/

1. Introduction

Dental enamel has become increasingly important in the fields of archaeological and forensic science for determining the dietary intake and geological provenance of humans (Bartelink et al., 2016; Bentley, 2006; Font et al., 2015) and animals (Brusgaard et al., 2019; Evans et al., 2019; Laffoon et al., 2015). The most common isotopic systems applied to dental enamel in both archaeological and modern contexts are strontium ($^{87}Sr/^{86}Sr$), oxygen ($\delta^{18}O$), and carbon ($\delta^{13}C$). Recent research, however, shows promising potential for various other isotope systems, such as neodymium ($^{143}Nd/^{144}Nd$: Plomp et al., 2019; Plomp et al., 2017), calcium ($\delta^{44/42}Ca$: Tacail et al., 2019; Tacail et al., 2017) and zinc ($\delta^{66}Zn$: Jaouen et al., 2018; Jaouen et al., 2016).

Elements enter the human body through the ingestion of food and water and become incorporated into the crystal lattice of bioapatite, the principle component of dental enamel, dentine, and bone. In the case of strontium, Sr^{2+} replaces Ca^{2+} due to comparable ionic radius and identical ionic charge (Zapanta LeGeros, 1981). Half of the ameloblasts, enamel forming cells, undergo apoptosis (disappear) during amelogenesis, the rest die after the maturation stage, i.e., during and after eruption (Giacaman et al., 2016; Matalová et al., 2015). As a result, enamel cannot be repaired or remodelled. Consequently, the isotopic signatures of dental enamel are indicative of the environment of the diet consumed during enamel formation. In human individuals, mineralisation of the permanent molars occurs between birth and circa 16 years of age, with slight variations observed between European, Asian and African populations (e.g., Nelson and Ash, 2010; Reid and Dean, 2006).

Extensive reference datasets or modelled predictive maps displaying *isotope landscapes* (isoscapes) are essential to allow accurate data interpretation in respect of potential provenance (Bataille et al., 2018; Bowen, 2010; Evans et al., 2010; Hoogewerff et al., 2019; West et al., 2010). For the Netherlands, a Sr isoscape and a reference $\delta^{18}O$ dataset based exclusively on archaeological samples have been published (Kootker et al., 2019; Kootker et al., 2016b). Although these reference datasets are invaluable for archaeological research, their applicability for anthropological forensic provenancing investigations are considered limited due to changing land usage, climate, environmental pollution, and the globalisation of diets. The use of environmental reference databases and isoscapes to make provenance determinations is based upon the assumption that the vast majority of the consumed foods, or Sr intake, is from local origin. An assumption that may be valid for archaeological research (but see e.g., Bakels and Jacomet, 2003; Kooistra et al., 2013; Wright, 2005 and references therein), but not necessarily for forensic research. The Netherlands is a densely populated and prosperous country with the 6th largest economy in the European Union (nominal GDP, 2019 data from IMF.org), supported by foreign trade (import and export). Here, due to the increasing globalisation of food supply, the establishment of nation-wide or international supermarket chains, and increasing urbanisation, an isotopic signature that reflects the more uniform supermarket-diet may impact anthropological forensic investigations. The possibility of a more uniform Sr isotope signature of modern people is strongly linked to the economic position of the Netherlands and its consequences with respect to the accessibility of (exotic) foods. This situation is certainly different in other parts of Europe and the rest of the world. This is demonstrated by $^{87}Sr/^{86}Sr$ ratios of modern keratin samples that show a correlation with geography, although for the strontium isotope system this may in part be a consequence of the tendency of hair keratin to incorporate exogeneous Sr through interactions with local tap water (e.g., bathing - Ehleringer et al., 2008; Nardoto et al., 2006; Tipple et al., 2018).

The increase in application of strontium isotopes in human forensic case work (Bartelink and Chesson, 2019; Bartelink et al., 2016; Font et al., 2015) calls for greater insights into the processes that cause spatial variation of Sr isotope ratios in the modern environment. Therefore, 94 modern human dental enamel and 124 tap water samples are presented in this study, and interpreted together with previously published data (n_{total} = 296, including Font et al., 2015; Plomp et al., 2019; Plomp et al., 2020). This paper presents the first extensive overview of the range of $^{87}Sr/^{86}Sr$ ratios that can be expected within modern Dutch individuals. The research then assesses the feasibility of using both water and enamel samples to aid in geolocating unidentified human remains in forensic investigations.

2. Strontium isotope system

The principle behind strontium isotope analyses of human remains is based on the premise that "You are (isotopically) what you eat". Strontium enters our system though ingestion of food and drinking water (Bentley, 2006; Erickson, 1985). Mass-dependent fractionation of $^{87}Sr/^{86}Sr$ is negligible because of the large atomic mass of strontium, and the fact that ^{87}Sr is only 1.16% heavier than ^{86}Sr (Faure, 1986). Moreover, any possible mass-dependent fractionation in $^{87}Sr/^{86}Sr$ is corrected during mass spectrometric measurement by the routine normalisation to a constant $^{88}Sr/^{86}Sr$ ratio (Faure, 1986). During weathering processes and soil formation, mineral breakdown is associated with some incongruent reactions such that bioavailable Sr isotope ratios will be similar but not identical to the underlying rocks. This means that Sr passes from bedrock to soil into biologically-available solutions of which the $^{87}Sr/^{86}Sr$ ratio may be accurately predicted using sophisticated models (see extensive discussion in Bataille et al., 2018). Although international trade and the import of food is not restricted to the modern period, the assumption is made that the vast majority of the food available for ancient populations were of local origin, allowing a direct comparison to be made between the human enamel $^{87}Sr/^{86}Sr$ ratios and biologically available $^{87}Sr/^{86}Sr$ ratios of the surrounding region that are linked to the underlying geology. For modern populations, however, the import and national distribution of foods potentially has a major impact on human $^{87}Sr/^{86}Sr$ ratios.

