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RESEARCH ARTICLE

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Strontium isotope ratios of human hair from the United States: Patterns and aberrations

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Rationale: Strontium isotope ratios ($^{87}Sr/^{86}Sr$) of hair may be a valuable tool to estimate human provenance. However, the systematics and mechanisms controlling spatial variation in $^{87}Sr/^{86}Sr$ of modern human hair remain unclear. Here, we measure $^{87}Sr/^{86}Sr$ of hair specimens from across the USA to assess the presence of geospatial relationships.

Methods: Ninety-eight human hair specimens were collected from salon/barbershop floors in 48 municipalities throughout the conterminous USA. [Sr] and $^{87}Sr/^{86}Sr$ ratios were measured from hair using quadrupole and multi-collector inductively coupled plasma mass spectrometers, respectively. The [Sr] and $^{87}Sr/^{86}Sr$ ratios of hair were compared with the measured [Sr] and $^{87}Sr/^{86}Sr$ ratios of tap waters from the collection locations. In addition, the $^{87}Sr/^{86}Sr$ ratio of hair was compared with the modeled ratios of bedrock and surface waters.

Results: Hair color was independent of the $^{87}Sr/^{86}Sr$ ratio, but related to [Sr]. The $^{87}Sr/^{86}Sr$ ratios of hair and leachate were not statistically different and were positively correlated; however, in several hair-leachate pairs, the ratios were conspicuously different. The $^{87}Sr/^{86}Sr$ ratios of both hair and leachate were linearly correlated with tap water. The $^{87}Sr/^{86}Sr$ ratio of hair was also significantly correlated with the modeled ratio of bedrock and surface waters, although the $^{87}Sr/^{86}Sr$ ratio of hair was most strongly correlated with the measured ratio of tap water.

Conclusions: The $^{87}Sr/^{86}Sr$ ratio of hair is related to the ratio of tap water, which varied geographically. The ratio of hair provided geographic information about an individual's recent residence. Differences in the $^{87}Sr/^{86}Sr$ ratios of hair and hair leachate may be concomitant with travel and could potentially be used as a screening tool to identify recent movements.

1 | INTRODUCTION

Reconstructing the provenance of an individual is important to many anthropologic and forensic investigations.[1-4] Stable isotope ratios recorded in human scalp hair have utility in reconstruction of the movement histories of individuals.[5-9] This is because hair keratin and its isotopic signatures are recorders of an individual's environment.[10] The applications of isotope analysis of hair are far-reaching considering the common occurrence of human hair at archeological sites and crime scenes.

Strontium isotope ratios ($^{87}Sr/^{86}Sr$) of human hair hold promise as an estimator of provenance and geographic movements. The $^{87}Sr/^{86}Sr$ ratios of human hair are thought to be largely controlled by local geology and the underlying bedrock of a region,[11] with the ratio of geologic materials varying geographically as a function of age and the ratio of Rb to Sr concentrations in the rock or soil.[12] The

Sr-isotopic signatures of underlying geology are transferred to soils and waters through chemical and physical weathering and become incorporated into biological materials with negligible change in the $^{87}Sr/^{86}Sr$ ratio from the source into an organic tissue.[12]

Sr-isotopic analysis of biological materials has been successfully applied to many provenance questions in the fields of archaeology,[13-22] ecology,[23-25] food science,[26-29] and forensic science.[30-34] These prior studies indicate that the $^{87}Sr/^{86}Sr$ ratios of organic tissues reflect the ratios of water and diet during the time period(s) when the biological materials formed, with negligible isotopic offsets between the source and tissue.[12] In other words, the $^{87}Sr/^{86}Sr$ ratios of organic tissues directly reflect those of the Sr source(s). The lack of Sr-isotopic variation between tissues and source has been shown in a variety of different organic tissues, including antlers,[35] teeth and bone,[36-43] and leaves.[44,45] Exposed keratin tissues, such as hair and nails, can also be affected by Sr contamination from the external environment.[11,46,47] Once exposed to the environment and detached from the organism's metabolism, the Sr concentration within hair increases with exposure time, reflecting continuous inputs of dust and aerosol particles into the keratin matrix.[46,48] This externally sourced Sr becomes embedded within the keratin and is not easily removed by bathing or washing.[49] The incorporation of exogenous Sr has been shown to alter the biological $^{87}Sr/^{86}Sr$ ratio of hair, and, thus, the endogenous geospatial information recorded by the isotopic signature.[47,50,51]

While the initial studies of Sr isotopes in modern human hair for geographic provenancing are promising, the application of the $^{87}Sr/^{86}Sr$ ratio of hair to reconstruct an individual's travel history presents a unique set of analytical and theoretical challenges as humans significantly modify their environment through the use of transported agricultural products and waters. Thus, the tissues of individuals that consume food from the "global supermarket" and drink/bathe with waters transported from distant regions are expected to have isotope ratios inconsistent with local isotopic signatures. These geospatial disconnections imparted on an individual's environment are transferred to the individual's hair and may undermine straightforward interpretations of geospatial isotopic relationships and models. Here, we measured $^{87}Sr/^{86}Sr$ ratios of human hair specimens that were collected throughout the USA. We compared these ratios with the measured $^{87}Sr/^{86}Sr$ ratios of tap waters from the collection locations and modeled the $^{87}Sr/^{86}Sr$ ratios of bedrock and surface waters to assess whether or not Sr-isotopic analysis can be used to estimate the provenance of an individual.

2 | EXPERIMENTAL

2.1 | Hair sample collection

Ninety-eight hair samples were collected in 48 municipalities of the coterminous USA, located within 16 states (Figure 1, Table 1). Specimens of cut hair from individuals were collected by gloved hand once material had accumulated on the floor. Up to three salon/barbershops in each municipality were visited and each collected hair specimen was immediately placed into a separate paper envelope. Hair specimens were collected during summer 2004. No demographic, dietary, socioeconomic, or travel history

