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The isotopic taphonomy of human remains

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Abstract

Isotopic analyses of O, H, C, N, Sr and Pb are being used for forensic investigations of human remains as indicators of geographic birthplace, residence and diet. Analysis of different tissues such as teeth for birthplace and hair for last residence and recent travel can provide a temporal context to an individual's life. However, the preservation of isotopic signatures during decomposition has not been systematically studied in a forensic context. In measurements of tooth enamel and hair from ten body donors to two human decomposition facilities, we demonstrate that light stable isotopes ($\delta^{18}O$ and $\delta^{13}C$) in the carbonate fraction and trace elements, $^{87}Sr/^{86}Sr$, and Pb isotopes are preserved during a year of either shallow burial or surface environmental exposure. Premolar and molar teeth are more consistent and predictive of geographic location than canine or incisor teeth, despite the fact that none of the teeth had any indication of taphonomic or diagenetic change from low rare earth elements and uranium concentrations. In hair from the same donors, elements that are structurally incorporated into keratin (O, H, C and N) are generally preserved, while trace elemental composition, $^{87}Sr/^{86}Sr$ and Pb isotopic signatures exchange with the local bioavailable soil pool. Despite best practices in leaching and cleaning protocols, we were unable to recover the perimortem Sr and Pb isotope composition of hair after environmental exposure longer than a few weeks or aqueous exposure. However, teeth and hair $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ isotopic analyses and $^{87}Sr/^{86}Sr$ and Pb isotopes in teeth remain useful indicators of geographic birthplace, residence and diet.

Introduction

As of 2017, there are 40,000 open cases of unidentified human remains in the United States.[1] Although DNA, fingerprints, and forensic anthropological profiles are available in many cases, these individuals remain nameless. Additional investigational leads are required to solve these cases, provide answers to their families and loved ones, and bring perpetrators to justice. Knowing where the individual lived and traveled, and what type of life he or she led, can be critical to focus investigative efforts. Isotopic analysis holds the promise of revealing just such clues and has led to identifications in several prominent cases (Meier-Augenstein and Fraser 2008, Ehleringer et al. 2010).

Oxygen and hydrogen isotopes are related to the hydrologic cycle and vary in known spatial patterns, while carbon and nitrogen isotopes are related to diet. Strontium isotopes are correlated to underlying soil signatures transmitted through the food we eat and the water we drink. Lead isotopes are also derived from soil, but can be strongly modified by anthropogenic activity and pollution. The spatial distribution of these isotopic landscapes ("isoscapes") for different elemental systems are decoupled, so they provide independent axes of information.

Elements are taken up in biological tissues with different turnover times; tooth enamel records prenatal and juvenile residence, bone records the last 5-10 years, and hair grows at a rate of approximately a centimeter per month. Combining multiple isotope systems and body tissues can elucidate a rich history for a person's geographic residence, diet, and pollution exposure.

Isotope analysis emerged out of geochemistry and has been employed in anthropology for several decades. It is now being applied in forensic casework to identify bodies or body parts (Meier-Augenstein and Fraser 2008, Ehleringer et al. 2010), but much of the foundational work is based on living humans (Ehleringer et al. 2008, Tipple et al. 2016) or bodies without premortem or perimortem baseline samples (Herrmann et al. 2015). Although successful in several case examples, the effect of exposure to an outdoor environment through postmortem changes had not been systematically studied in humans.

[1] https://www.identifyus.org/en, NamUs fact sheet, downloaded on July 19, 2017.

Objectives

The primary purpose of this research was: 1) to measure $\delta^{18}O$, $\delta^{2}H$, $\delta^{13}C$, $\delta^{15}N$, $\delta^{88}Sr$, $\delta^{44}Ca$, $^{87}Sr/^{86}Sr$, and Pb isotopes in teeth and hair samples of recently deceased human donors and compare perimortem samples to those after one year of environmental exposure and 2) to evaluate the accuracy of geolocation and dietary predictions with the known origins, travel, and lifestyle of individuals. The goal was to determine if taphonomic processes altered the perimortem signatures, and if both perimortem and postmortem isotope signatures gave accurate inferences about where the individuals were from and how they lived. In addition, we measured environmental samples of soil leachates, precipitation and tap water to characterize endmember contributions if perimortem isotopic signatures were not preserved.