3. Modern diet and food provenance

The average food consumption of the Dutch population is dominated by dairy products, followed by cereals and vegetables. These three food groups contain relatively high concentrations of strontium [Sr] and therefore dominate the dietary strontium input to the modern human population. Fruit, fish and meat are of lesser importance in the Dutch diet; the consumption of fish and fruit are even amongst the lowest in Europe (Geurts et al., 2017). The strontium concentrations of food sources vary considerably within the food types and between source regions (Burton et al., 1999), although clear trends are observed. The Sr concentrations of grains and cereals are, compared to other food sources, relatively high. On average, 2–10 ppm (μg/g) can be expected, depending on for example the Ca content of the taxa (Browning and Cowieson, 2013; Laursen et al., 2011; Schroeder et al., 1972). Edible plants and vegetables exhibit a wide range of strontium concentrations (1–500+ ppm), with the highest concentrations found in leafy vegetables (ATSDR, 2004; Burton et al., 1999; Schroeder et al., 1972). In contrast, dairy products contain lower strontium, between 0.4 and 11 ppm (Health Canada, 2007; Schroeder et al., 1972; Swoboda et al., 2008). The Sr content of the edible parts of fish and mammals is low due to the fact that fish and mammals discriminate against Sr in favour of Ca. Hence, only 20–40% of ingested Sr is absorbed (Sillen and Kavanagh, 1982 and references therein). For example, Rummel et al. (2012) report typical [Sr] of <1 ppm for beef in Europe. The Sr content of fish is ~10 times higher (1–23 ppm in Carvalho et al., 2005), comparable to that of cereal and dairy products.

The Netherlands both exports and imports food, including raw and processed products from the three main food groups. In 2018, dairy products were imported predominantly from Germany, Belgium, Ireland and France (www.zuivelnl.org). Due to the inadequate quality of wheat grown in the Netherlands (low protein content), the Dutch wheat supply finds its way to the cattle feed industry, rather than to the human consumers. The milling industry, is therefore largely dependent on imports of wheat from France and Germany to meet both human and animal demands (www.agrimatie.nl). Similarly, Dutch malting plants also rely on imported barley and malt from Belgium, Germany, Denmark and France (www.euromalt.be). The import of dairy products, wheat and grains is therefore expected to result in a significant input to Dutch foods that have $^{87}Sr/^{87}Sr$ ratios non-local to the Netherlands. The vast majority of the imported fruits and fresh vegetables are designated for re-export. Moreover, the leafy vegetables containing the highest Sr content are grown in greenhouses in the Netherlands throughout the year. The imported vegetable taxa, such as tomatoes, beans, and carrots, contribute less significantly to the Dutch diet and hence the dietary Sr intake.

The most notable effect of globalism and industrial scale food production on the $^{87}Sr/^{86}Sr$ ratios available to the modern human diet is that of the supermarket chains, distributing locally grown foods and imported food products throughout the country. The supermarket-diet will tend to lead to homogenisation of $^{87}Sr/^{86}Sr$ ratios in the region potentially overprinting the local geology (e.g., Ehleringer et al., 2008; Nardoto et al., 2006).

4. Tap water resources in the Netherlands

Water supply in the Netherlands is divided into 10 different areas managed by different drinking water companies. Four types of raw water sources are used: groundwater (58%), natural dune water (1%), riverbank filtration water (6%) and surface water (35%) (Vewin, 2017). The water companies produce and distribute circa 1150 million m^3 of drinking water per year. Vitens, Evides Waterbedrijf, and Brabant Water are responsible for the production 701 million m^3; providing clean drinking water to 10.3 million residents in 2016 (Vewin, 2017). Tap water in the coastal provinces of Noord-Holland, Zuid-Holland, and Zeeland is dominated by water abstracted from only infiltration, surface, and riverbank filtration water sites. All other provinces receive tap water from groundwater sources, with a few exceptions in Overijssel (riverbank infiltration), Gelderland (push moraine - infiltration), and Limburg (combination of all).

The Netherlands consists of a mixed geology, with Holocene sediments in the north and western part of the country and Pleistocene sediments in the south and eastern part of the country (Vos, 2015). These different deposits are expected to influence the isotopic values of particularly the groundwater resources (Zou et al., 2018). Holocene age sediments are expected to exhibit $^{87}Sr/^{86}Sr$ ratios approximating precipitation/seawater (~0.7091 - Veizer, 1989). Pleistocene sediments are likely to exhibit more radiogenic signatures due to their age and provenance (>0.7095, based on Kootker et al., 2016b). The Sr intake of humans can be expected to include a mixture of sources, such as public tap water, commercial natural mineral water, and water based beverages (Ehleringer et al., 2010; Montgomery et al., 2006). The $^{87}Sr/^{86}Sr$ ratios of tap water are expected to be at least partially related to the type of abstraction site, raw water source, and water quality. Strontium concentration and hardness of water are for instance directly related (Schroeder et al., 1972).

Ground water sources potentially exhibit a wide range of $^{87}Sr/^{86}Sr$ ratios. Groundwater from shallow aquifers is likely to reflect a mixed isotopic composition of recent rainwater and the infiltrated soil. In the Netherlands, however, ground water used for tap water production is generally abstracted at depths between 15 and 200+ m below surface (e.g., vewin.nl; vitens.nl). Groundwater, as well as riverbank filtration water, remain in the soil for years (e.g., at Oasen's abstraction sites: 2–50 years) before abstraction. Water-rock interaction during this period results in groundwater with a Sr isotope ratio that reflects the soil composition (Johnson and DePaolo, 1997; Zou et al., 2018). Hence, although river water used for the riverbank filtration sites from the Greater Rhine and other major river systems may have been transported across great distance, local geology dominates its isotopic signature due to the lengthy filtration process. The dune infiltration processes are also expected to affect isotopic compositions. Infiltration sites use different raw water sources: rainwater and, predominantly, pre-purified river water that is pumped to infiltration ponds in the dune areas and the infiltration period takes several months for the natural processes in the subsurface to improve the water quality. Consequently, the tap water $^{87}Sr/^{86}Sr$ ratios are expected to represent a mix of the infiltrated river water and that of the filtering sediments (and thus to be close to 0.7091, Kootker et al., 2016a, 2016b).

