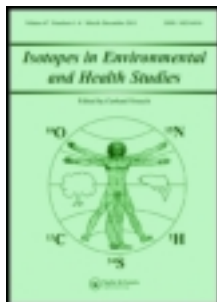


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## $\delta^2\text{H}$ and $\delta^{18}\text{O}$ of human body water: a GIS model to distinguish residents from non-residents in the contiguous USA

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An understanding of the factors influencing the isotopic composition of body water is important to determine the isotopic composition of tissues that are used to reconstruct movement patterns of humans. The  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of body water ( $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$ ) are related to the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of drinking water ( $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$ ), but clearly distinct because of other factors including the composition of food. Here, we develop a mechanistic geographical information system (GIS) model to produce spatial projections of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for the USA. We investigate the influence of gender, food, and drinking water on the predicted values by comparing them with the published values. The strongest influence on the predicted values was related to the source of  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values. We combine the model with equations that describe the rate of turnover to produce estimates for the time required for a non-resident to reach an isotopic equilibrium with a resident population.

**Keywords:** body water; diagnostic stable isotope application; human; isoscapes; isotope ecology; hydrogen-2; model; oxygen-18; population; USA; mechanistic model; stable isotopes

### 1. Introduction

The hydrogen (H) and oxygen (O) isotopic composition of body water ( $\delta_{\text{bw}}$ ) influences the isotopic composition of tissues that are commonly used to investigate the movement patterns of humans and animals [1–3]. For example, the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of hair, bones, and teeth have been used to identify the location of origin for unknown samples and to track migration and resource use in modern and ancient human populations [4–6]. In general,  $\delta_{\text{bw}}$  and these tissues are positively correlated with the isotopic composition of meteoric precipitation [7–11]. The isotopic composition of meteoric precipitation varies globally, with higher latitudes and higher elevations having lower  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values than the low-latitude and coastal areas [12–14]. As a result, the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of keratin-based tissues such as feathers [4], hair [5,15], and nails [16] have been used to predict the regions of origin for these samples.

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The slope of the relationship between the  $\delta^2\text{H}$  values of meteoric water and those of keratin-based tissues can approach unity [4], but see [3,17]. In contrast, the slope of  $\delta^{18}\text{O}$  values will never approach unity because of the incorporation of atmospheric  $\text{O}_2$  into the body water pool during cellular respiration and the subsequent flow of O into keratin and other tissues [7,18]. In a comparison between the isotopic compositions of drinking water ( $\delta_{\text{dw}}$ ) and hair ( $\delta_{\text{h}}$ ) for modern humans, the slope of the relationship between  $\delta^2\text{H}$  values was 0.27 and the slope of the relationship between  $\delta^{18}\text{O}$  values was 0.35, indicating that drinking water directly accounted for 27 and 35% of the H and O atoms in hair, respectively [5]. Drinking water was the largest single contributor of H and O to  $\delta_{\text{h}}$  values, but other factors, such as  $\delta_{\text{bw}}$ , may have influenced  $\delta_{\text{h}}$  values. The conclusions about the region of origin of a sample using  $\delta_{\text{h}}$  values could be improved if models describing the flow of H and O into the body water, and subsequently into hair, were more robust.

Models that estimate  $\delta_{\text{bw}}$  have been developed [7,18,19]. Basic models use a linear regression to relate  $\delta_{\text{bw}}$  to local drinking water [19,20]. Other models use mass balance principles to model the flow of H and O into and out of the body to estimate  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values [7,18,21,22].  $\delta_{\text{bw}}$  is influenced by  $\delta_{\text{dw}}$ , diet, and climate [7,9,19,21]. The early mass balance models incorporated only major influxes and effluxes of H and O, whereas the model developed by Kohn [7] was more complex and included the influence of temperature and humidity on the evaporative enrichment of body water.

As a theoretical exercise, we used the Kohn model as a framework to develop a mass balance model to estimate the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for humans. We combined this model with spatial maps that predict the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  of tap water and meteoric water in ARCGIS 9.1<sup>®</sup> to produce spatial representations of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for the contiguous USA [23]. We used the tap water maps as the best representation of the isotopic composition of the water consumed by humans in the contiguous USA. We compared these estimates with those produced using meteoric precipitation to evaluate differences in the estimated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values that are related to drinking water sources. In most cases, meteoric precipitation maps will not provide accurate representations of the isotopic composition of human drinking water obtained from groundwater and surface water. However, accurate maps of the isotopic composition of precipitation water are readily available for most areas of the globe, whereas the coverage of surface and groundwater maps is relatively limited. Thus, it is useful to assess the degree to which the use of precipitation maps, as opposed to that of true tap water maps, as input to the model influences the predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values.