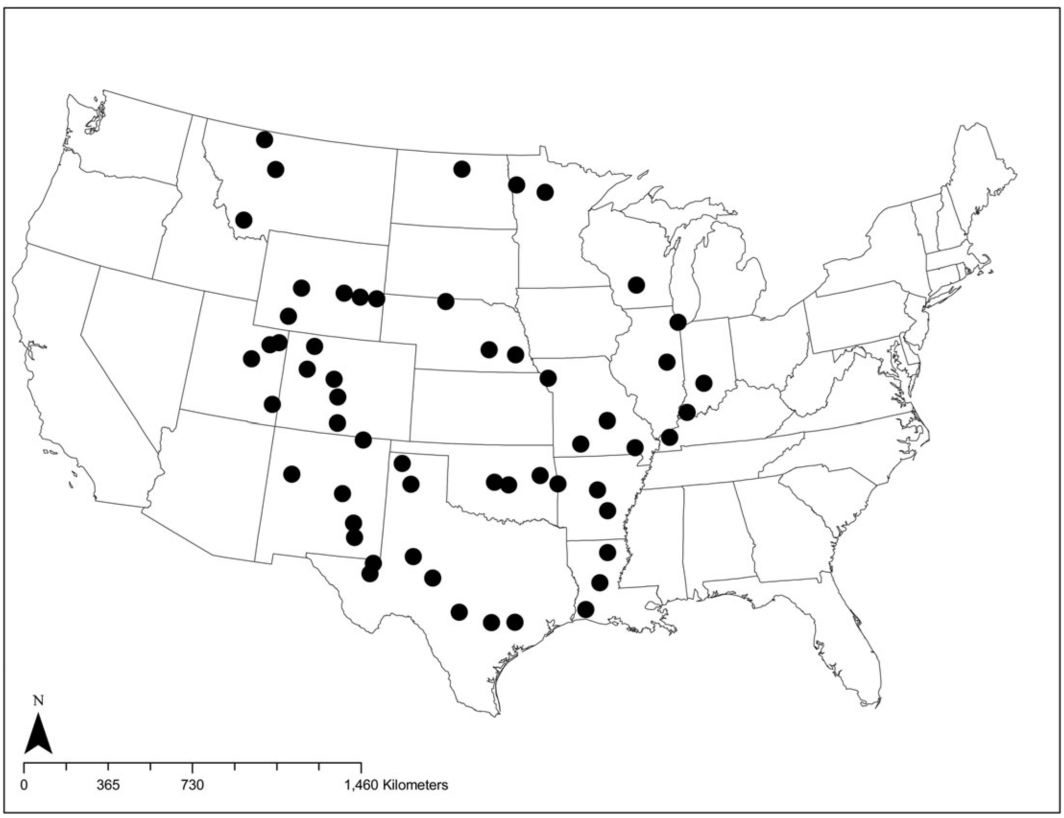

FIGURE 1 Hair and water sampling sites within the coterminous USA

TABLE 1 Hair specimen collection locations, colors, concentrations, and strontium isotope ratios

Identifier	City	State	Latitude	Longitude	Hair color	Hair [Sr] (μg g^{-1})	Hair $^{87}Sr/^{86}Sr$	Leachate $^{87}Sr/^{86}Sr$	$\Delta_{Hair-Leach}$
129	Price	UT	39.5987	−110.8223	Red	12.7	0.71083	0.71085	0.00002
130	Price	UT	39.5987	−110.8223	Brown	6.0	0.70931	0.70997	***0.00066***
132	Monticello	UT	37.8697	−109.3425	Brown	2.1	0.70943	0.70947	0.00004
133	Monticello	UT	37.8697	−109.3425	Brown	7.3	0.70821	0.70809	0.00012
135	Grants	NM	35.1506	−107.8508	Salt/pepper	3.4	0.71214	0.71197	0.00017
136	Grants	NM	35.1506	−107.8508	Black	14.3	0.70862	0.70898	***0.00036***
141	Vaughn	NM	34.5997	−105.2179	Brown	70.3	0.71224	0.71222	0.00001
142	Roswell	NM	33.4293	−104.5221	Salt/pepper	1.0	0.70807	0.71168	***0.00362***
143	Roswell	NM	33.4293	−104.5221	Salt/pepper	7.0	0.70821	0.70800	0.00021
145	Artesia	NM	32.8425	−104.4034	Red	10.9	0.70788	0.70788	0.00001
146	Artesia	NM	32.8425	−104.4034	Red	64.0	0.70760	0.70758	0.00002
148	Pecos	TX	31.4198	−103.4921	Salt/pepper	1.9	0.70862	0.70869	0.00007
149	Pecos	TX	31.4198	−103.4921	Brown	10.8	0.70861	0.70864	0.00003
150	Pecos	TX	31.4198	−103.4921	Black	9.1	0.70860	0.70854	0.00006
151	Odessa	TX	31.8552	−103.3747	Salt/pepper	10.3	0.70868	0.71794	***0.00926***
152	Odessa	TX	31.8552	−103.3747	Red	128.1	0.70862	0.70857	0.00004
154	Big Spring	TX	32.2500	−101.4786	Black	8.4	0.70874	0.70870	0.00004
155	Big Spring	TX	32.2500	−101.4786	Brown	88.8	0.70872	0.70873	0.00001
156	Big Spring	TX	32.2500	−101.4786	Salt/pepper	5.5	0.70886	0.70865	0.00021
157	San Angelo	TX	31.4255	−100.4965	Brown	-	0.70844	0.70847	0.00003
158	San Angelo	TX	31.4225	−100.4965	Salt/pepper	23.7	0.70894	0.70864	0.00030
163	Luling	TX	29.6818	−97.6494	Brown	1.1	0.70825	0.70820	0.00005
164	Luling	TX	29.6818	−97.6494	Brown	15.5	0.70827	0.70816	0.00011
166	Columbus	TX	29.7060	−96.5468	Brown	51.9	0.70817	0.70815	0.00002
167	Columbus	TX	29.7060	−96.5468	Brown	86.9	0.70779	0.70790	0.00010
169	Lake Charles	LA	30.1878	−93.2183	Salt/pepper	1.1	0.70864	0.70922	***0.00058***
170	Lake Charles	LA	30.1878	−93.2183	Salt/pepper	6.4	0.70844	0.70844	0.00000
172	Alexandria	LA	31.2599	−92.4982	Salt/pepper	10.7	0.70861	0.70860	0.00001
173	Alexandria	LA	31.2599	−92.4928	Brown	23.9	0.70849	0.70847	0.00002
177	Monroe	LA	32.4873	−92.0809	Black	1.7	0.70942	0.70907	***0.00035***
178	Monroe	LA	32.4873	−92.0809	Salt/pepper	11.9	0.71015	0.71010	0.00005
179	Pine Bluff	AR	34.2139	−91.9806	Black	27.7	0.70916	0.70912	0.00004
181	Pine Bluff	AR	34.2139	−91.9806	Brown	3.1	0.70915	0.70907	0.00008
182	Conway	AR	35.0909	−92.4372	Salt/pepper	0.3	0.71278	0.71274	0.00003
183	Conway	AR	35.0909	−92.4372	Brown	5.8	0.71320	0.71395	***0.00075***
186	Fort Smith	AR	35.3850	−94.4221	Brown	1.6	0.71123	0.71078	***0.00045***
188	Muskogee	OK	35.7411	−95.3077	Brown	12.6	0.70945	0.70957	0.00012
190	Muskogee	OK	35.7411	−95.3077	Salt/pepper	2.4	0.70974	0.70976	0.00002
191	Shawnee	OK	35.3500	−96.9116	Brown	14.1	0.71238	0.71240	0.00002
192	Shawnee	OK	35.3500	−96.9116	Brown	17.7	0.71227	0.71217	0.00010
194	Sayre	OK	35.4572	−97.6182	Black	18.9	0.70865	0.70866	0.00001
195	Sayre	OK	35.4572	−97.6182	Salt/pepper	8.6	0.70898	0.70895	0.00003
198	Amarillo	TX	35.2219	−101.8308	Brown	64.6	0.70916	0.70917	0.00002
199	Amarillo	TX	35.2219	−101.8308	Brown	92.7	0.70920	0.70918	0.00002
200	Dalhart	TX	36.0606	−102.3475	Red	13.0	0.70884	0.70891	0.00007
201	Dalhart	TX	36.0606	−102.3475	Brown	49.0	0.70900	0.70891	0.00008
203	Raton	NM	36.8926	−104.4409	Brown	13.6	0.70948	0.70951	0.00003
204	Raton	NM	36.8926	−104.4409	Red	10.3	0.70982	0.70996	0.00015
207	Alamosa	CO	37.4675	−105.8527	Salt/pepper	2.9	0.71469	0.70878	***0.00590***
208	Alamosa	CO	37.4675	−105.8527	Salt/pepper	5.4	0.70936	0.70937	0.00001