Surface waters are formed from a mixture of sources, such as groundwater discharge and surface runoff. Surface water extraction sites are equally compromised by human activities, e.g., agriculture, industry, medication, etc. (ILT, 2017). Due to the application of advanced purification techniques these pollutants can be efficiently removed. The exact effect of these contaminants on the Sr isotope composition of the purified tap water is unclear.

5. Experimental approach

5.1. Sampling strategy

5.1.1. Dental enamel

Extracted mandibular and maxillary third molars were donated to the Vrije Universiteit Amsterdam. An additional 94 modern individuals were added to existing datasets (Font et al., 2015; Plomp et al., 2019; Plomp et al., 2020). The use of the human dental elements for scientific research was approved by the Medical Ethics Review Committee of the Amsterdam UMC, location VUmc. Background information on the donors was obtained by anonymous questionnaires, providing information on the donor's geographical location at the time of tooth formation, as well dietary preferences and health. None of the individuals relocated during the formation and mineralisation period of the dental enamel.

After removing the outermost surface of the enamel, circa 1–3 mg of dental enamel powder was collected using a diamond-tipped burr precleaned in 10% HCl. The samples were taken from the mesial or distal lobe of either the buccal or lingual surface, depending on the physical quality of the molar and the presence of carious lesions. The samples were sealed in acid-cleaned polyethylene Eppendorf centrifuge tubes and transported to the class 100 clean laboratory at the Vrije Universiteit Amsterdam.

5.1.2. Tap water

A total of 124 tap water samples were taken between May and November 2018 (see Supplementary data for an overview of the sampled locations). 50 ml polyethylene centrifuge tubes were rinsed three times with tap water before circa 45 ml of tap water was collected. The samples were then transferred to the Vrije Universiteit Amsterdam and stored in a fridge at 4 °C to avoid evaporation. The vials were transported to the class 1000 clean laboratory at the Vrije Universiteit Amsterdam, where circa 10 ml of tap water was subsampled into 30 ml acid-cleaned Savillex PFA beakers. The subsamples were dried down over night, capped and stored. In addition, 60 tap water samples were selected for concentration measurements. For concentration measurements, a 2 ml aliquot of tap water was pipetted into a 10 ml acid-cleaned ICP Exetainer® vial and acidified with 118 µl 14 M HNO_3.

5.2. Analytical methods

5.2.1. Isotope analysis

The dental enamel samples and tap water residues were dissolved in 500 µl 3 M HNO_3 for ion exchange chromatography. Detailed descriptions of the Sr extraction/chromatography protocol and the sample loading protocol are provided in Kootker et al. (2016a). The strontium isotope compositions were measured on a ThermoFinnigan Triton *Plus* thermal ionisation mass spectrometer (TIMS) at the Vrije Universiteit Amsterdam. The ratios were determined using a static routine and were corrected for mass-fractionation to $^{86}Sr/^{88}Sr$ of 0.1194. The NBS987 standard gave a mean $^{87}Sr/^{86}Sr$ value of 0.710258 ± 0.000009 (n = 15). The current certified value of NBS987 is 0.71034 ± 0.00026 (certificate issue date 19 June 2007); however, the quoted accepted values vary significantly (between circa 0.71024 to 0.710263, e.g., Irrgeher et al., 2016; Linderholm et al., 2020; Stein et al., 1997). In this study, the measurements were all normalised to an accepted value of 0.710240. For every sample batch the correction factor was calculated ($0.710240/^{87}Sr/^{86}Sr_{measured\ NBS}$) and applied to the sample batch. The procedural blanks contained on average 38 pg strontium (n = 9). The dataset were analysed using SPSS 25.0 (IBM SPSS Statistics for Macintosh, Armonk, IBM Corp.).

5.2.2. Concentration measurements

The acidified 60 tap water samples were analysed using a Thermo X-Series II Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The aliquot solutions were introduced via a quartz dual cyclonic spray chamber equipped with a PFA-ST MicroFlow nebulizer (Elemental Scientific) with a sample uptake rate of about 100 μl min^{-1}. Changes in sensitivity over time were monitored and corrected for using the geological reference material BHVO-2, which was analysed after every second sample (modified after Eggins et al., 1997). Sr concentrations were calculated using a two-point calibration of blank and BHVO-2. Long-term precision determined with multiple geological reference materials is about 10% (RSD).

5.2.3. Geospatial methods

In order to assess potential spatial autocorrelations in the tap water and human enamel $^{87}Sr/^{86}Sr$ ratios, Global Moran's I analyses were performed in ArcGIS (version 10.6.1). Furthermore, cross validations of the measured and predicted (i.e. values that are established by removing one or more data locations and predict their associated data using the data at the rest of the locations) values were performed using ArcGIS′ Cross Validation Geoprocessing Tool.

6. Results and discussion

6.1. Sr isotope variation

Results are presented in the Supplementary data (Tables S1 and S2) and summarised in Table 1. The 124 tap water samples were complemented with 19 analyses from a previous project (IDIS – Identification by Isotope Analysis in Font et al., 2015 – n_{total} = 143). In addition, the 94 dental enamel samples were complemented with 59 samples previously published (Font et al., 2015; Plomp et al., 2019; Plomp et al., 2020 - n_{total} = 153). For the 9 samples from Plomp et al. (2020) the $^{87}Sr/^{86}Sr$ ratios were selected from the lateral lingual enamel where possible (mesial of buccal side of the wall (WM/WB) where selected if lingual side (WL) was unavailable due to presence of carious lesions).