We also investigated the influence of the isotopic composition of food on the estimated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values by comparing values estimated using two different source layers for food values. The first source layer linked the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food with the  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values. The second source layer used one  $\delta^2\text{H}$  value and one  $\delta^{18}\text{O}$  value for food for the entire USA. Food is transported widely across the USA and, thus, food consumed in one location may not be isotopically linked to a specific locale. As a result, individuals who live in areas with different  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values may have similarities in the isotopic composition of their food. Given the paucity of published values on the  $^2\text{H}$  and  $^{18}\text{O}$  composition of modern foods, it is difficult to assess the degree to which the source layers used here reflect reality, but rather the layers are selected to represent end-member cases allowing us to test the sensitivity of the model to uncertainty in the spatial distribution of food isotopic compositions.

We compared the estimated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values with the published values to test the various permutations of our model. This method of testing model robustness also has limitations due to the dearth of published values. However, this technique allows for an initial test of the model, with future versions of the model being improved after a coordinated collection of body water samples from humans across large geographical areas. Lastly, we demonstrate that this model could be used to infer movement patterns of humans across broad geographical gradients, distinguishing residents at isotopic equilibrium from recent non-resident arrivals.

## 2. Development of a spatial model

### 2.1. Development of a human body water model

We developed a model that estimates the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for a human based on the daily influxes and effluxes of H and O. The body water pool represents the total water content of the body including intra- and extracellular water. At the scale of the whole body, the  $\delta_{\text{bw}}$  pool is assumed to be one pool and isotopically uniform. A 40-year-old male who has a mass of  $\sim 82$  kg has a total body water pool of  $\sim 41$  kg, and a 40-year-old female who has a mass of  $\sim 74$  kg has a total body water pool of  $\sim 31$  kg [24]. Our body water model is based on the O mass balance model developed by Luz *et al.* [25] and modified by Schoeller *et al.* [22] for H and O. The sources contributing H and/or O to body water include drinking water, free water in food, H and O bound in ingested organic molecules (food), atmospheric  $\text{O}_2$ , and atmospheric water vapour. Outputs of H and/or O include liquid water in the form of urine, sweat, and faecal water, water vapour loss associated with breathing and evaporation across the skin, and carbon dioxide loss. We used the following mass balance equations to estimate  $\delta_{\text{bw}}$  and we have represented the isotopic enrichments as molar ratios ( $R$ ). At steady-state conditions where the flow of H and O into the body water of a human equals the flow of H and O out of the human, the  $^2\text{H}$  and  $^{18}\text{O}$  composition of body water can be estimated for a one-input and one-output system as

$$x_{\text{in}} \times R_{\text{in}} = x_{\text{out}} \times R_{\text{out}} \times \alpha_{\text{bw}}, \quad (1)$$

where  $x$  is the amount of the input or output,  $R$  is the ratio of the heavy stable isotope to the light stable isotope ( $R_{\text{in}} = \text{inputs}$  and  $R_{\text{out}} = \text{outputs}$ ), and  $\alpha_{\text{bw}}$  is the fractionation between body water and the output. Since there are multiple inputs and outputs of H and O and the fractionations associated with each output can be estimated,  $R_{\text{bodywater}}$  ( $R_{\text{bw}}$ ) can be estimated at steady-state conditions by

$$R_{\text{bw}} = \frac{\sum_{i=1}^n x_{\text{in},i} \times R_{\text{in},i}}{\sum_{g=1}^x x_{\text{out},g} \times \alpha_{\text{out},g}}. \quad (2)$$

Inserting the specific inputs and outputs for  $^2\text{H}$  in body water into the above equation yields

$$R_{^2\text{H}_{\text{bw}}} = \frac{(x_{\text{dw}}R_{\text{dw}} + x_{\text{fw}}R_{\text{fw}} + x_{\text{v}}R_{\text{v}} + x_{\text{wvg}}R_{\text{wvg}})}{(x_{\text{bwl}}\alpha_{\text{bwl}} + x_{\text{tv1}}\alpha_{\text{tv1}} + x_{\text{rwl}}\alpha_{\text{rwl}})} \quad (3)$$

and for  $^{18}\text{O}$  in body water results in

$$R_{^{18}\text{O}_{\text{bw}}} = \frac{(x_{\text{dw}}R_{\text{dw}} + x_{\text{fw}}R_{\text{fw}} + x_{\text{v}}R_{\text{v}} + x_{\text{wvg}}R_{\text{wvg}} + x_{\text{O}_2}R_{\text{O}_2})}{(x_{\text{CO}_2}\alpha_{\text{CO}_2} + x_{\text{bwl}}\alpha_{\text{bwl}} + x_{\text{tv1}}\alpha_{\text{tv1}} + x_{\text{rwl}}\alpha_{\text{rwl}})}, \quad (4)$$

where  $x$  is the mole fraction of drinking water (dw), food water (fw), H or O bound in food (v), water vapour gain (wvg), diatomic O inspired from the atmosphere ( $\text{O}_2$ ), breath water expelled (bwl), transcutaneous water loss (tv1), carbon dioxide expelled to the atmosphere ( $\text{CO}_2$ ), and remaining water loss (rwl) including urine, faecal water, and sweat. We combined urine, faecal water, and sweat loss into one term because all were assumed to be unfractionated relative to body water, whereas transcutaneous water loss, carbon dioxide loss, and breath water were all assumed to be related to body water by fractionation factors (Tables 1 and 2).

