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Hydrogen and Oxygen Stable Isotope Ratios of Milk in the United States

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Models of hydrogen and oxygen incorporation in human tissues recognize the impact of geographic location on the isotopic composition of fluid intake, but inputs can include nonlocal beverages, such as milk. Milk and cow drinking water were collected from dairies, and commercially available milk was purchased from supermarkets and fast food restaurants. It was hypothesized that milk water δ^2H and $\delta^{18}O$ values record geographic location information. Correlations between milk water isotope ratios and purchase location tap water were significant. However, the amount of variation in milk δ^2H and $\delta^{18}O$ values explained by tap water was low, suggesting a single estimation of fluid input isotope ratios may not always be adequate in studies. The δ^2H and $\delta^{18}O$ values of paired milk and cow drinking water were related, suggesting potential for geographical origin assignment using stable isotope analysis. As an application example, milk water $\delta^{18}O$ values were used to predict possible regions of origin for restaurant samples.

KEYWORDS: Stable isotope; hydrogen; oxygen; dairy; milk; region of origin

INTRODUCTION

Recent process-based models for the incorporation of hydrogen and oxygen atoms into animal hair keratin have been developed and tested for humans (*1, 2*) and a small mammal (*3*). These models follow the incorporation of stable isotopes from an animal's dietary and drinking water inputs to its body water pool to predict the isotopic composition of proteinaceous tissues. The drinking water consumed by an animal is derived from an isotope landscape, or isoscape (*4*), which describes predictable geographic variations in the stable isotope values of hydrogen (δ^2H) and oxygen ($\delta^{18}O$) within meteoric water (*5*) and tap water (*6*). Because animals drink local waters, the water in their tissues reflects the local δ^2H and $\delta^{18}O$ values (*4*). As such, the stable isotope analysis of proteinaceous tissues such as keratin is useful for investigations of the geographic region of origin of animals and humans, with applications in the fields of animal ecology (*7*), anthropology (*8*), archeology (*9*), forensics (*10*), and wildlife crime (*11*).

Current predictive hair keratin models recognize four major O inputs to the body water pool of a human consumer: (1) drinking water, (2) food, (3) food water, and (4) molecular O_2 (*1, 2, 12*). The first three sources provide the basis for H atoms in the body water pool as well. The isotopic composition of O_2 does not vary worldwide (*13*) and is constant in model calculations. As a first approximation, modelers assume that modern humans consume a global or continental supermarket diet (*1*), and these models use constant δ^2H and $\delta^{18}O$ values for food and food water inputs

regardless of consumer location (however, see refs *2* and *8* for discussions of local food inputs). The input responsible for most isotopic variation in final hair keratin values is drinking water. Thus, the geographic location of a consumer is important when process-based models are applied to predict final tissue δ^2H and $\delta^{18}O$ values.

Whereas tap water can be considered one of the main components of fluid intake, the average modern human consumes a large variety of beverages. For example, Americans regularly imbibe alcoholic beverages, coffee, tea, fruit juices and drinks, milk, soft drinks, and bottled water (*14*). Although some beverages (e.g., coffee and fruit drinks) are brewed or reconstituted with local tap water, the source of water in other beverages (e.g., milk) could be far distant from the consumer. Incorporating a single model parameter to account for the impact of consumer location on the isotopic composition of liquid inputs may not be appropriate for cases in which large quantities of nonlocal beverages are imbibed.

In this survey, we collected paired milk and cow drinking water samples from dairies to assess the relationship between water inputs and milk outputs for dairy cows. We also purchased milk from grocery retailers and multiple outlets of a national fast food restaurant across the contiguous United States and compared the stable isotope ratios of water extracted from these milk samples to local tap water. We hypothesized that the δ^2H and $\delta^{18}O$ values of milk water are related to cow drinking water and record geographic information regarding the location of cows. We also hypothesized that there is isotopic variation in commercially available milk and that the δ^2H and $\delta^{18}O$ values of milk produced locally to a consumer should be related to those of purchase