Table 1
Descriptive statistics for the human dental enamel, and tap water datasets.

Statistics	Tap water	Trimmed	Enamel	Trimmed
N	143	119	153	151
Mean $^{87}Sr/^{86}Sr$	0.70927	0.70902	0.70935	0.70936
Standard deviation (1σ)	0.00067	0.00029	0.00026	0.00024
Variance	0.00000	0.00000	0.00000	0.00000
Minimum	0.70837	0.70837	0.70847	0.70877
Maximum	0.71278	0.70970	0.70995	0.70995
Range	0.00441	0.00133	0.00148	0.00118
Median	0.70909	0.70898	0.70935	0.70938

The observed range of $^{87}Sr/^{86}Sr$ ratios was larger for tap water than for enamel in the Netherlands. For tap water, the range was 0.70837–0.71278 ($\Delta Sr_{max-min}$ = 0.004). Modern human enamel samples ranged from 0.70847 to 0.70995 ($\Delta Sr_{max-min}$ = 0.0015). Both datasets contained outliers, as assessed by inspection of a boxplot. Removal of the outliers did not result in normally distributed datasets, as assessed by Shapiro-Wilk's test ($p < 0.05$). Therefore, Mann-Whitney *U* test was run to determine if there were differences in $^{87}Sr/^{86}Sr$ ratios between dental enamel and tap water. Median $^{87}Sr/^{86}Sr$ ratios were statistically significantly different between the two sample types (U = 14,758, z = −5.189, p = 0.000). This difference may be translated into the expectation that tap water does not control the Sr isotope composition of human dental enamel.

6.2. Tap water

The distribution of the tap water $^{87}Sr/^{86}Sr$ ratios is displayed in Fig. 1B. Multiple samples from one city or village show variable, but overall limited differences in Sr isotope composition, ΔSr ranging between 0.000003 and 0.000445 (see Supplementary data, Table S3). Little variation is seen in tap water from Waternet (n = 3, all 0.7089), PWN (n = 7, 0.7088–0.7089, $\Delta Sr_{max-min}$ = 0.0001), Dunea (n = 9, 0.7089–0.7094 $\Delta Sr_{max-min}$ = 0.0005), Oasen (n = 9, 0.7088–0.7093, $\Delta Sr_{max-min}$ = 0.0005), Waterbedrijf Groningen (n = 8, 0.7086–0.7092, ΔSr = 0.0006), and Evides Waterbedrijf (n = 7, 0.7088–0.7096, $\Delta Sr_{max-min}$ = 0.0008). Except for Waterbedrijf Groningen, the water companies mentioned above almost exclusively rely on surface water, riverbank filtration and infiltration abstraction sites (KWR, 2017) that are situated in an underlying geology that is dominated by Holocene river and coastal sediments, explaining the limited variation in observed $^{87}Sr/^{86}Sr$ ratios. The groundwater abstraction sites in Noord-Brabant, operated by Brabant Water, sample waters from the thick layers of clay, sand, and loam (brabantwater.nl) that are known to exhibit a wide range of $^{87}Sr/^{86}Sr$ ratios. This is reflected in the observed tap water isotope compositions, ranging from 0.7089–0.7098 (n = 12, ΔSr = 0.0009).

The largest variations in tap water $^{87}Sr/^{86}Sr$ ratios are seen in water managed by WMD Drinkwater (n = 8, 0.7086–0.7103, $\Delta Sr_{max-min}$ = 0.0017), Vitens (n = 53, 0.7084–0.7106, $\Delta Sr_{max-min}$ = 0.0023), and WML (n = 27, 0.7084–0.7128, $\Delta Sr_{max-min}$ = 0.0044). All WMD Drinkwater groundwater stations abstract water from coarse sand and Pleistocene boulder clay layers, which are expected to exhibit more radiogenic Sr isotope ratios. This is reflected in the observed tap water $^{87}Sr/^{86}Sr$ ratios. However, some WMD Drinkwater samples exhibit low Sr isotope ratios (0.7086). Vitens covers almost 50% of the Netherlands, and therefore abstracts groundwater from all types of sediments (Holocene clays – Pleistocene loam) and consequently distribute tap water with varying Sr isotope ratios. For example, tap water from Zwolle (Overijssel), is abstracted at a depth of circa 120–160 m from a local groundwater source (*Engelse Werk* - vitens.nl). These sediments belong to the Upper North Sea Group that are expected to exhibit Sr isotope ratios comparable to seawater (0.7091, TNO-NITG, 2004; Veizer, 1989). Kampen is located 15 km west of Zwolle. Here, tap water is extracted about 15 km south of Zwolle from a push moraine (Veluwe). This ice push ridge consists of radiogenic Pleistocene sediments, which is reflected in the high $^{87}Sr/^{86}Sr$ ratios of the Kampen tap water samples (n = 2, 0.7104). The most complex tap water abstraction and distribution system is found in the province of Limburg. Hence, tap water represents a mix of groundwater, riverbank filtration, infiltration and surface water. This diversity, in combination with the various types of sediments present (e.g., Holocene fluviatile sediments, Pleistocene loess) results in a wide range of observed tap water $^{87}Sr/^{86}Sr$ ratios (i.e., 0.7084–0.7128).