We created the model and then parameterised it for an adult male and for an adult female eating an average American diet (Table 1). This model was based on the Kohn model [7] that estimated the  $\delta^{18}\text{O}_{\text{bw}}$  values for animals. We modified the Kohn model to more closely match the expected values for humans and we also expanded the model to predict  $\delta^2\text{H}_{\text{bw}}$  values. We

Table 1. Quantity, symbol, unit, values, and literature source used in the four parameterisations ( $V_{-1}^a$ ,  $V_{-2}^b$ ,  $V_{-3}^c$ , and  $V_{-4}^d$ ) of the body water model.

Quantity	Symbol	Unit	$V_{-1}$	$V_{-2}$	$V_{-3}$	$V_{-4}$	Source
Mass	$m_b$	kg	83	75	83	83	[24]
Height	$h$	cm	180	164	180	180	[24]
Total energy expenditure	$P_j$	$\text{kJ d}^{-1}$	Equation (5)	Equation (5)	Equation (5)	Equation (5)	[26]
Age	$Y$	years	40	40	40	40	[24]
Activity level	$A$		1.6	1.6	1.6	1.6	[26]
Mass of food	$m_f$	kg	Equation (6)	Equation (6)	Equation (6)	Equation (6)	[7]
Extraction efficiency	$\phi$		1	1	1	1	[7]
Digestibility	$\gamma$		0.85	0.85	0.85	0.85	[7]
Fraction of carbohydrate in diet	$w_c$		0.53	0.53	0.53	0.53	[24]
Fraction of fat in diet	$w_f$		0.32	0.32	0.32	0.32	[24]
Fraction of protein in diet	$w_p$		0.15	0.15	0.15	0.15	[24]
Amount of H and O in food	$m_v$	mol	Equation (7)	Equation (7)	Equation (7)	Equation (7)	[7]
Amount of diatomic oxygen	$m_{O_2}$	mol	Equation (8)	Equation (8)	Equation (8)	Equation (8)	[7]
Amount of water in food	$m_{f_w}$	mol	Equation (9)	Equation (9)	Equation (9)	Equation (9)	[7]
Fraction of water content in food	$W_{cf}$		0.65	0.65	0.65	0.65	[7]
Air flow through lungs	$\phi_{air}$	l	Equation (10)	Equation (10)	Equation (10)	Equation (10)	[7]
Amount of H and O gain in lungs	$m_{wvg}$	mol	Equation (11)	Equation (11)	Equation (11)	Equation (11)	[7]
Ambient relative humidity	$H_a$		MRH <sup>c</sup>	MRH	MRH	MRH	MRH
Lung relative humidity	$H_l$		1.0	1.0	1.0	1.0	
Ambient temperature	$t_a$	°C	MAT <sup>f</sup>	MAT	MAT	MAT	MAT
Ambient thermodynamic temperature	$T_k$	K	Convert	Convert	Convert	Convert	Convert $t_a$ to $T_k$
Lung temperature	$t_l$	°C	33	33	33	33	
Amount of H and O in drinking water	$m_{d_w}$	mol	Equation (12)	Equation (12)	Equation (12)	Equation (12)	[24]
Total amount of flow of H and O	$m_{WFj}$	mol	211.11	183.3	211.11	211.11	[24]
Amount of O lost in CO <sub>2</sub>	$m_{CO_2}$	mol	Equation (13)	Equation (13)	Equation (13)	Equation (13)	[7]
Surface area	$S$	m <sup>2</sup>	Equation (14)	Equation (14)	Equation (14)	Equation (14)	[24]

Amount of H and O lost transcutaneously	$m_{tv1}$	mol	Equation (15)	Equation (15)	Equation (15)	Equation (15)	[7]
Amount of H and O lost through breathing	$m_{bw1}$	mol	Equation (16)	Equation (16)	Equation (16)	Equation (16)	[7]
Amount of remaining H and O lost	$m_{rw1}$	mol	Equation (17)	Equation (17)	Equation (17)	Equation (17)	[7]
$\delta^{18}\text{O}$ diatomic oxygen	$\delta^{18}\text{O}_{\text{O}_2}$	‰	15.3	15.3	15.3	15.3	[27]
$\delta^2\text{H}$ drinking water	$\delta^2\text{H}_{\text{dw}}$	‰	Tap <sup>g</sup>	Tap	Tap	Precip. <sup>h</sup>	[23]; www.waterisotopes.org OIPC
$\delta^{18}\text{O}$ drinking water	$\delta^{18}\text{O}_{\text{dw}}$	‰	Tap	Tap	Tap	Precip.	[23]; www.waterisotopes.org OIPC
$\delta^2\text{H}$ food	$\delta^2\text{H}_{\text{fd}}$	‰	Equation (18)	Equation (18)	-115	Equation (18)	
$\delta^{18}\text{O}$ food	$\delta^{18}\text{O}_{\text{fd}}$	‰	Equation (18)	Equation (18)	32	Equation (18)	
$\delta^2\text{H}$ food water	$\delta^2\text{H}_{\text{fw}}$	‰	Equation (19)	Equation (19)	Equation (19)	Equation (19)	[28]
$\delta^{18}\text{O}$ food water	$\delta^{18}\text{O}_{\text{fw}}$	‰	Equation (20)	Equation (20)	Equation (20)	Equation (20)	[28]