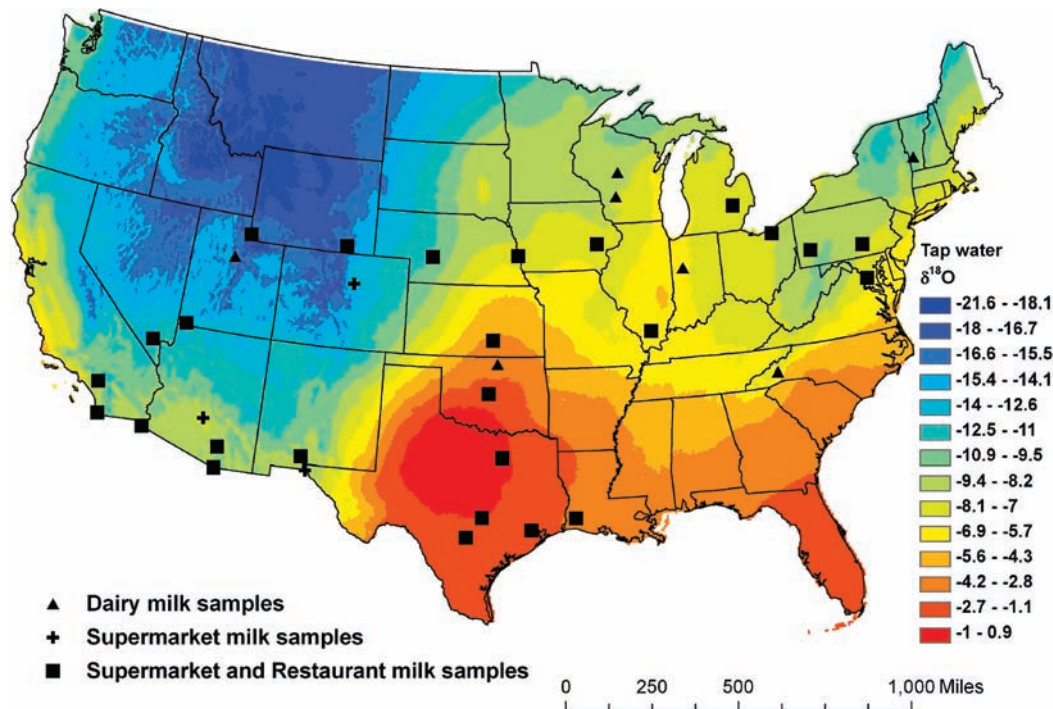*Corresponding author [telephone (801) 755-7990; fax (801) 277-1204; e-mail lesley@isoforensics.com].

Figure 1. Locations for the paired dairy milk and cow drinking water samples (triangles) and purchased milk samples collected within the contiguous United States shown on a predicted tap water oxygen isoscape (see ref 6). Cities where both supermarket and restaurants samples were purchased are denoted by squares. Only supermarket samples were purchased in cities denoted by crosses.

location tap water. We first describe the general patterns observed in the measured δ^2H and δ^{18}O values of water extracted from milk before presenting a potential application of the stable isotope analysis of milk.

MATERIALS AND METHODS

Sample Acquisition and Preparation. Paired milk and cow drinking water samples were collected by colleagues and acquaintances that had previously established connections with dairies. Volunteers were provided with 2 oz plastic bottles, which were filled with milk and tap water and then frozen. Frozen samples were shipped with ice packs to the University of Utah via overnight delivery. Samples were thawed but cold at delivery. Samples were immediately refrozen upon receipt. Commercially available milk was purchased from traditional grocery retailers ("supermarkets") in cities located throughout the contiguous United States. In each supermarket, we purchased one container of whole milk from each brand that offered milk in 1 qt or smaller containers. In stores that did not stock quart-sized milk containers, we purchased a half-gallon container of the store brand instead. In a subset of the surveyed cities we also collected the milk drinks provided with children's fast food meals, which were purchased from multiple outlets of the same restaurant chain. Upon purchase, all milk samples were stored on wet ice until returned to the laboratory and frozen.

Frozen purchased milk, as well as paired milk and water samples, were later thawed. A 1 mL subsample of each milk sample was distributed onto clean glass wool (15), and water was cryogenically extracted using a vacuum distillation line (16). A 0.5 mL aliquot of extracted water was transferred to a 1.8 mL crimp-top GC vial and sealed; a 0.5 mL aliquot of each cow drinking water sample was transferred to a GC vial without distillation. Sealed samples were stored in a cool, dark location prior to analysis. We also collected and analyzed tap water from every purchase city, as detailed previously (17).

Stable Isotope Analysis. Samples were analyzed in duplicate at the University of Utah Stable Isotope Ratio Facility for Environmental Research (SIRFER; http://sirfer.net) on a ThermoFinnigan-MAT Delta Plus XL isotope ratio mass spectrometer (Bremen, Germany) with a high-temperature conversion elemental analyzer (TC/EA) attached. Samples were pyrolyzed at 1400 °C to produce H_2 and CO gas. Resultant gases were

separated on a 1 m, 0.25 in. (outer diameter), molecular sieve 5 Å gas chromatography column (Costech Analytical, Valencia, CA). Water samples were introduced to the pyrolysis column using a PAL autosampler (LEAP Technologies, Carrboro, NC) and analyzed alongside a set of three laboratory water reference materials previously calibrated to the Vienna Standard Mean Ocean Water (VSMOW) scale. The analytical precisions for samples were ±1.55‰ and ±0.17‰ for H and O, respectively.