Based on the observed spatial patterning, tap water $^{87}Sr/^{86}Sr$ ratios correlate with the local geology of the abstraction site. These results

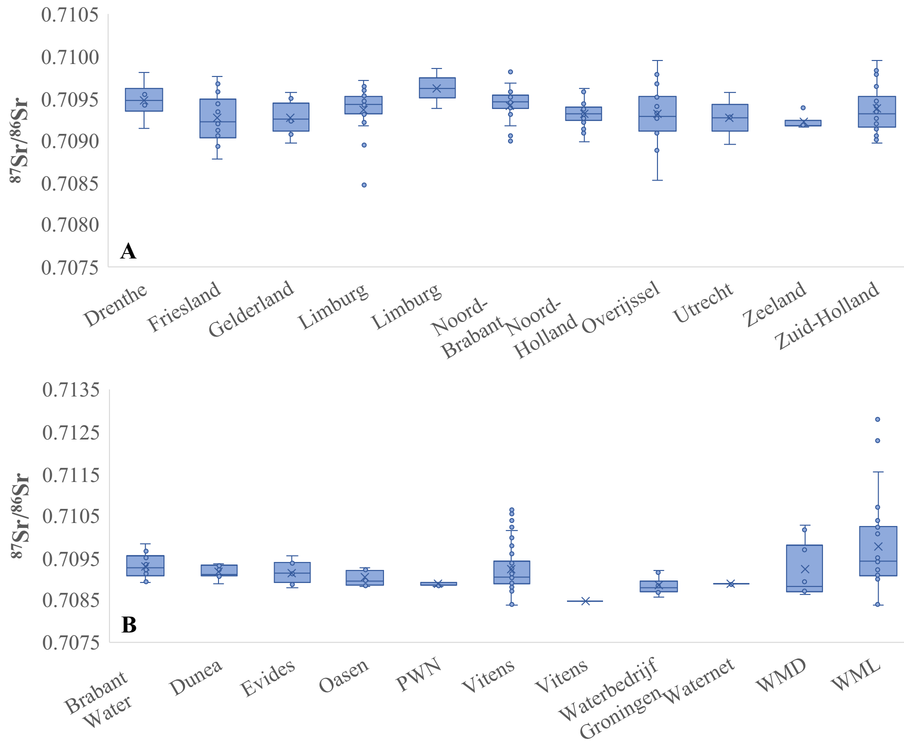

Fig. 1. Tukey's schematic box and whisker showing A) the variation in modern human dental enamel between the different provinces of origin (n = 153), and B) the variation in tap water $^{87}Sr/^{86}Sr$ between the 10 water companies in the Netherlands (n = 143). Key: the boxes represent the interquartile range (IQR: Q3–Q1), the central line indicates the median. The whiskers represent Q1–1.5 IQR and Q3 + 1.5 IQR. The circles represent individual data points.

are consistent with previous research (Kamenov and Curtis, 2017; Montgomery et al., 2006; Voerkelius et al., 2010).

6.3. Human dental enamel

The distribution of the modern human dental enamel $^{87}Sr/^{86}Sr$ ratios is displayed in Fig. 1A. The $^{87}Sr/^{86}Sr$ ratios range between 0.7085 and 0.7100 (n = 153). The greatest variance in human dental enamel $^{87}Sr/^{86}Sr$ ratios is seen in the samples from the provinces of Overijssel (n = 17, 0.7085–0.7100, $\Delta Sr_{max-min}$ = 0.0014), Limburg (n = 23, 0.7085–0.7099, $\Delta Sr_{max-min}$ = 0.0014), Friesland (n = 19, 0.7088–0.7098, $\Delta Sr_{max-min}$ = 0.0010), and Zuid-Holland (n = 24, 0.7090–0.7099, $\Delta Sr_{max-min}$ = 0.0010, see Supplementary data Table S4). The observed variation therefore seems to be independent of the degree of urbanisation, of which the highest degrees are found in Noord-Holland, Zuid-Holland and Noord-Brabant (Vanham et al., 2016).

Two individuals from Almelo (Overijssel) and Heerlen (Limburg) exhibit low $^{87}Sr/^{86}Sr$ ratios (0.7085) that deviate significantly from the other values. In one molar (Almelo1) an amalgam filling was present. To date, no research has been executed to investigate the effect filling materials on the isotopic integrity of dental enamel. The effect of the carious lesion itself is considered of limited importance, as the biogenic $^{87}Sr/^{86}Sr$ ratio of the unaffected enamel is not overwritten during the remineralisation process (Plomp et al., 2020). The absence of dental fillings in the other individual (IDIS5, Heerlen) suggests that the isotopic composition reflects the dietary Sr intake. Nevertheless, the reasons behind these low $^{87}Sr/^{86}Sr$ ratios remain unclear. Although $^{87}Sr/^{86}Sr$ ratios of 0.7085 are significantly different from the mean (>1.5 ∗ IQR), this study does show that they occur within the modern Dutch population. Excluding them from the dataset could result in erroneous interpretations in which individuals are mistakenly defined as non-Dutch.

6.4. Tap water contribution to dietary Sr

The Sr concentrations in 60 tap water samples range from 0.035 to 0.357 ppm (μg/g), with a mean of 0.174 and a median of 0.157 ppm (Supplementary data Table S5). Based on a healthy intake of 2 l of local drinking water per day, tap water is expected to contribute between circa 70 to 720 μg (average 340 μg) strontium to the human dietary intake. Compared to the average Sr intake from the most important food sources in the Netherlands, the contribution of tap water is not expected to dominate the isotope composition of human dental enamel. For reference, the dietary Sr intake of an adult Canadian is estimated between 1337 and 1869 μg per day (CDW, 2018). Data from the average diet in the Netherlands is currently unavailable. However, a non-dominant contribution of local tap water to the strontium isotope composition of modern dental enamel is supported by the data displayed in Fig. 2. Human dental enamel and tap water samples were available from 24 locations. A Spearman's rank-order correlation test showed no statistically significant correlation between the tap water and dental enamel samples, $r_s(22) = 0.230$, $p = 0.279$. There is no evidence of major systematic changes in the water distribution network in the past 4 decades, hence, based on this dataset the conclusion can be drawn that the Sr isotope signature of the human dental enamel samples in the Netherlands are not controlled by the consumed tap water $^{87}Sr/^{86}Sr$ ratios.