<sup>a</sup>V\_1, model for an adult male created using the estimated  $\delta$  values of tap water.

<sup>b</sup>V\_2, model for an adult female created using the estimated  $\delta$  values of tap water.

<sup>c</sup>V\_3, model for an adult male created using the estimated  $\delta$  values of tap water and the  $\delta$  values of food held constant.

<sup>d</sup>V\_4, model for an adult male created using the estimated  $\delta$  values of precipitation.

<sup>e</sup>MRH, mean relative humidity.

<sup>f</sup>MAT, mean annual temperature.

<sup>g</sup>Tap represents the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values of local tap water modelled by Bowen *et al.* [23].

<sup>h</sup>Precip. represents the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of local precipitation, estimated using OIPC at www.waterisotopes.org.

<sup>i</sup>OIPC, online isotopes in precipitation calculator.

Table 2. Equation number, value, unit of measure, and source of constants used to predict  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values in body water models V\_1<sup>a</sup>, V\_2<sup>b</sup>, V\_3<sup>c</sup>, and V\_4<sup>d</sup>.

Equation	Constants	Value	Unit	Source
Equation (5)	$c_{1,1}$	66		[26]
Equation (5)	$c_{2,1}$	13.8		[26]
Equation (5)	$c_{3,1}$	5.0		[26]
Equation (5)	$c_{4,1}$	6.8		[26]
Equation (5)	$c_5$	4.1868		[26]
Equation (5)	$c_{1,2}$	655		[26]
Equation (5)	$c_{2,2}$	9.6		[26]
Equation (5)	$c_{3,2}$	1.9		[26]
Equation (5)	$c_{4,2}$	4.7		[26]
Equation (6)	$k_1$	17.3	$\text{kJ kg}^{-1}$	[7]
Equation (6)	$k_2$	39.7	$\text{kJ kg}^{-1}$	[7]
Equation (6)	$k_3$	20.1	$\text{kJ kg}^{-1}$	[7]
Equation (7)	$f_{1,1}$	30.9	$\text{mol kg}^{-1}$	[7]
Equation (7)	$f_{2,1}$	60.0	$\text{mol kg}^{-1}$	[7]
Equation (7)	$f_{3,1}$	11.0	$\text{mol kg}^{-1}$	[7]
Equation (7)	$f_{1,2}$	15.4	$\text{mol kg}^{-1}$	[7]
Equation (7)	$f_{2,2}$	2.0	$\text{mol kg}^{-1}$	[7]
Equation (7)	$f_{3,2}$	3.0	$\text{mol kg}^{-1}$	[7]
Equation (8)	$k_4$	0.00216	$\text{mol O}_2 \text{kJ}^{-1}$	[7]
Equation (9)	$k_5$	55.56	$\text{mol kg}^{-1}$	[7]
Equation (10)	$k_6$	22.4	$\text{l mol}^{-1}$	[7]
Equation (10)	$k_7$	0.20	% efficiency lungs	[7]
Equation (10)	$k_8$	0.21	% O <sub>2</sub> in atm	[7]
Equations (11) and (16)	$k_9$	10		[7]
Equations (11) and (16)	$k_{10}$	0.686		[7]
Equations (11) and (16)	$k_{11}$	0.027		[7]
Equations (11) and (16)	$k_{12}$	760	Torr	[7]
Equation (13)	$k_{13}$	2.0		[7]
Equation (14)	$k_{14}$	3600		[24]
Equation (15)	$k_{15}$	0.14	$\text{g min}^{-1}\text{m}^{-2}$	[24]
Equation (15)	$k_{16}$	18	Moles $\text{g}^{-1}$	
Equation (18)	$a_{1,1}$	-30 <sup>e</sup>	%o	[29]
Equation (18)	$a_{2,1}$	-167 <sup>e</sup>	%o	[30]
Equation (18)	$a_{3,1}$	-33 <sup>e</sup>	%o	[31]
Equation (18)	$a_{1,2}$	27 <sup>e</sup>	%o	[29]
Equation (18)	$a_{2,2}$	22 <sup>e</sup>	%o	[32]
Equation (18)	$a_{3,2}$	33 <sup>e</sup>	%o	[33]
Equation (19)	$k_{17}$	0.69	$\delta^2\text{H}_{\text{fw}}/\delta^2\text{H}_{\text{dw}}$	[28]
Equation (19)	$k_{18}$	3.8	%o	[28]
Equation (20)	$k_{19}$	4.0	%o	[28]
Equation (21)	$k_{20}$	1158.8		[34]
Equations (21) and (22)	$k_{21}$	10 <sup>9</sup>		[34]
Equation (21)	$k_{22}$	1620.1		[34]
Equations (21) and (22)	$k_{23}$	10 <sup>6</sup>		[34]
Equation (21)	$k_{24}$	794.84		[34]
Equations (21) and (22)	$k_{25}$	10 <sup>3</sup>		[34]
Equation (21)	$k_{26}$	161.04		[34]
Equation (21)	$k_{27}$	2.9992		[34]
Equation (22)	$k_{28}$	-7.685		[34]
Equation (22)	$k_{29}$	6.7123		[34]
Equation (22)	$k_{30}$	1.6664		[34]
Equation (22)	$k_{31}$	0.35041		[34]
Hydrogen $\alpha$ values	$\alpha_{\text{Hwater-vapour}}$	Variable		[34]
	$\alpha_{\text{Htv1-bw}}$	0.935		[22]
	$\alpha_{\text{Hbw1-bw}}$	0.946		[22]
Oxygen $\alpha$ values	$\alpha_{\text{Owater-vapour}}$	Variable		[34]
	$\alpha_{\text{Otv1-bw}}$	0.981		[22]
	$\alpha_{\text{Obw1-bw}}$	0.991		[22]
	$\alpha_{\text{OCO}_2\text{-bw}}$	1.038		[35]