Stable Isotope Notation. Stable isotope abundances are reported in δ notation as parts per thousands (‰), where

$$\delta = (R_A/R_S - 1) \times 1000$$

and R_A and R_S are the molar ratios of the rare to abundant isotope (e.g., ^{2}H/^{1}H) in the sample and standard, respectively. The international standard for both hydrogen and oxygen stable isotope analysis is VSMOW.

RESULTS

We collected paired milk and cow drinking water samples from eight locations in six states and the territory of Puerto Rico (**Figure 1**). We purchased milk samples from supermarkets in 30 cities within 18 states (**Figure 1**). Two or more brands of milk were purchased in 10 cities, for a total of 45 samples. We also collected milk from outlets of the same national fast food restaurant in 26 of the 30 cities, which were located in 17 states (**Figure 1**). Statistics for the purchased supermarket and restaurant milk sample data sets are summarized in **Table 1**.

There was a significant linear relationship between the measured δ^2H and δ^{18}O values of water extracted from the milk samples collected at dairies, described by the equation δ^2H = $9.2\delta^{18}$O + 11‰ (data not shown; $r^2 = 0.986$, $P < 0.0001$). The stable isotope ratios of paired milk and cow drinking water samples were related; the simple linear regression equation describing the covariation in milk and cow drinking water δ^2H values was δ^2H$_{milk}$ = $0.96\delta^2$H$_{water}$ + 9‰ (**Figure 2a**). The equation for δ^{18}O values was δ^{18}O$_{milk}$ = $0.86\delta^{18}$O$_{water}$ + 1.1‰ (**Figure 2b**). The slope of the δ^2H$_{milk}$ versus δ^2H$_{water}$ line was not

Table 1. Statistics for the Measured Hydrogen and Oxygen Stable Isotope Ratios of Water Extracted from Milk Purchased in U.S. Supermarkets and Fast Food Restaurants

	supermarket milk		restaurant milk	
	δ^2H (‰)	$\delta^{18}O$ (‰)	δ^2H (‰)	$\delta^{18}O$ (‰)
maximum	−16	−3.6	−27	−4.0
minimum	−108	−13.0	−107	−13.2
range	92	9.4	80	9.3
mean	−48	−6.5	−58	−7.8
σ	21	2.1	28	3.1

significantly different from that of a 1:1 line, but the slope of the $\delta^{18}O_{milk}$ versus $\delta^{18}O_{water}$ line was ($F_{1,12} = 19.8$, $P < 0.001$). The mean difference between milk and cow drinking water was 11‰ for δ^2H values (1σ = 3‰) and 2.2‰ for $\delta^{18}O$ values (1σ = 0.7‰). The δ^2H and $\delta^{18}O$ values of water extracted from dairy milk samples were isotopically enriched relative to cow drinking water.

There was a significant linear relationship between the measured δ^2H and $\delta^{18}O$ values of water extracted from supermarket milk samples, described by the equation $\delta^2H = 9.5\delta^{18}O + 14‰$ (**Figure 3a**; $r^2 = 0.953$, $P < 0.0001$). There was also a significant linear relationship between the measured δ^2H and $\delta^{18}O$ values of water extracted from restaurant milk samples, described by the equation $\delta^2H = 9.0\delta^{18}O + 12‰$ (**Figure 3b**; $r^2 = 0.981$, $P < 0.0001$). The supermarket and restaurant regression lines were not statistically different from one another, nor were they different from the regression line for dairy milk samples. The slopes of the supermarket and restaurant lines were different than the slope of the regression line generated from the δ^2H and $\delta^{18}O$ values of purchase location tap water samples ($F_{1,72} = 15.4$, $P < 0.001$; and $F_{1,53} = 8.5$, $P < 0.01$, respectively). The tap water line was described by the equation $\delta^2H = 8.1\delta^{18}O + 6‰$ (data not shown; $r^2 = 0.984$, $P < 0.0001$).

As seen in **Figure 3b**, restaurant samples clustered in three distinct groups, described in **Table 2**. The presence of three distinct groups in the supermarket milk data set (**Figure 3a**) was not obvious through visual examination of the data set. The correlations between the stable isotope ratios of water extracted from milk versus tap water collected at purchase locations were significant ($P < 0.01$ in all cases). Tap water described a moderate amount of variation in milk water δ^2H and $\delta^{18}O$ values in both the supermarket and restaurant data sets; coefficients of determination (r^2) ranged from 0.30 to 0.49 (data not shown). As expected, on the basis of the clusters of data evident in the restaurant data set (**Figure 3b**), the r^2 values were lower for the restaurant milk versus tap water regressions than for the supermarket milk versus tap water regressions.