(Continues)

TABLE 1 (Continued)

Identifier	City	State	Latitude	Longitude	Hair color	Hair [Sr] (μg g^{-1})	Hair $^{87}Sr/^{86}Sr$	Leachate $^{87}Sr/^{86}Sr$	$\Delta_{Hair\text{-}Leach}$
209	Alamosa	CO	37.4675	−105.8527	Salt/pepper	5.1	0.70931	0.70911	0.00020
211	Salida	CO	38.5362	−105.9917	Brown	6.0	0.71350	0.71340	0.00010
212	Salida	CO	38.5362	−105.9917	Brown	2.4	0.71395	0.71371	0.00024
213	Leadville	CO	39.2487	−106.2921	Salt/pepper	0.7	0.71260	0.71403	***0.00143***
214	Leadville	CO	39.2487	−106.2921	Brown	4.1	0.71962	0.71918	***0.00044***
215	Leadville	CO	39.2487	−106.2921	Brown	3.3	0.71277	0.71225	***0.00052***
218	Rifle	CO	39.5333	−107.7830	Brown	9.9	0.71096	0.71098	0.00002
219	Rifle	CO	39.5333	−107.7830	Brown	30.0	0.71098	0.71094	0.00004
220	Rifle	CO	39.5333	−107.7830	Brown	9.2	0.71103	0.71098	0.00004
221	Craig	CO	40.5127	−107.5534	Red	14.1	0.71092	0.71044	***0.00048***
224	Vernal	UT	40.4496	−109.5024	Blond	12.4	0.71450	0.71454	0.00004
225	Vernal	UT	40.4496	−109.5024	Blond	12.1	0.71384	0.71378	0.00006
260	Rugby	ND	48.3686	−99.9956	Brown	0.3	0.70813	0.70816	0.00003
261	Crookston	MN	47.7812	−96.6102	Brown	0.9	0.71640	0.71636	0.00004
263	Crookston	MN	47.7812	−96.6102	Salt/pepper	0.2	0.71653	0.71587	***0.00066***
264	Bemidji	MN	47.4717	−94.8809	Red	2.3	0.71522	0.71518	0.00004
265	Bemidji	MN	47.4717	−94.8809	Brown	0.6	0.71410	0.71413	0.00003
266	Bemidji	MN	47.4717	−94.8809	Salt/pepper	0.8	0.71235	0.71238	0.00003
278	Baraboo	WI	43.4700	−89.7415	Salt/pepper	0.4	0.71077	0.70966	***0.00112***
282	Chicago	IL	41.7915	−87.5936	Black	0.9	0.70933	0.70939	0.00006
283	Chicago	IL	41.7915	−87.5936	Brown	10.1	0.71005	0.70994	0.00010
284	Chicago	IL	41.7915	−87.5936	Salt/pepper	0.4	0.71024	0.70988	***0.00036***
285	Mahomet	IL	40.1883	−88.4000	Brown	3.5	0.70934	0.70928	0.00005
293	Evansville	IN	38.0210	−87.5711	Brown	1.6	0.71045	0.71056	0.00011
296	Paducah	KY	37.0851	−88.5985	Brown	0.2	0.70980	0.70972	0.00009
304	Ozark	MO	37.0237	−93.2060	Salt/pepper	0.2	0.71025	0.71022	0.00002
305	Ozark	MO	37.0237	−93.2060	Brown	0.5	0.70996	0.70976	0.00019
306	St. Joseph	MO	39.7554	−94.8365	Brown	8.4	0.70901	0.70899	0.00002
307	St. Joseph	MO	39.7554	−94.8365	Brown	1.2	0.70915	0.70903	0.00011
309	St. Joseph	MO	39.7554	−94.8365	Red	2.6	0.70894	0.70902	0.00008
310	Lincoln	NE	40.7356	−96.5999	Blond	7.7	0.71013	0.71033	0.00020
315	Grand Island	NE	40.9302	−98.0474	Brown	4.2	0.71184	0.71183	0.00001
316	Grand Island	NE	40.9302	−98.0471	Brown	12.3	0.71167	0.71164	0.00003
319	Grand Island	NE	40.9302	−98.0471	Brown	5.0	0.71184	0.71183	0.00001
320	Valentine	NE	42.8725	−100.5493	Red	4.1	0.70962	0.70963	0.00001
321	Valentine	NE	42.8725	−100.5493	Brown	13.4	0.70975	0.70968	0.00007
328	Lusk	WY	42.7622	−104.4517	Brown	9.2	0.71024	0.71035	0.00011
330	Douglas	WY	42.7596	−105.3854	Brown	1.7	0.71227	0.71258	0.00032
334	Casper	WY	42.8492	−106.3023	Brown	7.6	0.71220	0.71216	0.00004
335	Casper	WY	42.8492	−106.3023	Blond	4.3	0.71202	0.71212	0.00010
336	Casper	WY	42.8492	−106.3023	Brown	1.6	0.71223	0.71215	0.00008
337	Casper	WY	42.8492	−106.3023	Blond	1.5	0.71153	0.71178	0.00025
341	Lander	WY	42.8331	−108.7300	Brown	6.2	0.71411	0.71413	0.00002
342	Lander	WY	42.8331	−108.7300	Brown	3.5	0.71772	0.70795	***0.00978***
344	Rock Springs	WY	41.5907	−109.2193	Brown	3.1	0.71124	0.71125	0.00001
345	Rock Springs	WY	41.5907	−109.2193	Red	27.0	0.71126	0.71125	0.00000
346	Rock Springs	WY	41.5907	−109.2193	Brown	14.7	0.71131	0.71130	0.00001
347	Rock Springs	WY	41.5907	−109.2193	Red	27.0	0.71119	0.71118	0.00001

Difference ($\Delta_{Hair\text{-}Leach}$) was calculated as the absolute $^{87}Sr/^{86}Sr$ ratio of leached hair minus the $^{87}Sr/^{86}Sr$ ratio of leachate.

information was obtained from the individuals from whom the cut hair specimens originated. Our initial assumption was that these individuals were residents of the municipality in which the specimens were collected, did not travel widely, and that they had no unique dietary patterns. Hair color was noted (Table 1). The hair specimens varied in texture and thickness, with lengths from 5 to 15 cm. The envelopes were stored at room temperature and in the dark from the date of collection.