<sup>a</sup>V\_1, model for an adult male created using the estimated  $\delta$  values of tap water.<sup>b</sup>V\_2, model for an adult female created using the estimated  $\delta$  values of tap water.<sup>c</sup>V\_3, model for an adult male created using the estimated  $\delta$  values of tap water and the  $\delta$  values of food held constant.<sup>d</sup>V\_4, model for an adult male created using the estimated  $\delta$  values of precipitation.<sup>e</sup>These values can be modified to increase the fit of the model to the measured values.



then estimated the mole fractions and fractionation factors that were subsequently input into Equations (3) and (4) to estimate the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  for humans. This model was constructed and tested using spreadsheet software before incorporating it into a geographical information system (GIS) platform.

### 2.1.1. Calculation of the amount of each input

We used the Harris–Benedict equations [26] to estimate the daily total energy expenditure ( $P_j$ ) for adult males ( $j = 1$ ) and adult females ( $j = 2$ ):

$$P_j = (c_{1,j} + c_{2,j}m_b + c_{3,j}h - c_{4,j}Y)Ac_5, \quad (5)$$

where  $m_b$  is the mass,  $h$  is the height,  $Y$  is the age, and  $A$  is the activity level of the individual (see Tables 1 and 2 for the values and descriptions for all variables and constants used in Equations (5)–(22)). Next, we determined the mass of food consumed ( $m_f$ ) by the individual to fuel  $P_j$ :

$$m_f = \frac{P_j}{[\phi\gamma(k_1w_c + k_2w_f + k_3w_p)]}, \quad (6)$$

where  $\phi$  is the extraction efficiency,  $\gamma$  is the digestibility of the diet,  $w_c$  is the fraction of carbohydrate in the diet,  $w_f$  is the fraction of fat in the diet, and  $w_p$  is the fraction of protein in the diet ( $w_c + w_f + w_p = 1$ ). We then calculated the amount of H ( $q = 1$ ) and O ( $q = 2$ ) in the mass of food consumed ( $m_{v,q}$ ) as

$$m_{v,q} = m_f\phi\gamma(f_{1,q}w_c + f_{2,q}w_f + f_{3,q}w_p). \quad (7)$$

Next, we calculated the amount of  $\text{O}_2$  ( $m_{\text{O}_2}$ ) required to fuel the estimated  $P_j$ :

$$m_{\text{O}_2} = k_4P_j. \quad (8)$$

We then determined the amount of food water ( $m_{\text{fw}}$ ) in the mass of food consumed as

$$m_{\text{fw}} = \frac{k_5m_fW_{\text{cf}}}{1 - W_{\text{cf}}}, \quad (9)$$

where  $W_{\text{cf}}$  is the fraction of water in food. Next, we calculated the air flow through the lungs ( $\text{O}_{\text{air}}$ ) as

$$\text{O}_{\text{air}} = \frac{k_6m_{\text{O}_2}}{(k_7k_8)}. \quad (10)$$

We then determined the amount of water gain ( $m_{\text{wvg}}$ ) across the lungs as