Methods

Human body donors were enrolled at the Anthropology Research Facility (ARF) at the University of Tennessee, Knoxville (n=6) and the Forensic Anthropological Research Facility (FARF) at Texas State, San Marcos (n=5). Intake samples of hair, bone, and teeth were taken, and the donors were placed in an outdoor environment on the surface (n=7) or in a shallow grave (n=4) and allowed to decompose naturally for approximately one year. Hair was sampled at Accumulated Degree Hours (ADH) 250, 500, 1000 and 2000, the major taphonomic stages. At the end of the study period, the donors were recovered and additional samples of hair, teeth and bone were collected. Hair mats from an additional ten donors at FARF were compared to intake samples to increase sample size. Precipitation, tap water, well water, surface soil grab samples, and soil cores to a depth of 12" were also collected.

We measured $\delta^{13}C$ and $\delta^{18}O$ of carbonate in chemically cleaned tooth enamel and bone by gas bench IRMS. $\delta^{13}C_{VPDB}$ and $\delta^{18}O_{VPDB}$ of both carbonate and inorganic components of ground soil from the dried, powdered <2 mm fraction was measured by EA-IRMS. After chemical cleaning twice with a 3:1 chloroform: methanol mixture, hair was ground in a liquid nitrogen-cooled ball mill. $\delta^{13}C$ and $\delta^{15}N$ of hair were measured by EA-IRMS, and $\delta^{18}O$ and $\delta^{2}H$ were measured by TC-EA-IRMS after equilibration to local humidity in parallel with standards. All data was corrected to international scales using two-point normalization with frequent matrix-matched certified and in-house secondary standards run as unknowns.

Bioavailable components of soil were extracted with an overnight 1 M ammonium acetate leach. Hair samples were cleaned in chloroform-methanol mixture as described above, and then measured in three components whenever sufficient sample was available: bulk hair, a leachate solution from three sequential leaches by 0.1 M HCl, and the solid residual material. This protocol is designed to separate the endogenous and exogenous trace elements and strontium (Tipple et al. 2013). Major and trace elements of all samples were analyzed by Q-ICPMS using validated methods. $^{87}Sr/^{86}Sr$ and Pb isotopes in tooth enamel, bone, hair, precipitation, tap water and bioavailable soil leaches were measured by MC-ICPMS after ion chromatographic separation of the element of interest. Full measurement details of analyses are available in the Department of Justice technical report currently being prepared.

Results

Tooth enamel

There is no systematic variation in $\delta^{13}C$ between intake and recovery tooth enamel samples. There is some variation in $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ in tooth enamel, but is more strongly related to dental element (canine / incisor versus premolar / molar) than to exposure condition (surface vs burial) or exposure time (intake vs recovery). More than 75% of the teeth were within 3.5‰ of the expected tap water smoothed value of Bowen et al. (2007), after conversion using the equation of Ehleringer et al. (2009), which is small relative to the estimated 22.5‰ range of tap water values in the US. All of the teeth deviating from the model values >3.5 ‰ were canine or incisors.

Donors had a wide range of birthplaces, including Minnesota, Michigan, California, Georgia, and Texas. The $^{87}Sr/^{86}Sr$ values in tooth enamel from 0.709-0.711, much less than either bedrock maps or average modeled water values for catchments (Bataille and Bowen 2012). The variations in Pb isotopes are far larger between individuals than within dental elements.

Hair

The $\delta^{13}C$, $\delta^{18}O$, and $\delta^{2}H$ hair values are to first order preserved through environmental exposure one year postmortem (Fig 1). $\delta^{15}N$ values systematically increase over time outside of analytical error or external sample reproducibility, although the amount of change is smaller than estimates of one trophic level (~3.5‰).