2.2 | Water sample collection

Tap water samples were collected at the same time as the hair specimens described above. The majority of tap water samples used in this study were previously analyzed and reported in Chesson et al[52] (Table 2). For some municipalities, we analyzed additional tap water samples that were collected during the initial 2004 sampling effort (Table 3). In addition, several municipalities were revisited and additional tap water samples were collected from the same locations as presented by Chesson et al[52] (Table 3).

2.3 | Cleaning

Hair specimens were rinsed with ultrapure water. Following the International Atomic Energy Agency (IAEA)-recommended[53] cleaning method with subsequent modifications by Tipple et al,[54] approximately 50 mg of each hair specimen was placed into a 15-mL centrifuge tube and sequentially sonicated for 10 min in 3 mL acetone (HPLC-grade, EMD; Darmstadt, Germany), ultrapure water, acetone again, and finally 0.1 M hydrochloric acid (Aristar® Plus, BDH® Chemicals, VWR, Radnor, PA, USA). After each 10-min sonication, the solute was decanted into another centrifuge tube, with all decanted solutes combined into a single centrifuge tube and labeled as the “leachate”. The ultrapure water was purified using a Milli-Q Academic A10® system (EMD Millipore, Billerica, MA, USA) with a resistivity >18 Ωm.

Leached hair specimens were allowed to dry at room temperature for 72 h within a laminar flow hood. After drying, the leached hair specimens were stored in clean centrifuge tubes until preparation for isotope analysis.

2.4 | Sample preparation for Sr-isotopic analysis

The leached hair specimens were digested using an Ethos EZ® microwave digestion system (Milestone, Inc., Shelton, CT, USA) following Tipple et al.[54] A certified reference material (TORT-2, Lobster Hepatopancreas Reference Material for Trace Metals, National Research Council, Ottawa, Ontario, Canada) and a method blank were digested along with the hair specimens under identical conditions.

2.5 | Strontium abundance analysis

All Sr elemental abundances were measured via inductively coupled plasma quadrupole-mass spectrometry (ICP-MS; Agilent 7500ce; Agilent Technologies, Santa Clara, CA, USA) following Tipple et al.[54] A standard reference solution T-205 (USGS, Reston, VA, USA) was measured as an external calibration standard at least five times within each analytical sequence. The long-term reproducibility for T-205 and differences relative to the accepted value indicated that the Sr concentrations of hair specimens were accurate to within 10%. The secondary reference material was TORT-2, which has a certified [Sr] of $45.2 \pm 1.9\ \mu g\ g^{-1}$ (2σ) and the measured [Sr] of TORT-2 was $52.4 \pm 9.6\ \mu g\ g^{-1}$ (1σ, $n = 7$).

2.6 | Sr-isotopic analysis

All Sr-isotope ratio measurements were performed using a Neptune *Plus* multi-collector inductively coupled plasma mass spectrometer (Thermo Fisher Scientific, Bremen, Germany). Samples were introduced using an online Sr purification method following Tipple et al[54] for hair digest and leachate and Chesson et al[52] for water. Each sample analysis was followed by a blank to monitor the efficiency of the crown ether Sr resin column for Sr extraction. The $^{87}Sr/^{86}Sr$ ratios of samples and reference materials were blank- and interference-corrected, and then normalized for instrumental mass discrimination using a defined $^{86}Sr/^{88}Sr$ ratio of 0.1194.

The samples were analyzed for $^{87}Sr/^{86}Sr$ alongside sets of reference materials and blanks. The reference materials included one primary reference and a secondary reference to monitor measurement accuracy and reproducibility. The primary reference material was SRM® 987 (0.71034 ± 0.00026 [95% CI]; National Institute of Standards and Technology, Gaithersburg, MD, USA) and the secondary reference material was TORT-2. The samples and SRM® 987 were analyzed at a ratio of 5:1. The measured $^{87}Sr/^{86}Sr$ ratios of SRM® 987 and TORT-2 were 0.71030 ± 0.00002 (1σ, $n = 132$) and 0.70926 ± 0.00002 (1σ, $n = 3$), respectively.

2.7 | Statistical and spatiotemporal analysis

Statistical analysis was completed using JMP® Pro 13 (SAS, Cary, NC, USA) for Mac OS X. The measured and modeled [Sr] and $^{87}Sr/^{86}Sr$ ratios were compared using paired t-tests. P-values were considered significant at the $\alpha = 0.05$ level. Spatial mapping and analysis were carried out using ArcGIS 10.4 (ESRI, Redlands, CA, USA). GIS data layers for the $^{87}Sr/^{86}Sr$ ratios of bedrock and surface waters were from Bataille and Bowen[55] and developed from maps presented in Beard and Johnson[12] by West et al.[30] The GIS data layers from Bataille and Bowen[55] are publicly available (waterisotopes.org).

3 | RESULTS AND DISCUSSION

3.1 | Strontium abundance in hair

The average [Sr] of hair was $14.0 \pm 22.6\ \mu g\ g^{-1}$ (1σ, $n = 97$) and the concentration ranged from 0.2 to $128.1\ \mu g\ g^{-1}$ (Table 1). Some Sr abundances measured in this study were elevated relative to previously reported [Sr] of hair.[51,56-62] However, most previous studies reported [Sr] from hair sampled at the nape of the neck and

TABLE 2 Water sample collection locations, measured Sr-isotope ratios and concentrations of tap water, and modeled Sr-isotope ratios of bedrock or surface water