$$m_{\text{wvg}} = \frac{[H_a k_9^{(k_{10} + k_{11}t_a)} \text{O}_{\text{air}}]}{(k_{12}k_6)}, \quad (11)$$

where  $H_a$  is the ambient relative humidity and  $t_a$  is the ambient temperature. We used a value of 58% for  $H_a$  and a value of 17.6°C for  $t_a$  to develop the model. Next, we calculated the amount of drinking water ( $m_{\text{dw}}$ ) necessary to balance the daily water budget by using the total water flow ( $m_{\text{WF}j}$ ) for an individual as

$$m_{\text{dw}} = m_{\text{WF}j} - m_{\text{wvg}} - m_{v,q} - m_{\text{fw}}. \quad (12)$$

We used a mean total water flow of 3.81 day<sup>-1</sup> for an adult male and of 3.31 day<sup>-1</sup> for an adult female [24].

### 2.1.2. Calculation of the amount of each output

We calculated the amount of each output of H and O. We began by determining the amount of oxygen lost as carbon dioxide ( $m_{\text{CO}_2}$ ) as

$$m_{\text{CO}_2} = m_{\text{O}_2} - \frac{m_{\text{v},q}}{k_{13}} - m_{\text{v},2}. \quad (13)$$

Next, we calculated the amount of water lost as transcutaneous water vapour ( $m_{\text{tvI}}$ ) by using the Dubois formula to calculate the surface area ( $S$ ) for the individual [24]:

$$S = \sqrt{\frac{hm_b}{k_{14}}}, \quad (14)$$

and we used the following equation to calculate  $m_{\text{tvI}}$  as

$$m_{\text{tvI}} = \frac{k_{15}S}{k_{16}}. \quad (15)$$

Humans lose  $0.14 \text{ g min}^{-1} \text{ m}^{-2}$  of water through their skin [22] and we assumed that clothing decreased the amount of water lost by 50%. We converted the amount of water lost per minute to a daily water loss [22,24]. Next, we determined the amount of water lost through breathing ( $m_{\text{bwI}}$ ) as

$$m_{\text{bwI}} = \frac{[H_1 k_9^{(k_{10}+k_{11}t_1)} \text{O}_{\text{air}}]}{(k_{12}k_6)}, \quad (16)$$

where  $H_1$  is the relative humidity in the lungs (100%) and  $t_1$  is the temperature in the lungs ( $33^\circ\text{C}$ ). Lastly, we calculated the moles of water lost as urine and sweat and in the faeces. In all the calculations, we referred to these three outputs as remaining water loss ( $m_{\text{rwl}}$ ):

$$m_{\text{rwl}} = m_{\text{WFj}} - m_{\text{tvI}} - m_{\text{bwI}}. \quad (17)$$

### 2.1.3. Estimation of the isotopic composition and fractionation factors for all inputs

We estimated the isotopic composition and the fractionation factors of all inputs of H and O. We assumed that the  $\delta^{18}\text{O}$  value of  $\text{O}_2$  absorbed in the lungs was 15.3‰ [27]. Next, we estimated the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food consumed. We developed two parameterisations, one in which, the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  of food ( $\delta_{\text{fd},q}$ ) were linked by the following equations to the  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  Equation (18). Since much of the food consumed in the USA is not of a local origin and to determine the importance of food to predict the  $\delta$  values of body water, we also calculated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  using static  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food based on our and other published values [32,36]. We used a value of  $-115\text{‰}$  for the  $\delta^2\text{H}$  of food ( $\delta^2\text{H}_{\text{fd}}$ ) and that of  $32\text{‰}$  for the  $\delta^{18}\text{O}$  of food ( $\delta^{18}\text{O}_{\text{fd}}$ ):

$$\begin{aligned} \delta_{\text{fd},q} = & \left[ \frac{f_{1,q} w_c m_f \phi \gamma}{m_{\text{v},q}} (\delta_{\text{dw},q} + a_{1,q}) \right] + \left[ \frac{f_{2,q} w_1 m_f \phi \gamma}{m_{\text{v},q}} (\delta_{\text{dw},q} + a_{2,q}) \right] \\ & + \left[ \frac{f_{3,q} w_p m_f \phi \gamma}{m_{\text{v},q}} (\delta_{\text{dw},q} + a_{3,q}) \right]. \end{aligned} \quad (18)$$

Next, using the values reported by Boner and Förstel [28], we calculated the  $\delta^2\text{H}$  of food water ( $\delta^2\text{H}_{\text{fw}}$ ) and  $\delta^{18}\text{O}$  food water ( $\delta^{18}\text{O}_{\text{fw}}$ ):

$$\delta^2\text{H}_{\text{fw}} = k_{17} \times \delta^2\text{H}_{\text{dw}} + k_{18}, \quad (19)$$

$$\delta^{18}\text{O}_{\text{fw}} = \delta^{18}\text{O}_{\text{dw}} + k_{19}. \quad (20)$$