6.5. Relation between [Sr] and $^{87}Sr/^{86}Sr$

The different water sources used for the production of tap water exhibit different levels of Mg and Ca. Hard water contains high levels of Mg and Ca, the degree of which is expressed in Deutsche Härte (dH).Water hardness varies per region. Sr and Ca contents are generally controlled by similar mineral systematics, hence Sr and Ca are highly correlated and waters high in calcium (>dH) are expected to contain higher

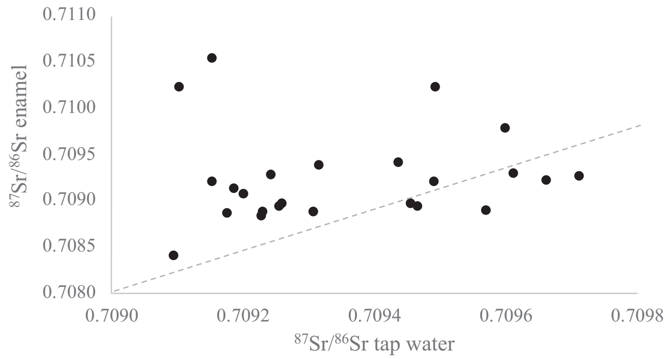

Fig. 2. Human dental enamel and tap water $^{87}Sr/^{86}Sr$ ratios from the same sample locations ($r_s(22) = 0.230$, $p = 0.279$). Dashed line represents a 1 to 1 relationship. For several sample locations multiple enamel samples were available and an average was taken to compare to the tap water values. Error bars (2SD) for those locations are smaller than the symbols. Details in Supplementary data Table S6.

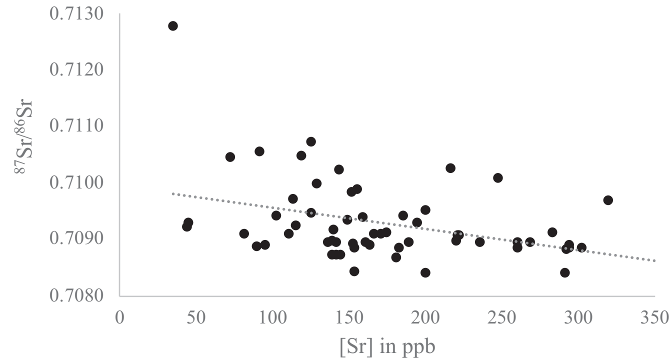

Fig. 4. $^{87}Sr/^{86}Sr$ ratios and strontium concentration data for 60 tap water samples. The weak negative correlation is statistically significant ($r_s(58) = -0.385$, $p = 0.002$).

concentrations of strontium. A Spearman's rank-order correlation was run to assess the relationship between [Sr] and tap water hardness. Tap water hardness was obtained for all 60 locations (waterhardheid.nl). If a range of dH values was given, the average dH values were included in the analysis. There was a statistically significant positive correlation between [Sr] and the water hardness ($r_s(58) = 0.420$, $p = 0.001$; Fig. 3). Accordingly, hard Dutch tap water is generally slightly higher in [Sr] than soft water. The contribution of tap water to dietary Sr might therefore be higher in regions with harder water (>dH). There is a statistically significant negative correlation between [Sr] and $^{87}Sr/^{86}Sr$ ratio ($r_s(58) = -0.385$, $p = 0.002$; Fig. 4). This translates into the tendency for tap water with low [Sr] to exhibit relatively high $^{87}Sr/^{86}Sr$ ratios. The data presented here, however, have too weak a correlation and the dataset is too small for use in a forensic context. Consequently, high human dental enamel $^{87}Sr/^{86}Sr$ ratios do not automatically exclude a provenance in regions with very hard waters. This is strengthened by the fact that there is a moderate negative, but statistically insignificant, correlation between tap water Sr content and dental enamel $^{87}Sr/^{86}Sr$ ratios from the same city (n = 12, Pearson's correlation, $r(10) = -0.32$, $p = 0.308$, see Supplementary data S7 and S8).

6.6. Implications for forensic provenancing

The $^{87}Sr/^{86}Sr$ ratios for the tap water and the human enamel samples were visualized in ArcGIS (Fig. 5A and B). The extreme outliers in the tap water dataset are clearly visible. The tap water $^{87}Sr/^{86}Sr$ ratios show that there is weak clustering when using all data (Moran's I = 0.323, z = 2.193, $p = 0.03$). However, when averaging the $^{87}Sr/^{86}Sr$ ratios from the same location, the data are not statistically different from randomly dispersed (Moran's I = 0.488, z = 1.270, $p = 0.204$). The Moran's I index for the human enamel $^{87}Sr/^{86}Sr$ ratios was −0.030 (z = −0.416, $p = 0.677$) and the cross validation showed no predictive strength. Both analyses could not be improved by using averaged $^{87}Sr/^{86}Sr$ ratios for the locations. The lack of spatial autocorrelation can be explained by the fact that $^{87}Sr/^{86}Sr$ ratios are supposed to be predominantly determined by the underlying geology. The human enamel data, however, seem unrelated to the geology, possibly due to the supermarket-effect. In contrast, tap water strontium isotope compositions are related to the regional geological characteristics. Therefore, more detailed isoscapes, following the model of Bataille et al. (2018), which include additional variables such as environment and geography, could be developed to predict tap water $^{87}Sr/^{86}Sr$ ratios for the Netherlands. However, due to the absence of correlation between human dental enamel and tap water $^{87}Sr/^{86}Sr$ ratios (Fig. 2), a predictive human enamel isoscape for the Netherlands for forensics research purposes seems unachievable. The lack of correlation between water and human enamel suggests that even if the water or human enamel database were expanded significantly (>*5) an accurate Sr isotope landscape map for the Netherlands cannot be established for forensics research purposes.