City	State	Latitude	Longitude	Tap water[1] $^{87}Sr/^{86}Sr$	Tap water[1] [Sr] (μg g^{-1})	Bedrock[2] $^{87}Sr/^{86}Sr$	Bedrock[3] $^{87}Sr/^{86}Sr$	Local water[3] $^{87}Sr/^{86}Sr$	Catchment[3] $^{87}Sr/^{86}Sr$	HUC 8[3] $^{87}Sr/^{86}Sr$
Price	UT	39.599	−110.822	0.7112	251.0	0.7059	0.7139	0.7134	0.7085	0.7081
Monticello	UT	37.870	−109.343	0.7082	116.3	0.7059	0.7113	0.7113	0.7126	0.7081
Grants	NM	35.151	−107.851	0.7134	691.1	0.7080	0.7113	0.7113	0.7081	0.7078
Vaughn	NM	34.600	−105.218	0.7123	713.4	0.7080	0.7115	0.7115	0.7089	0.7089
Roswell	NM	33.429	−104.522	0.7077	934.4	0.7049	0.7113	0.7113	0.7084	0.7083
Artesia	NM	32.843	−104.403	0.7098[#]	6.8	0.7049	0.7113	0.7113	0.7089	0.7084
Pecos	TX	31.420	−103.492	0.7086	1326.5	0.7049	0.7113	0.7113	0.7086	0.7084
Odessa	TX	31.855	−103.375	0.7086	1725.6	0.7049	0.7113	0.7113	0.7090	0.7089
Big Spring	TX	32.250	−101.479	0.7087	1513.3	0.7059	0.7114	0.7114	0.7078	0.7084
San Angelo	TX	31.423	−100.497	0.7085	1903.1	0.7080	0.7113	0.7113	0.7075	0.7075
Luling	TX	29.682	−97.649	0.7083	281.8	0.7054	0.7090	0.7092	0.7075	0.7075
Columbus	TX	29.706	−96.547	0.7082	643.1	0.7049	0.7113	0.7113	0.7088	0.7080
Lake Charles	LA	30.188	−93.218	0.7083	305.8	0.7049	0.7113	0.7113	0.7113	0.7091
Alexandria	LA	31.260	−92.493	0.7110	23.9	0.7049	0.7113	0.7113	0.7113	0.7090
Monroe	LA	32.487	−92.081	0.7111	38.1	0.7049	0.7113	0.7113	0.7113	0.7096
Pine Bluff	AR	34.214	−91.981	0.7093	234.9	0.7049	0.7113	0.7113	0.7094	0.7083
Conway	AR	35.091	−92.437	0.7151	23.5	0.7080	0.7132	0.7150	0.7139	0.7090
Fort Smith	AR	35.385	−94.422	0.7138	34.4	0.7080	0.7194	0.7181	0.7103	0.7090
Muskogee	OK	35.741	−95.308	0.7102	143.6	0.7080	0.7194	0.7181	0.7102	0.7089
Shawnee	OK	35.350	−96.912	0.7129	105.7	0.7080	0.7192	0.7179	0.7102	0.7096
Sayre	OK	35.457	−97.618	0.7088	467.9	0.7080	0.7113	0.7113	0.7102	0.7095
Amarillo	TX	35.222	−101.831	0.7092	1063.1	0.7049	0.7113	0.7113	0.7091	0.7084
Dalhart	TX	36.061	−102.348	0.7089	788.6	0.7049	0.7113	0.7113	0.7091	0.7070
Raton	NM	36.893	−104.441	0.7096	231.8	0.7059	0.7113	0.7113	0.7082	0.7060
Alamosa	CO	37.468	−105.853	0.7099	67.7	0.7049	0.7115	0.7115	0.7080	0.7076
Salida	CO	38.536	−105.992	0.7135	259.9	0.7242	0.7113	0.7113	0.7112	0.7100
Leadville	CO	39.249	−106.292	0.7149	38.9	0.7080	0.7113	0.7113	0.7112	0.7105
Rifle	CO	39.533	−107.783	0.7110	441.5	0.7054	0.7113	0.7113	0.7106	0.7106
Craig	CO	40.513	−107.553	0.7132	38.7	0.7059	0.7113	0.7113	0.7132	0.7121
Vernal	UT	40.450	−109.502	0.7130	26.8	0.7059	0.7113	0.7113	0.7094	0.7100
Rugby	ND	48.369	−99.996	0.7089	52.3	0.7059	0.7113	0.7113	0.7113	0.7113
Crookston	MN	47.781	−96.610	0.7170	160.2	0.7320	0.7048	0.7057	0.7120	0.7133
Bemidji	MN	47.472	−94.881	0.7190[#]	37.5	0.7320	0.7614	0.7360	0.7207	0.7138
Baraboo	WI	43.470	−89.742	0.7112	50.2	0.7101	0.7146	0.7119	0.7116	0.7116
Chicago	IL	41.792	−87.594	0.7100	100.0	0.7091	0.7104	0.7100	0.7100	0.7100
Mahomet	IL	40.188	−88.400	0.7095	296.4	0.7091	0.7221	0.7223	0.7093	0.7095
Evansville	IN	38.021	−87.571	0.7109	214.2	0.7080	0.7195	0.7182	0.7155	0.7099
Paducah	KY	37.085	−88.599	0.7110	68.8	0.7059	0.7113	0.7113	0.7109	0.7096
Ozark	MO	37.024	−93.206	0.7123	11.5	0.7080	0.7089	0.7089	0.7091	0.7089
St. joseph	MO	39.755	−94.837	0.7091	252.9	0.7080	0.7192	0.7093	0.7092	0.7093
Lincoln	NE	40.736	−96.600	0.7107	256.6	0.7059	0.7122	0.7130	0.7092	0.7094
Grand Island	NE	40.930	−98.047	0.7119	256.6	0.7059	0.7077	0.7077	0.7078	0.7077
Valentine	NE	42.873	−100.549	0.7099	150.2	0.7049	0.7115	0.7114	0.7097	0.7110
Lusk	WY	42.762	−104.452	0.7116[#]	4.9	0.7049	0.7088	0.7082	0.7096	0.7082
Douglas	WY	42.760	−105.385	0.7140[#]	306.3	0.7054	0.7113	0.7113	0.7093	0.7097
Casper	WY	42.849	−106.302	0.7123[#]	415.8	0.7059	0.7139	0.7132	0.7097	0.7096
Lander	WY	42.833	−108.730	0.7257[#]	13.4	0.7073	0.7113	0.7113	0.7099	0.7100
Rock Springs	WY	41.591	−109.219	0.7112[#]	262.9	0.7059	0.7113	0.7113	0.7101	0.7090

[1]Originally reported in Chesson et al[52]

[2]Predicted from a model by Beard and Johnson[12]

[3]Predicted from a model by Bataille and Bowen[55]

[#]Average of initial measurement reported in Chesson et al[52] and additional measurements reported in Table 3.

TABLE 3 Water sample collection locations, initial and replicate analyses of the strontium isotope ratios of tap water

City	State	Collected	Measured water
Lusk	WY	2004	0.7155#
		2004	0.71081
		2004	0.71077
		2013	0.71076
		2013	0.71082
		2013	0.71080
		Average	0.7116
		1σ	0.0019
Douglas	WY	2004	0.7159#
		2004	0.71155
		2004	0.71605
		2013	0.71588
		2013	0.71420
		2013	0.71041
		Average	0.7140
		1σ	0.0025
Casper	WY	2004	0.7123#
		2013	0.71224
		2013	0.71228
		2013	0.71225
		Average	0.7123
		1σ	0.0000
Lander	WY	2004	0.7244#
		2004	0.72410
		2004	0.72466
		2013	0.72852
		2013	0.72575
		2013	0.72660
		Average	0.7257
		1σ	0.0017
Rock Springs	WY	2004	0.7114#
		2013	0.71114
		2013	0.71109
		Average	0.7112
		1σ	0.0002
Evanston	WY	2004	0.7162#
		2013	0.71279
		2013	0.71276
		Average	0.7139
		1σ	0.0020
Artesia	NM	2004	0.7143#
		2004	0.70759
		2004	0.70762
		Average	0.7098
		1σ	0.0039
Bemidji	MN	2004	0.7178#
		2004	0.71953
		2004	0.71967
		Average	0.7190
		1σ	0.0010

#Originally reported in Chesson et al.[52]

not hair collected from salons or barbershops. Specimens from salon/barbershop floors are expected to represent material from the distal end of hair strands. Thus, it is likely that the hair specimens collected in this study had been exposed to the external environment for a longer period of time, resulting in elevated values of [Sr]. Furthermore, the data presented here were from samples collected throughout the United States (Figure 1). Previous studies of [Sr] of hair have focused on more constrained geographic areas and, thus, the specimens collected in this work may display elevated [Sr] due the additional variability that becomes apparent when a larger geographic area is examined.