DISCUSSION

Milk Records the Isotopic Composition of Cow Drinking Water. We observed that the δ^2H and $\delta^{18}O$ values of paired milk and cow drinking water samples are strongly related in a predictable manner, as seen in **Figure 2**. As we expected, the stable isotope ratios of water within cow milk record geographical origin information (i.e., the stable isotope ratios of local drinking water). However, the δ^2H and $\delta^{18}O$ values of paired milk and cow drinking water did not exactly match due to the contribution of food and atmospheric O_2 to body water; for both H and O, the water within cow milk was enriched relative to drinking water.

As discussed above, the δ^2H and $\delta^{18}O$ values of body water in any animal are a mixture of hydrogen and oxygen atoms derived from drinking water, food, food water, and molecular O_2 (*12*).
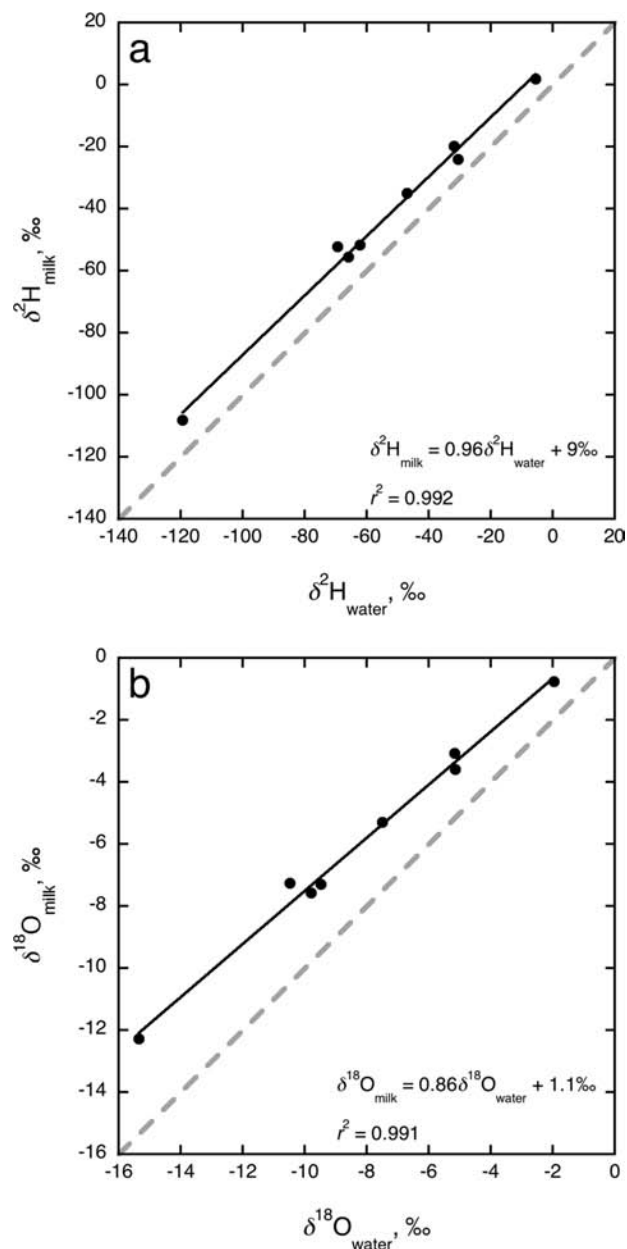

Figure 2. δ^2H (**a**) and $\delta^{18}O$ (**b**) values of paired dairy milk and cow drinking water samples. The simple linear regressions are shown by the solid black lines. The thick dashed gray lines represent the 1:1 lines for a theoretical perfect match between the isotopic composition of paired milk and cow drinking water samples.