Both $\delta^{18}O$ and $\delta^{2}H$ in hair showed some variability outside of the external sample reproducibility during the course of the study period, but there was no systematic trend over time. The median change between recovery and intake

samples for δ^2H was -3.0 ±4.1‰ (1 sd, n=14) and for $\delta^{18}O$ was -0.1 ±1.0‰ (1 sd, n=14). The δ^2H offset between intake and recovery samples is consistent with offsets due to freezing (Gordon et al. 2016).

Using hair isotope prediction maps presented in Ehleringer et al. (2008), 15 of 19 of the $\delta^{18}O$ values of intake samples (79%) were within 1‰ of model predictions. Using a more detailed body water conversion calculation (Ehleringer et al. 2008) to convert measured hair values to local tap water did not substantially improve the model's predictions. There were significant differences between the measured δ^2H_{VSMOW} of hair with that of the Ehleringer prediction map, consistent with the normalization issues discussed in Coplen and Qi (2012).

By contrast, $^{87}Sr/^{86}Sr$ and Pb isotopes in hair are poorly preserved. Changes in $^{87}Sr/^{86}Sr$ strongly indicates exchange with the soil bioavailable pool (Fig 1).

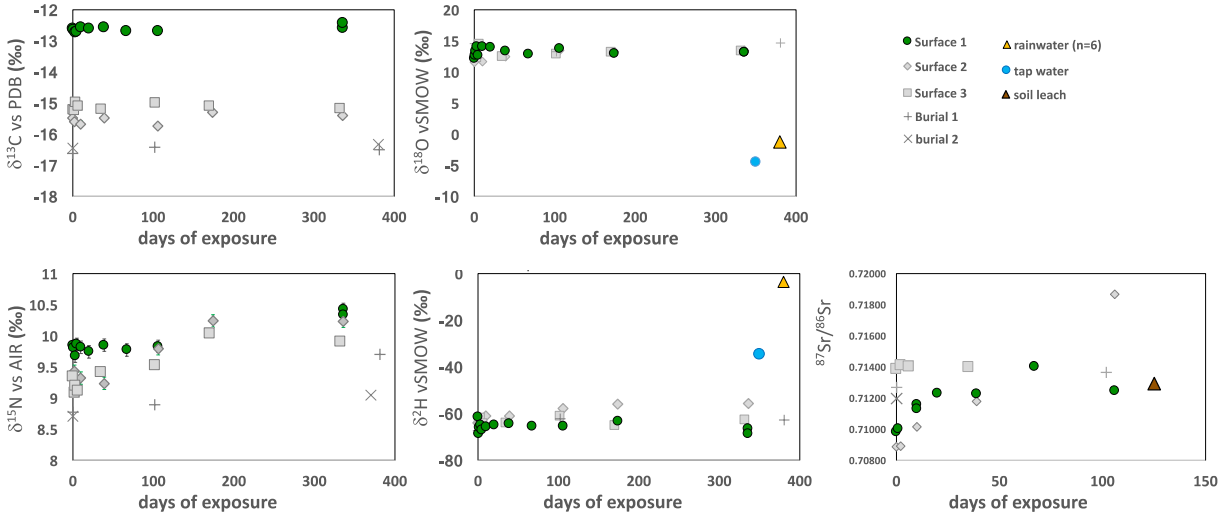

Fig. 1. Variation in isotopes of hair in Tennessee Anthropology Research Facility donors over the course of one year of outdoor exposure. For clarity, one individual is highlighted in green, but trends are typical among donors. Potential mixing endmembers from the local environment are also shown. $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$ and δ^2H are from bulk hair. When possible, $^{87}Sr/^{86}Sr$ is from solid residual fraction. Additional donors are shown to illustrate donor range.