Published archaeological and modern biosphere isoscapes, including the one from the Netherlands, used specific samples that correlate with the underlying subsurface geology, i.e. small rodents, soil, and vegetation (e.g., Evans et al., 2010; Laffoon et al., 2012; Willmes et al., 2014; Kootker et al., 2016b). Although the consumed tap water in the Netherlands is related to the local geology, the modern human Sr intake appears to have become detached from the local geological strontium isotope composition probably due to the globalising diet and the import of food. Hence, the main principle behind the application of the strontium isotope system as a proxy for mobility appears to be invalid in this modern, densely populated society and possibly in similar densely populated cities and countries worldwide. Larger and less densely populated countries, with more complex geology, need to be examined to establish if human Sr isotope ratio variation is also dampened by globalisation of the food supply.

The effective application of Sr isotopes in modern populations for forensic provenancing studies therefore strongly depends on the existence of a database containing a large number of modern human enamel data from individuals with a known provenance history. Based on the extended database presented in this study, a $^{87}Sr/^{86}Sr$ range for the Netherlands is established, varying between 0.7085 and 0.7100 (n = 153), with 98.0% of the individuals (n = 151) ranging between 0.7088 and 0.7099. The established Dutch $^{87}Sr/^{86}Sr$ range can be used in forensic human identification cases to exclude a Dutch provenance or (recent) residence. Future research is required to assess whether local dietary plant samples provide a better proxy for human enamel Sr isotope ratios than tap water. Additional isotopic data on local and imported food resources may help to gain more insight into the

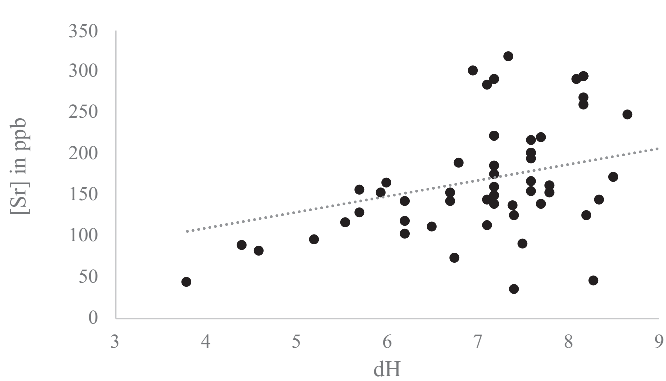

Fig. 3. Strontium concentration data and water hardness in Deutsche Härte (Dh) for 60 tap water samples. The weak positive correlation is statistically significant ($r_s(58) = 0.420$, $p = 0.001$).

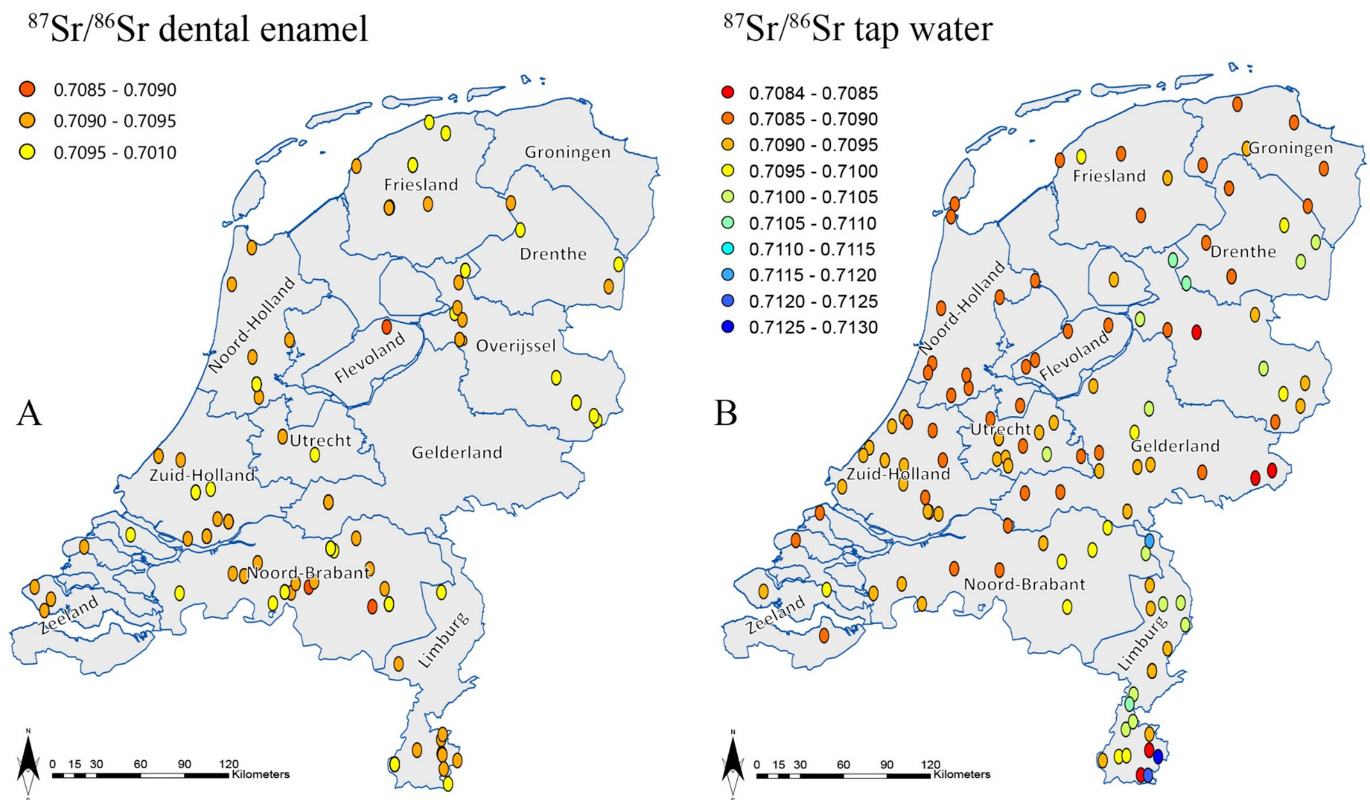

Fig. 5. A: Graphical distribution of modern human enamel $^{87}Sr/^{86}Sr$ ratios in the Netherlands. B: Graphical distribution of tap water $^{87}Sr/^{86}Sr$ ratios in the Netherlands. Samples from a single location with comparable Sr isotope compositions are depicted as a single symbol. Individual data in Supplementary data Tables S1 and S2.

observed limited spatial variation of the Sr isotope composition of human dental enamel in the Netherlands, allowing for a more accurate interpretation of forensic Sr isotope data.