Some H and O atoms are passively diffused into the body water pool [i.e., from drinking water (*18*)]; other atoms are added only through the production of water during metabolism [i.e., H from food; O from food and molecular O_2 (*3, 19*)]. The δ^2H and $\delta^{18}O$ values of plant cellulose, like that consumed by cows, are typically enriched (*20*); atmospheric O_2 has a $\delta^{18}O$ value of ca. +23.5‰ (*13*). Thus, the isotopic composition of metabolic water produced by a cow will generally be isotopically enriched relative to its drinking water. Although drinking water represents the largest contribution to a cow's body water pool, the impact of the enriched metabolic water could change total body water δ^2H and $\delta^{18}O$ values so that the values no longer exactly match the isotopic composition of the cow's drinking water (*19,21,22*). The magnitude of the enrichment between milk water and drinking water should be a function of total water flux in the cow: the higher the water flux, the more the milk water and drinking water
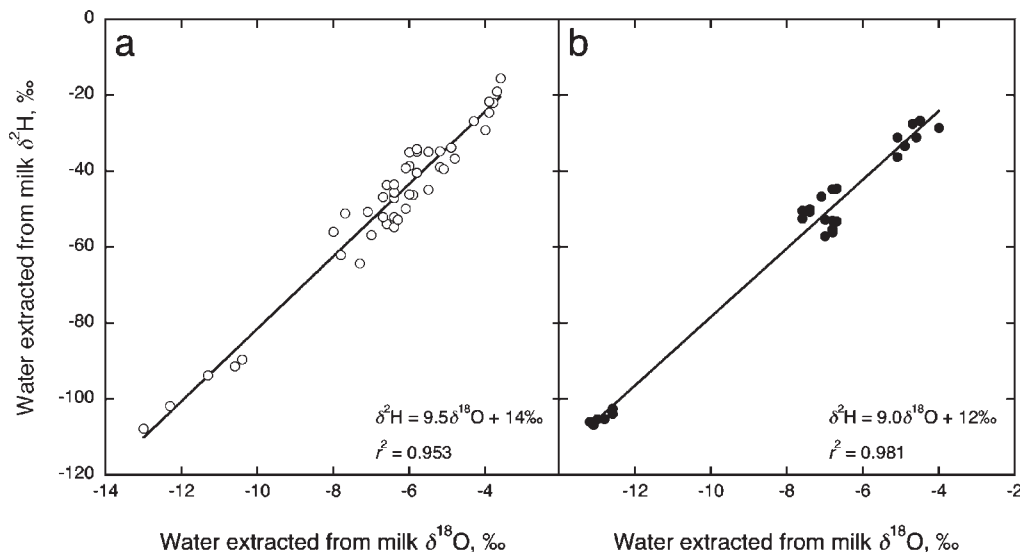
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Figure 3. δ^2H and $\delta^{18}O$ values of water extracted from milk samples collected from supermarkets (open circles, **a**) and multiple outlets of a national fast food restaurant (solid circles, **b**). The simple linear regressions are shown by the solid black lines.

Table 2. Calculated Means and Standard Deviations for Three Groups in the Restaurant Milk Data Set, Identified by Visual Examination (see **Figure** 3b)

group	δ^2H (‰) mean	σ	$\delta^{18}O$ (‰) mean	σ	n
1	−31	3	−4.7	0.4	7
2	−51	4	−7.0	0.3	13
3	−105	0	−12.9	0.3	6

should be similar because the contribution of drinking water relative to metabolic water in the body water pool increases.

Other studies have documented a 2−6‰ enrichment between cow drinking water and milk water $\delta^{18}O$ values (*21−23*), which is corroborated by our paired O data (mean enrichment = 2.2‰). It has been previously suggested the predictable relationship between the stable isotope ratios of milk and cow drinking water documented in this survey and elsewhere could be used to investigate the region of origin of milk samples from unknown locations (*21*). We demonstrate a possible geolocation application, using the stable isotope analysis of milk samples from restaurants, below.

Geography Affects Milk Water δ^2H and $\delta^{18}O$ Values at Regional Scales. We present two possible explanations for the modest explanatory power of the correlations between milk and tap water δ^2H and $\delta^{18}O$ values (r^2 = 0.30−0.49; data not shown). First, the commercial producers that provide milk to American consumers in supermarkets and restaurants use regional distribution systems to transport milk across a large area. Therefore, because of the distance that milk is transported across a region, the δ^2H and $\delta^{18}O$ values of milk water and the tap water at purchase location need not necessarily be linked. Alternatively, large commercial milk operations can combine milk produced in many locations prior to distribution. This mixing of milk from throughout a large area should weaken the link between milk water δ^2H and $\delta^{18}O$ values and a specific consumer's local tap water.

Traditionally, the dairy industry was concentrated in three main regions of the United States: the northeast, the Great Lake states, and the Corn Belt (*24*). The past 30 years has seen rapid expansion into western states, as evidenced by increasing milk production in Idaho, New Mexico, and Washington (*25*). Although large numbers of fluid milk processing plants can still be found in the traditional dairy regions, plants are now located coast to coast (*26*). The dairy industry has simultaneously undergone a period of rapid consolidation, whereby many smaller dairies have been replaced by larger, multiple-herd operations (*24, 25, 27*). The combination of geographic expansion with commercial consolidation has created regions where a single milk processor can distribute large quantities of milk across the region while simultaneously acting as mixers for milk produced throughout a region.