The mean [Sr] of tap water from municipalities studied here was 393.1 ± 474.6 μg L^{-1} (1σ, $n = 48$,) and the concentration ranged from 4.9 to 1903.1 μg L^{-1} (Table 2). We found that the [Sr] values of hair and tap water were significantly different (paired t-test, p <0.0001, t(96) = 8.111753), yet were linearly correlated ($R^2 = 0.46$, p <0.0001). The [Sr] of hair was roughly an order of magnitude lower than that of tap water from the same collection location (Figure 2). These data suggest that the amount of Sr in an individual's tap water relates to the amount in an individual's hair. Thus, in regions with elevated [Sr] of tap water, it would be expected that the [Sr] of hair would also be elevated. However, the [Sr] of hair may also relate to hair color. The cross-plot of [Sr] of hair against [Sr] of tap water, distinguishing hair coloration, showed two distinct data trends (Figure S1, supporting information). The group of data with a large number of more pigmented specimens (e.g., dark brown, red, etc.) showed a positive relationship between the two variables, while the other showed no relationship. The trend that showed no relationship between the two variables included many hair specimens that were

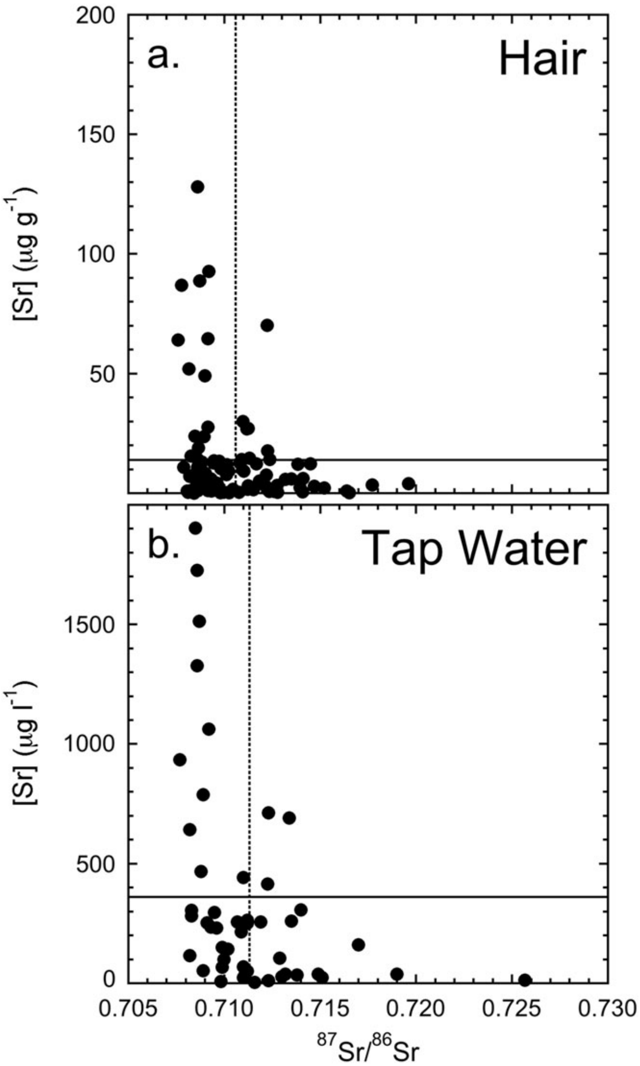

FIGURE 2 Strontium concentration [Sr] against the $^{87}Sr/^{86}Sr$ ratios of hair (A) and tap water (B). Solid and dotted lines indicate the average [Sr] and $^{87}Sr/^{86}Sr$ values, respectively

salt-and-pepper (i.e., grey or a mixture of pigmented and non-pigmented hair) in color. We found that salt-and-pepper colored hair had the lowest [Sr] of all hair colors, with an average of 4.8 μg g^{-1} (Table 1). While not statistically significant, these trends may indicate a relationship between hair color and [Sr]. The amount of melanin, the primary pigment found in human hair, has been linked to increased incorporation of elements.[58] Salt-and-pepper hair has reduced amounts of melanin or can completely lack the pigment, which could be the cause of the [Sr] in these samples being relatively reduced compared with the darker pigmented hair specimens. However, numerous hair samples with visible coloration had reduced [Sr] relative to similarly colored hair samples within the dataset. These samples displayed the same trend as the salt-and-pepper hair (Figure S1, supporting information). One possibility is that these visibly colored samples with reduced [Sr] do not represent the natural color of the hair, but rather the color of artificial dyes. At present this explanation cannot be further explored due to the anonymous nature of the collection methods.

3.2 | Sr-isotope ratios of hair

The average $^{87}Sr/^{86}Sr$ ratio of hair was 0.71059 ± 0.00234 (1σ, n = 98) and the ratio ranged from 0.70760 to 0.71962 (Table 1). This range was larger than previous reports of $^{87}Sr/^{86}Sr$ of hair specimens collected within the same city or geographic region.[47,50,51,54] Considering the large geographic area represented in this study and the known relationship between $^{87}Sr/^{86}Sr$ and geography, the observation of such a large range of $^{87}Sr/^{86}Sr$ ratios of hair might be expected (Figure 2).

The wide variation in the $^{87}Sr/^{86}Sr$ ratios of hair suggests a significant geographic control on Sr-isotopic composition. Previous research has indicated that the $^{87}Sr/^{86}Sr$ ratio of hair reflects a combination of both endogenous and exogenous sources of Sr.[54,58] However, there remains no consensus on the geospatial mechanism controlling the $^{87}Sr/^{86}Sr$ of hair. As an example, Font and colleagues[51] found endogenous Sr to be the most significant contributor to the geospatial signal encoded in the $^{87}Sr/^{86}Sr$ of hair, while several other studies conversely found strong relationships between exogenous sources and the $^{87}Sr/^{86}Sr$ of hair.[11,47,50] Therefore, separating the factors contributing to the $^{87}Sr/^{86}Sr$ of hair is paramount for utilizing this analytical tool in anthropologic studies.[5,6]