Next, we converted the above estimated  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values into  $R$  values. We then estimated the  $R$  value for the water vapour gain in the lungs ( $R_{\text{Hwvg}}$  and  $R_{\text{Owvg}}$ ) using the following equations for each element [34]:

$$R_{\text{Hwvg}} = \frac{R_{\text{Hdw}}}{[\text{Exp}((k_{20}T_k^3/k_{21} - k_{22}T_k^2/k_{23} + k_{24}T_k/k_{25} - k_{26} + k_{27}k_{21}/T_k^3)/k_{25})]}, \quad (21)$$

$$R_{\text{Owvg}} = \frac{R_{\text{Odw}}}{[\text{Exp}((k_{28} + k_{29}k_{25}/T_k - k_{30}k_{23}/T_k^2 + k_{31}k_{21}/T_k^3)/k_{25})]}, \quad (22)$$

where  $T_k$  is the temperature in kelvin.

#### 2.1.4. Estimation of the isotopic composition and fractionation factors for all outputs

Lastly, we estimated the isotopic composition and fractionation factors for all outputs. We used the empirically derived estimate for the equilibrium fractionation ( $a_{\text{CO}_2-\text{bw}} = 1.038$ ) between the  $\delta^{18}\text{O}_{\text{bw}}$  and  $\delta^{18}\text{O}$  values of  $\text{CO}_2$  [35]. We used the fractionation factors developed by Schoeller *et al.* [22] for the fractionation between body water and water lost transcutaneously (H:  $\alpha_{\text{Htvl}-\text{bw}} = 0.935$  and O:  $\alpha_{\text{Otl}-\text{bw}} = 0.981$ ) and for the fractionation between body water and water lost through breathing (H:  $\alpha_{\text{Hbw1}-\text{bw}} = 0.946$  and O:  $\alpha_{\text{Obw1}-\text{bw}} = 0.991$ ). We assumed that urine, sweat, and faecal water were unfractionated relative to body water [22]. The amounts,  $R$  values, and  $\alpha$  values for all inputs and outputs were used in Equations (3) and (4) to predict the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values.

#### 2.1.5. Validation of the estimates of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ of body water

We used  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values for Chicago, IL [22], and Houston, TX [37], to develop our spreadsheet model and we compared our estimated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values with the published values for Chicago [22] and with the published  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for Houston [37]. The published  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values for Chicago, IL, were  $-48$  and  $-6.6\text{‰}$ , respectively. The published  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  for Houston, TX, were  $-16$  and  $-2.0\text{‰}$ , respectively.

## 2.2. Creation of a spatial map of the predicted $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of body water

We input all equations into ArcGIS 9.1 ModelBuilder<sup>®</sup> to produce spatially explicit representations of the equilibrium  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for the contiguous USA. This produced mechanistic estimates of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  for each pixel within a raster that covers the contiguous USA. Since ambient temperature and humidity levels affect the estimates of  $\delta_{\text{bw}}$ , spatial representations included data layers of mean annual temperature (MAT) and mean relative humidity (MRH) (CRU CL 2.0;  $10' \times 10'$ ; <http://www.cru.uea.ac.uk/cru/data/tmc.htm>) and data layers for the estimated  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values.

We created four parameterisations of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values as maps, representing combinations of sex and diet. The first two parameterisations were an adult male (V\_1) and an adult female (V\_2). We linked the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food to local drinking water (Equation (18)) for parameterisations V\_1 and V\_2; the source of the estimated  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values was the spatial map of tap water created by Bowen *et al.* [23] for the USA. The third parameterisation was a male (V\_3) with fixed values for food inputs ( $\delta^2\text{H} = -115\text{‰}$  and  $\delta^{18}\text{O} = 32\text{‰}$ ). Lastly, differences between the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of local tap water and local precipitation can be  $>40\text{‰}$  for H and  $>7.0\text{‰}$  for O [23]. Therefore, the fourth parameterisation (V\_4) was a male with drinking water inputs equal to those of precipitation and not to those of tap water. Here, we

used the estimated  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of local precipitation based on the Online Isotopes in Precipitation Calculator ([www.waterisotopes.org](http://www.waterisotopes.org)). We compared the four parameterisations with the published  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values to test and validate each parameterisation.