The evidence for a regional milk distribution pattern is especially strong in the restaurant milk data set. While the correlations between milk water and tap water stable isotope ratios were significant, the amount of variance in restaurant milk water δ^2H and $\delta^{18}O$ values explained by tap water at purchase location was low (r^2 = 0.32 and 0.30, respectively; data not shown). Water extracted from the restaurant milk samples fell into three distinct groups (**Figure 3b** and **Table 2**), and each group contained samples purchased across a large range of tap water isotope values. For example, group 3 had the lowest mean δ^2H value (−105‰) and included milk purchased in Arizona, Utah, and Wyoming. The tap water δ^2H values for these purchase locations spanned a ~65‰ range. However, the purchase location tap water δ^2H values for group 3 were, on average, the lowest values measured from the tap water data set. In general, the isotopic composition of milk purchased in states with low tap water stable isotope ratios was likely to be low as well, suggesting a regional pattern in the distribution of milk.

Example Geolocation Application Using Restaurant Milk. As an example application of stable isotope analysis for beverage origin assignment, we used the restaurant data set to predict dairy cow location based on milk water $\delta^{18}O$ values. Using the equation describing covariation in the $\delta^{18}O$ values of paired dairy milk and cow drinking water samples (**Figure 2b**), we predicted the $\delta^{18}O$ value of drinking water for cows that could have produced the milk samples in each of the three restaurant milk sample groups (based on group mean; **Table 2**). The calculated drinking water $\delta^{18}O$ value ± 3σ about the respective group mean (**Table 2**) was then used to predict the potential geographic regions of origin of the dairy cows. Predicted regions for each group, based on a tap water oxygen isoscape of the United States (*6*), are shown in **Figure 4**. Sample purchase locations have been color-coded to match predicted regions to highlight the general correspondence
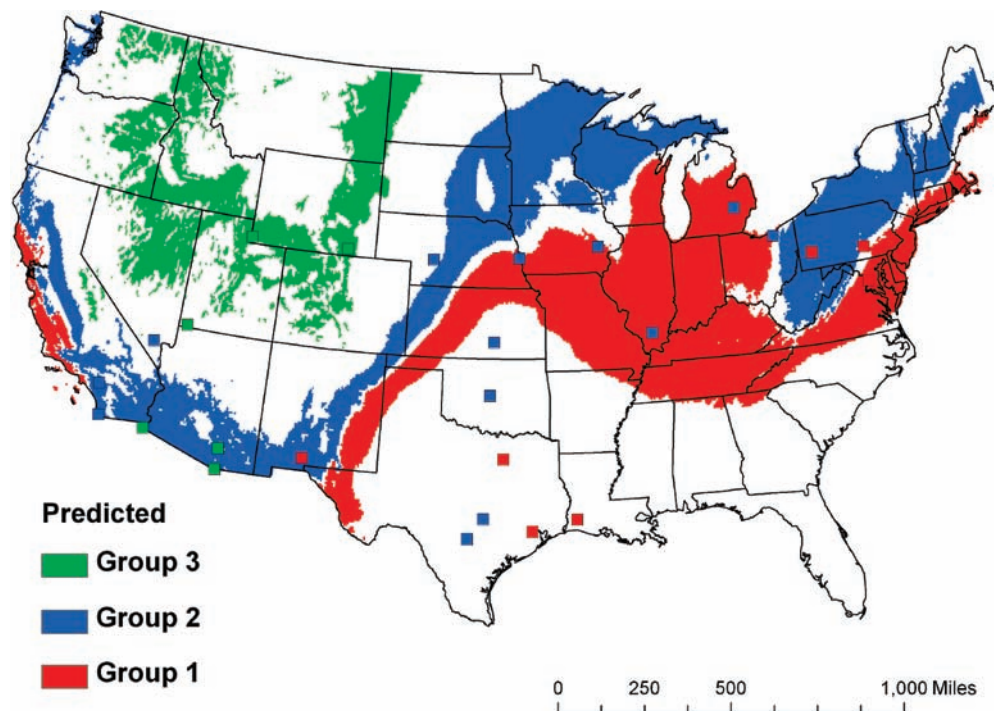
Figure 4. Predicted regions of origin for cows that could have produced the milk purchased from multiple outlets of a national fast food restaurant. Groups were identified from **Figure 3b** and are described in **Table 2**. The $\delta^{18}O$ value of cow drinking water was predicted using group mean $\delta^{18}O$ values and the equation $\delta^{18}O_{milk} = 0.86\delta^{18}O_{water} + 1.1‰$ (**Figure 2b**); error was estimated as 3σ for each group (**Table 2**). The prediction regions were constructed using a tap water oxygen isoscape (*6*). Purchase locations, denoted by squares, have been color-coded to match prediction regions.