Comparing the $^{87}Sr/^{86}Sr$ ratios of leached hair and the leachate resulting from cleaning may allow for isolation of endogenous and exogenous Sr-isotopic signatures. It has been suggested that the $^{87}Sr/^{86}Sr$ of leached hair and the $^{87}Sr/^{86}Sr$ of leachate may be somewhat representative of endogenous and exogenous Sr signatures, respectively.[51,54] We found that the average $^{87}Sr/^{86}Sr$ ratio of hair leachate was 0.71055 ± 0.00230 (1σ, n = 97) and the ratio ranged from 0.70758 to 071918 (Table 1). The $^{87}Sr/^{86}Sr$ ratios of hair and leachate were not significantly different (paired t-test, p = 0.7581, t(97) = −0.30888). These results were consistent with previous studies, which found the $^{87}Sr/^{86}Sr$ ratios of hair and leachate to be generally similar.[51,54] Font and colleagues[51] analyzed the $^{87}Sr/^{86}Sr$ of hair leached with 2 M HNO_3 and the resulting leachate and found the ratios of the hair and the leachate from a single individual were not distinct and were within analytical error. The individual in question had limited geographic movements and the authors of that study suggested that the lack of distinction between the $^{87}Sr/^{86}Sr$ ratios of the hair and the leachate was indicative of similar endogenous and exogenous Sr sources.[51] In addition, Font et al[51] compared the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate from mixed hair samples collected from two barbershops and found that in one case the ratios were not different, while in the other case they were different. The authors interpreted the difference between the ratios of leached hair and leachate for one case as a consequence of variability from the mixture of multiple individuals represented in the hair sample.[51]

Here, we suggest that the difference in the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate may provide evidence of geographical movement. We found the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate were linearly correlated (R = 0.77, p <0.0001). The majority of paired measurements appeared to not be different; however, a subset of specimens had distinct $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate (Figure 3). All the specimens in our study were collected ensuring distinction of individuals, so differences between $^{87}Sr/^{86}Sr$ of leached hair and leachate were not related to sampling a mixture of individuals' hair. Previously, Tipple and colleagues[54] noted difference in the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate from 22 individuals from a single city and suggested that distinctions between endogenous and exogenous Sr may relate to travel. Here, we collected hair from fewer individuals within one location but from multiple locations, spanning a wide geographic range. Together, these two datasets suggest that a comparison of the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate may be used as a preliminary screening tool to identify individuals who had recently traveled.

As our goal in this study was to assess the capacity of the $^{87}Sr/^{86}Sr$ ratio of hair to record geographic information, it is important to identify and remove suspected "travelers" from the dataset. To identify suspected "travelers" we excluded leached hair and leachate pairs with a difference in their $^{87}Sr/^{86}Sr$ ratios greater than 0.00034 ($\Delta_{Hair\text{-}Leach}$) (Figure 3). While this cutoff was somewhat arbitrary (i.e., greater than 99% CI of SRM® 987) and warrants further investigation, we were able to remove 17 pairs that did not meet this threshold. Interestingly, specimens from the suspected "travelers" had significantly lower [Sr] than the remaining population (Student's t-test, p <0.0001, t(94.5) = −4.05071) with nearly 50% of the suspected "travelers" being from salt-and-pepper hair samples. This may suggest that pigmentation is related to the capacity of hair to bind Sr, and thus its capacity to record recent travel movements. Pigmentation and its relationship to $^{87}Sr/^{86}Sr$ and travel movements warrant additional study. Nonetheless, once suspected "travelers" were removed from consideration, we found the $^{87}Sr/^{86}Sr$ ratios of leached hair and leachate pairs were not different (paired t-test, p = 0.1258, t(80) = 1.54725) and linearly correlated (R = 0.998, p <0.0001). Additional research would be required to assess the cause(s) of the variation observed in the $^{87}Sr/^{86}Sr$ ratios between some leached hair and leachate pairs.

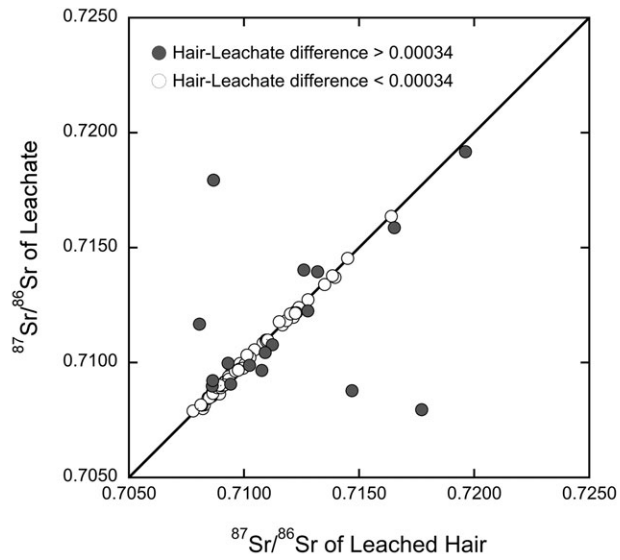

FIGURE 3 Cross-plot of the $^{87}Sr/^{86}Sr$ ratios of leachate and leached hair. Open and solid circles represent leached hair and leachate pairs with differences in $^{87}Sr/^{86}Sr$ <0.00034 and ≥0.00034, respectively. Solid line is the 1:1 line

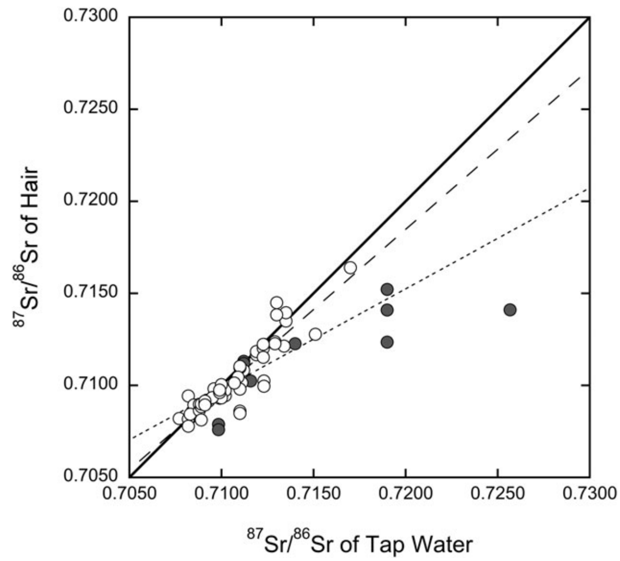

FIGURE 4 Cross-plot of the $^{87}Sr/^{86}Sr$ ratios of hair and tap water. Solid circles represent locations where the variation in the $^{87}Sr/^{86}Sr$ ratios of replicated tap water samples is >0.001. Dotted line is the linear relationship using all data and is described by the $^{87}Sr/^{86}Sr$ ratio of hair = ($^{87}Sr/^{86}Sr$ of tap water)0.55 + 0.32 ($R = 0.80$, p <0.0001). Hashed line is a linear relationship when locations with known variability in the $^{87}Sr/^{86}Sr$ ratios of tap water are removed. It is described by $^{87}Sr/^{86}Sr$ of hair = ($^{87}Sr/^{86}Sr$ of tap water)0.87 + 0.09 ($R = 0.83$, p <0.0001). Solid line is the 1:1 line

3.3 | Relationships between the Sr-isotope ratios of hair and tap water

The mean $^{87}Sr/^{86}Sr$ ratio of tap water from municipalities studied here was 0.71132 ± 0.00322 (1σ, $n = 48$) and the ratios ranged from 0.70770 to 0.72567 (Chesson et al[52] and this study; Tables 2 and 3). We found that the $^{87}Sr/^{86}Sr$ ratios of hair and tap water were significantly different (paired t-test; $p = 0.0005$, t(97) = 3.630528) and linearly correlated ($R^2 = 0.80$; p <0.0001). Similar results were found when the hair-leachate pairs for "travelers" were removed from consideration (paired t-test; $p = 0.0009$, t(80) = −3.46618). However, when considering the screened dataset, the linear relationships were relatively stronger ($R = 0.83$; p <0.0001).