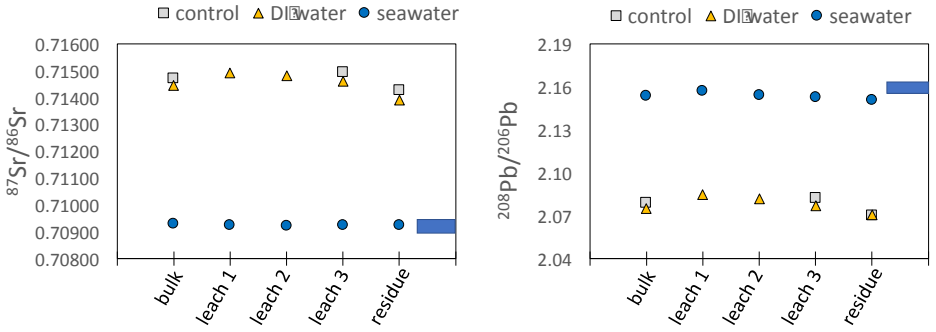

Fig. 2. Measured Sr and Pb isotopes in two hair samples from a single donor. Samples were submerged in either deionized water or lead-spiked seawater for three days and then cleaned as normal. The control samples had no aqueous exposure. The blue bars show the measured values of the seawater solution. The three sequential leaches use 0.1 M HCl (Tipple et al 2016).

To further evaluate Sr and Pb preservation, we submerged hair samples from one donor for three days in two solutions, deionized water and a lead-spiked seawater solution. The $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$, and δ^2H values were all the same within analytical error, despite the fact that the two endmember solutions were isotopically distinct. However, Sr and Pb isotope values of the solid residual were similar to the parent solution, despite using an aggressive leaching method proposed to remove exogenous Sr from hair (Tipple et al. 2013, Fig 2). The solid residual fraction of both the deionized water and seawater exposed samples had similar concentrations of most major and trace elements.

Discussion

There was no systematic variability in light stable and radiogenic isotope ratios in teeth linked to environmental exposure up to one year. Canine and incisor teeth showed a larger $\delta^{18}O$ range than premolars and molars, and $\delta^{18}O$ measurements had a greater discrepancy from either tap water or precipitation model estimates. Strontium isotope measurements of tooth enamel had a much narrower range of values than that predicted for $^{87}Sr/^{86}Sr$ by isoscape models of the US for bedrock (±carbonate), local water, or flux-weighted catchment water. A flux-weighted catchment water model averaged over watersheds (Bataille and Bowen 2012) showed the best agreement with the measurements, but overall $^{87}Sr/^{86}Sr$ was a less specific indicator of geographic origin than $\delta^{18}O$.

Hair analyses showed a more complicated story for both preservation and prediction. $\delta^{13}C$ values were well preserved over time, while $\delta^{15}N$ values showed increases of up to 1‰ over a year of exposure, with increases starting after the first month. This amount of change would not have changed the trophic level interpretation of these individuals. However, it is unclear if the increase in $\delta^{15}N$ continues beyond the study period. At both sites and in both burial and surface placements, these changes would bias dietary interpretations to suggest more animal or fish protein was included in the diet. It is currently unclear what the controlling parameters on these changes are, but this could have important implications for anthropologic dietary interpretations.

Both Sr and Pb isotopes in hair changed over time in burial and surface environments. We were unable to recover perimortem isotope compositions, despite aggressive leaching. Similar protocols for sequential leaching of carbonates to remove diagenetic alteration are assumed to recover primary isotope compositions when sequential leaches approache a stable isotope value (Liu et al 2013). However, our submersion experiment demonstrates new criteria are needed to determine if pervasive resetting of the isotope composition and trace element concentrations occurred.

$\delta^{18}O$ and $\delta^{2}H$ hair values were preserved over one year, and $\delta^{18}O$ values generally correspond to the geographic region of last known residence. The predicted region of last residence depends strongly on details of the conversion of measured $\delta^{18}O$ in hair to $\delta^{18}O$ in tap water. Due to the significant taphonomic changes in hair, we recommend that Sr and Pb isotopes are not used in determining geographic origination of unknown human remains unless the remains are very recent and have been known not to have been exposed to groundwater or precipitation.

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