between purchase and potential source locations (**Figure 4**). Although we currently have no information on the processing, distribution, and/or supply system(s) used by the national fast food restaurant chain to challenge or verify our predictions, this exercise nevertheless highlights the potential usefulness of stable isotope analysis for tracing the production and transport of milk.

Stable Isotope Analysis of Milk May Be Useful for Predicting Regions of Origin. We found that milk water records the isotopic composition of the cow's environmental drinking water. Therefore, it may be possible to predict the region of origin for a milk sample, an application of stable isotope analysis we have explored using the restaurant milk data set. We predicted that milk collected from restaurants during this survey was produced in all five of the top five milk-producing states in the United States (*24*). However, predicted regions were also consistent with states without large amounts of milk production, such as Nevada and West Virginia (*28*). On the basis of the stable isotope analysis of extracted milk water alone, we cannot exclude these states from predictions. The use of other chemical analyses (*23*) or the stable isotope analysis of milk solids (*22*) may help to refine these predictions, with obvious implications for future work to expand the use of stable isotope analysis for the geographical origin assignment of milk.

The δ^2H and $\delta^{18}O$ values of water extracted from dairy milk samples were not equal to those of the drinking water of the cow. Furthermore, the stable isotope ratios of commercially available milk that could have been transported long distances were not necessarily related to those of local tap water at purchase location. The transportation of milk some distance from production region to purchase location means the stable isotopic composition of milk is not always reliably linked to a consumer's geographic location. Thus, the use of a single parameter, based on consumer location, to model the stable isotope ratios of a consumer's fluid intake to predict the isotopic composition of proteinaceous tissues may not always be adequate in animal studies. However,

the application of milk water isotopes to source the region of origin of milk is promising, as we have demonstrated in an example exercise using restaurant milk samples.

LITERATURE CITED

(1) Ehleringer, J. R.; Bowen, G. J.; Chesson, L. A.; West, A. G.; Podlesak, D. W.; Cerling, T. E. Hydrogen and oxygen isotope ratios in human hair are related to geography. *Proc. Natl. Acad. Sci. U.S.A.* **2008**, *105*, 2788–2793.

(2) Bowen, G. J.; Ehleringer, J. R.; Chesson, L. A.; Thompson, A. H.; Podlesak, D. W.; Cerling, T. E. Dietary and physiological controls on the hydrogen and oxygen isotope ratios of hair from mid-20th century indigenous populations. *Am. J. Phys. Anthropol.* **2009**, *139*, 494–504.

(3) Podlesak, D. W.; Torregrossa, A.-M.; Ehleringer, J. R.; Dearing, M. D.; Passey, B. H.; Cerling, T. E. Turnover of oxygen and hydrogen isotopes in the body water, CO_2, hair, and enamel of a small mammal. *Geochim. Cosmochim. Acta* **2008**, *72*, 19–35.

(4) West, J. B.; Bowen, G. J.; Cerling, T. E.; Ehleringer, J. R. Stable isotopes as one of nature's ecological recorders. *Trends Ecol. Evol.* **2006**, *21*, 408–414.

(5) Bowen, G. J.; Revenaugh, J. Interpolating the isotopic composition of modern meteoric precipitation. *Water Resour. Res.* **2003**, *39*, 1299–1311.

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(6) Bowen, G. J.; Ehleringer, J. R.; Chesson, L. A.; Stange, E.; Cerling, T. E. Stable isotope ratios of tap water in the contiguous United States. *Water Resour. Res.* **2007**, *43*, W03419.

(7) Hobson, K. A.; Van Wilgenburg, S. L.; Larson, K.; Wassenaar, L. I. A feather hydrogen isoscape for Mexico. *J. Geochem. Explor.* **2009**, doi: 10.1016/j.gexplo.2009.02.002.

(8) Thompson, A. H.; Chesson, L. A.; Podlesak, D. W.; Bowen, G. J.; Cerling, T. E.; Ehleringer, J. R. Stable isotope analysis of modern human hair collected from Asia (China, India, Mongolia and Pakistan). *Am. J. Phys. Anthropol.* **2010**, doi: 10.1002/ajpa.21162.