Numerous studies have observed relationships between the $^{87}Sr/^{86}Sr$ ratio of hair and possible exogenous sources of Sr in the environment (e.g.,[5,11,47,50,51,54,63]). Recently, a significant relationship between the $^{87}Sr/^{86}Sr$ ratios of hair and tap water was observed within a single city that had municipal water with several distinct ratios.[47] Tipple and colleagues[47] found a significant link between the $^{87}Sr/^{86}Sr$ ratios of hair from individual donors and the ratios of water in the regions of the city where the donors resided. Given that significant amounts of Sr can be incorporated into hair keratin after eruption of strands from the scalp,[58] the authors suggested that bathing with tap water influences the $^{87}Sr/^{86}Sr$ ratio of hair.[47]

It has been previously shown that Sr displays negligible isotopic variation between Sr source and organic tissue(s), with the $^{87}Sr/^{86}Sr$ ratio of organic tissues directly reflecting that of the source(s) (e.g.,[12,23,35,64]). Here, we found that the $^{87}Sr/^{86}Sr$ ratios of hair and tap water were strongly related (Figure 4), but these ratios were significantly different, potentially pointing towards additional factors that affect the $^{87}Sr/^{86}Sr$ ratio of hair. However, we note that measurements of the $^{87}Sr/^{86}Sr$ ratio of tap waters showed significant spatial and temporal variation in some locations (Table 3). Previous studies have shown significant variation in the $^{87}Sr/^{86}Sr$ and other isotope ratios of tap water on the local scale due to water management practices.[47,52,65-69] While the hair samples presented in Figure 4 did not suggest travel movement based on the isotopic comparisons of the hair-leachate pairs (i.e., difference in $^{87}Sr/^{86}Sr$ of pairs <0.00034), it is possible that spatial and temporal variation in the $^{87}Sr/^{86}Sr$ ratio of tap water added to the uncertainty in drawing geographic correlations between individuals and their environments. Drawing a comparison between the $^{87}Sr/^{86}Sr$ ratio of tap water and the ratio of either hair or leachate may be impossible in some cases due to underlying and insufficiently sampled variability in the $^{87}Sr/^{86}Sr$ ratio of tap water. With isotopically variable (1σ >0.0010) water samples removed, the relationship between the $^{87}Sr/^{86}Sr$ ratios of tap water and leached hair has a slope of 0.87 ($R = 0.93$, p <0.0001) – much closer to the theoretical slope of 1. Given the known isotopic variability in water supply within some metropolitan areas, additional sampling resolution may be required in these locations to understand the geospatial relationships between the $^{87}Sr/^{86}Sr$ ratio of tap water and that of human hair.

3.4 | Comparison between $^{87}Sr/^{86}Sr$ ratios of hair and modeled bedrock/water

As expected, the measured $^{87}Sr/^{86}Sr$ ratio of hair was significantly positively correlated with the modeled ratios of bedrock and surface waters (Table 3). We found that the Beard and Johnson[12] and Bataille and Bowen[70] models of $^{87}Sr/^{86}Sr$ of bedrock explained approximately 36% and 11% of the variation observed in the $^{87}Sr/^{86}Sr$ ratios of hair, respectively (Figure S2, supporting information). When compared with the modeled $^{87}Sr/^{86}Sr$ data of surface water, we found that the

Bataille and Bowen[70] estimates of the $^{87}Sr/^{86}Sr$ ratios of local water, flux-weighted small catchment water, and flux-weighted averaged water from HUC 8 hydrologic units (i.e., watershed water) explained 10%, 19% and 32% of the variation observed in the $^{87}Sr/^{86}Sr$ ratios of leached hair, respectively (Figure S3, supporting information). While the Beard and Johnson[12] model of the $^{87}Sr/^{86}Sr$ ratios of bedrock and the Bataille and Bowen[70] model of the $^{87}Sr/^{86}Sr$ ratios of watershed water explained similar levels of isotopic variation in the hair, the slope of the relationship between the ratios of (estimated) watershed water and hair was closer to 1 ($m = 0.74$) than the slope of the relationship between the ratios of (estimated) bedrock and hair ($m = 0.18$).

Various water management practices complicate predictions of the $^{87}Sr/^{86}Sr$ ratios of human tissues based on the ratios of local water. Modeled estimates of bioavailable $^{87}Sr/^{86}Sr$ ratios largely rely on geologic bedrock mapping and estimates of rock weathering based on rock type.[55,70,71] While these models are appropriate for natural systems, they may not be entirely appropriate for developed regions and human-modified systems. Specifically, human communities may draw on surface waters, groundwater, or transported water from other regions depending on needs and the local hydrogeographic situation. Communities that utilize groundwater and/or transported water exclusively or in a significant proportion versus surface water have been found to have the largest differences between measured and expected $^{87}Sr/^{86}Sr$ ratios of water[52] and are particularly difficult to geospatially model. Given that the rock type and mineralogy of a groundwater aquifer may be dissimilar to that of the surrounding surficial geologic units, it is not unexpected that the modeled $^{87}Sr/^{86}Sr$ ratios of water and the measured ratios of human tissues could be dissimilar. As an example, Tipple and colleagues[47] demonstrated that a single water system may supply groundwater to some service areas and not others, and may change the relative proportion of groundwater supplied depending on season or other climatic factors. In their study, the tap water sourced from groundwater had a higher $^{87}Sr/^{86}Sr$ ratio than the surface water.[47] Furthermore, water supply systems may utilize water that has been transported large distances. For instance, water systems for coastal California cities often transport snowmelt-derived water 200 miles or more from the Sierra Nevada Mountains to distant metropolitan regions. This water management strategy causes the measured and modeled $^{87}Sr/^{86}Sr$ ratios of water to be dissimilar.[52] Despite these complexities, geospatial modeling of bioavailable $^{87}Sr/^{86}Sr$ ratios is required to use the Sr-isotope system as a quantitative tool in provenance studies of modern humans. Additional modeling efforts will be necessary to account for the complex mixing of Sr isotopes between natural and anthropogenic systems. Our findings suggest that next-generation prediction models for the Sr-isotopic composition of water need to incorporate additional layers for applications towards modern humans, including the underlying infrastructure of water systems in municipalities and its effects on the $^{87}Sr/^{86}Sr$ ratios of tap water available to residents. While integrating these natural and anthropogenic factors into a mechanistic model might be challenging in the near future, calibrating multivariate models using machine-learning regression methods can significantly improve the predictive power.[72]

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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