7. Conclusion

The accurate interpretation of $^{87}Sr/^{86}Sr$ ratios in archaeological and forensic case studies requires sufficient background information, or a bioavailable baseline. For the successful application of strontium isotope analysis of human tissues, the dietary intake and the underlying local geology should be correlated. While this may be valid for archaeological studies, forensic provenancing studies have to address the challenge of a globalising supermarket-diet and its homogenising effect on human $^{87}Sr/^{86}Sr$ ratios. The current Sr isoscapes and baseline datasets are based on modern vegetation samples and/or archaeological faunal samples that reflect the strontium isotope composition of the geological subsurface. However, forensic isotope investigations require tailored baseline $^{87}Sr/^{86}Sr$ datasets and isoscapes as human isotopic signatures do not necessarily reflect the local geological values.

This study examined tap water and modern dental enamel with the aim to place constraints on the expected local strontium isotope signature in the Netherlands and its spatial variation. Tap water values (0.7084–0.7128, n = 143) were found to reflect the local geology, as they exhibited expected Sr isotope values, and outliers could be explained by divergent abstraction processes that were not influenced by the local geology. However, the tap water dataset does not correlate with the modern human dental enamel $^{87}Sr/^{86}Sr$ ratios, i.e., tap water is not dominating the dietary Sr intake. This is likely the result of the low Sr concentrations in Dutch tap water and the increasing internationalisation of the food distribution. The latter is especially important for the Netherlands, a country which depends heavily on the import of food. Hence, for forensic provenance investigations in the Netherlands, tap water should not be used as comparative reference material for modern dental enamel for Sr isotopes. Where possible, future studies should use human tissues to establish an isoscape to determine the range of expected isotopic ratios. The Dutch range in $^{87}Sr/^{86}Sr$ in human enamel is between 0.7085 and 0.7100 (n = 153), with 98.0% of the individuals ranging between 0.7088 and 0.7099 (n = 151). Although clear spatial variations in $^{87}Sr/^{86}Sr$ within the Netherlands are absent, the baseline data for the Netherlands are of great importance for forensic human identification cases using strontium isotopes as an investigative tool. Additional isotopic data on the bioavailable Sr from imported and local foods will allow better insights into the isotopic variation of modern human dental enamel. In combination with the application of a multi-isotope approach (Sr-Pb-C-H-O), such information is needed for more accurate isotopic baselines that are representatives of human isotopic variation. This will improve interpretations of modern isotopic data and the application of isotopic techniques to forensic case studies.

Funding

The research was supported by the European Research Council under the European Union Seventh Framework Programme (FP7/2007–2013)/ERC grant agreement no. 319209.

CRediT authorship contribution statement

Lisette M. Kootker: Conceptualization, Investigation, Methodology, Formal analysis, Writing - original draft, Visualization, Project administration. **Esther Plomp:** Conceptualization, Investigation, Methodology, Formal analysis, Writing - original draft, Project administration, Visualization. **Saskia T.M. Ammer:** Formal analysis, Visualization, Writing - original draft. **Vera Hoogland:** Investigation. **Gareth R. Davies:** Funding acquisition, Resources, Writing - review & editing.

Declaration of competing interest

The authors have declared that no competing interests exist.

Acknowledgements

This research would have been impossible without the donations of dental elements dental that were facilitated by the Amsterdam UMC, location VUmc, in Amsterdam (J. Baart and T. Forouzanfar); Ikazia Hospital and Erasmus Medical Centre in Rotterdam (J. Kwakman, G. van Beek, and E. Wolvius); Universitair Medisch Centrum Maastricht and Zuyderland (C. Wilkert and L. Poort); Medisch Centrum Leeuwarden and Tandzorg Steenwijk (J. de Visscher, D.S. Joffe, and W. Meijer); Scheper Ziekenhuis Emmen (D.P. Dietvorst); Medisch Spectrum Twente and Tandartsenpraktijk G.C. Muller (A.M. Ettema, A.-G. Westhoff, and F.C. Bakker); Tandheelkunde Eemland (M. Dorrepaal); Tandartspraktijk Strang (E. Veen); and the Elkerliek Ziekenhuis, Máxima Medisch Centrum, Jeroen Bosh Ziekenhuis, and AmphiA-Breda (G. Dicker, K. de Vries, C. Griëntschnig, G. Mensink, and J.C. de Folter). Special thanks to the donations provided by Jan Paul Klijntunte (Drenthe and South-Holland) as well as Pieter Korver (South-Holland). Gertjan Dicker and the Nederlands Tandartsenblad are thanked for advertising our request to collect extracted molars. We thank our colleagues for the collection of water samples: Fraukje Brouwer, Margot Daleman, Joyce van Dijk, Maaike de Haas, Kinie Esser, Anja Fisher, Michael Gress, Maarten Groenendijk, Stijn Heeren, Alice Knaf, Jason Laffoon, Stefanie Luginbuehl, Coen Nienaber, Godfried Scheijvens, Richard Smeets, Ward Teertstra, Vincent van der Veen, Bas van der Wagt, and Jan Wijbrans. Peter van Diepenbeek (NWL) is acknowledged for providing detailed insights into the water management in Limburg. We thank the anonymous reviewers for their constructive comments and suggestions.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.138992.

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