(9) Wilson, A. S.; Taylor, T.; Ceruti, M. C.; Chavez, J. A.; Reinhard, J.; Grimes, V.; Meier-Augenstein, W.; Cartmell, L.; Stern, B.; Richards, M. P.; Worobey, M.; Barnes, I.; Gilbert, M. T. P. Stable isotope and DNA evidence for ritual sequences in Inca child sacrifice. *Proc. Natl. Acad. Sci. U.S.A.* **2007**, *104*, 16456–16461.

(10) Meier-Augenstein, W.; Fraser, I. Forensic isotope analysis leads to identification of a mutilated murder victim. *Sci. Justice* **2008**, *48*, 153–159.

(11) Tobe, S. S. Determining the geographic origin of animal samples. In *Forensic Science in Wildlife Investigations*; Linacre, A., Ed.; CRC Press: Boca Raton, FL, 2009; pp 127−156.

(12) Ritz, P.; Cole, T. J.; Davies, P. S. W.; Goldeberg, G. R.; Coward, W. A. Interactions between ^{2}H and ^{18}O natural abundance variations and DLW measurements of energy expenditure. *Am. J. Physiol.* **1996**, *271*, E302–E308.

(13) Kroopnick, P.; Craig, H. Atmospheric oxygen: isotopic composition and solubility fractionation. *Science* **1972**, *175*, 54–55.

(14) USDA. Food availability (per capita) data system, http://www.ers.usda.gov/Data/FoodConsumption/.

(15) West, J. B.; Ehleringer, J. R.; Cerling, T. E. Geography and vintage predicted by a novel GIS model of wine δ^{18}O. *J. Agric. Food Chem.* **2007**, *55*, 7075–7083.

(16) West, A. G.; Patrickson, S. J.; Ehleringer, J. R. Water extraction times for plant and soil materials used in stable isotope analysis. *Rapid Commun. Mass Spectrom.* **2006**, *20*, 1317–1321.

(17) Chesson, L. A.; Podlesak, D. W.; Erkkila, B. R.; Cerling, T. E.; Ehleringer, J. R. Isotopic consequences of consumer food choice: hydrogen and oxygen stable isotope ratios in foods from fast food restaurants versus supermarkets. *Food Chem.* **2010**, *119*, 1250–1256.

(18) Murphy, M. R. Symposium: Nutritional factors affecting animal water and waste quality. *J. Dairy Sci.* **1992**, *75*, 326–333.

(19) Kohn, M. J. Predicting animal δ^{18}O: accounting for diet and physiological adaptation. *Geochim. Cosmochim. Acta* **1996**, *60*, 4811–4829.

(20) Roden, J.; Lin, G.; Ehleringer, J. A mechanistic model for interpretation of hydrogen and oxygen isotope ratios in tree-ring cellulose. *Geochim. Cosmochim. Acta* **2000**, *64*, 21–35.

(21) Kornexl, B. E.; Werner, T.; Rossmann, A.; Schmidt, H.-L. Measurement of the stable isotope abundances in milk and milk ingredients - a possible tool for origin assignment and quality control. *Z. Lebensm.-Unters. -Forsch.* **1997**, *205*, 19–24.

(22) Camin, F.; Perini, M.; Colombari, G.; Bontempo, L.; Versini, G. Influence of dietary composition on the carbon, nitrogen, oxygen and hydrogen stable isotope ratios of milk. *Rapid Commun. Mass Spectrom.* **2008**, *22*, 1690–1696.

(23) Renou, J.-P.; Deponge, C.; Gachon, P.; Bonnefoy, J.-C.; Coulon, J.-B.; Garel, J.-P.; Vérité, R.; Ritz, P. Characterization of animal products according to geographic origin and feeding diet using nuclear magnetic resonance and isotope ratio mass spectrometry: cow milk. *Food Chem.* **2004**, *85*, 63–66.

(24) Blayney, D. P. The changing landscape of U.S. milk production, www.ers.usda.gov/publications/sb978/sb978.pdf.

(25) Rutt, M. E. *On the Moo-ve: Testing for Spatial Agglomeration Economies in the U.S. Diary Industry*; Kansas State University: Manhattan, KS, 2007.

(26) Glory, A. Current status of dairy food markets, www.cpdmp.cornell.edu/CPDMP/Pages/Workshops/Syracuse06/.

(27) Miller, J. J.; Blayney, D. P. Diary backgrounder, www.ers.usda.gov/Publications/LDP/2006/07Jul/LDPM14501/.

(28) USDA. National Agricultural Statistics Service Quick Stats, http://www.nass.usda.gov/Data_and_Statistics/Quick_Stats/index.asp.