### 2.3. Application of the spatial body water model to distinguish locals from visitors

We combined parameterisation V\_1 of the GIS body water model with the published turnover constants of human body water to estimate the length of time required for a resident of Houston, TX, who relocated to Chicago, IL, and Salt Lake City, UT, to become indistinguishable from residents of these two cities. We assumed that the turnover of bulk human body water is a one-pool system and we predicted  $\delta^2\text{H}_{\text{bw}}$  at time  $t$  as

$$\delta^2\text{H}_{\text{bw}}^t = \delta^2\text{H}^{\text{eq}} + a e^{-\lambda t}, \quad (23)$$

where  $\delta^2\text{H}^{\text{eq}}$  is the predicted isotope equilibrium (i.e. steady-state) value for each location (Chicago or Salt Lake City),  $a$  is the difference between the final ( $\delta^2\text{H}^{\text{eq}}$ ) and initial isotope values ( $\delta^2\text{H}^{\text{init}}$ ), and  $t$  is the time since arrival from Houston.  $\lambda$  is a first-order rate constant that can be calculated from the published values of the half-life of body water ( $t_{1/2}$ ). The measured half-life of body water for a male adult is between 5.9 and 7.5 days [38].  $\lambda$  can be calculated as

$$\lambda = \frac{\ln(2)}{t_{1/2}}. \quad (24)$$

Based on the published  $\delta_{\text{bw}}$  values and the associated uncertainties, we assumed that the non-resident was indistinguishable from residents when the  $\delta^2\text{H}_{\text{bw}}$  value of the non-resident was  $\leq 4.5\%$  different from the  $\delta^2\text{H}_{\text{bw}}$  values of the residents [22,39].

## 3. Results

### 3.1. Steady-state body water model predictions

The spreadsheet body water calculations for adult males and females estimated the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values within the expected values for individuals residing in both Houston, TX, and Chicago, IL (Table 3). Only slight differences were predicted to occur between the estimated  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for men and women. After consideration of water input, the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food had the greatest influence on  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values (Table 3). Despite these differences, the ranges of estimated values were still within the expected range of values for each location (Table 3). Therefore, we accept that the body water model presented here reasonably captures the expected ranges in body water isotope ratios.

### 3.2. Steady-state body water model projections

We selected four parameterisations of the body water model ( $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$ ). The predicted values for males (V\_1), females (V\_2), and males with the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food held constant (V\_3) were similar, and when the modelled values were regressed against the measured values, the slope of the lines were indistinguishable from 1 for all the three models (Table 4; Figure 1). The parameterisation resulting in the greatest difference between the predicted and observed values was V\_4, the parameterisation in which the predicted  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of precipitation were used for drinking water. Here, the slope of the relationship between the measured and predicted values was significantly less than 1 (Table 4; Figure 1).

Table 3. Measured  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values, measured  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values, and predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values ( $\pm 1\sigma$ ) for Houston and Chicago calculated for males and females with food linked to local drinking water (Equation (18)) and for  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food held constant ( $\delta^2\text{H}_{\text{fd}} = -115\text{‰}$ ;  $\delta^{18}\text{O}_{\text{fd}} = 32\text{‰}$ ).

Location	Estimated		Observed $\delta^2\text{H}_{\text{bw}}$	Predicted $\delta^2\text{H}$ body water				Observed $\delta^{18}\text{O}_{\text{bw}}$	Predicted $\delta^{18}\text{O}$ body water				Source
	$\delta^2\text{H}_{\text{dw}}$	$\delta^{18}\text{O}_{\text{dw}}$		Food linked to $\delta^2\text{H}_{\text{dw}}$		$\delta^2\text{H}_{\text{fd}} = -115\text{‰}$			Food linked to $\delta^{18}\text{O}_{\text{dw}}$		$\delta^{18}\text{O}_{\text{fd}} = 32\text{‰}$		
			Males	Females	Males	Females	Males	Females					
Chicago	-48	-6.6	$-37 \pm 5$	-41	-41	-37	-38	$-4.8 \pm 0.6$	-4.6	-4.7	-4.2	-4.3	[22]
Houston	-16	-2.0	$-13.5 \pm 2.7$	-12	-12	-11	-11	$-1.4 \pm 1.3$	-1.0	-1.0	-0.8	-0.7	[37]

Note: The last column is the source for the measured  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values and  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values.

Table 4. Latitude and longitude, estimated  $\delta^2\text{H}_{\text{dw}}$  and  $\delta^{18}\text{O}_{\text{dw}}$  values, measured  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values, and predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values from the four parameterisations of the body water model ( $\pm 1\sigma$ ).

Location	Latitude	Longitude	Modelled		Measured	Modelled $\delta^2\text{H}$ body water				Lon <sup>e</sup>	Kohn <sup>f</sup>	Source
			$\delta^2\text{H}_{\text{dw}}$	$\delta^{18}\text{O}_{\text{dw}}$	$\delta^2\text{H}_{\text{bw}}$	V_1 <sup>a</sup>	V_2 <sup>b</sup>	V_3 <sup>c</sup>	V_4 <sup>d</sup>			
Albuquerque, NM	35.0844	-106.6506	-77	NA	-78	-68	-67	-61	-70	NA	NA	[1]
Chicago, IL	41.8500	-87.6500	-48	-6.6	-37 ± 5	-40	-40	-37	-44	-3.3	-4.7	[22]
Harvard, MA	42.5000	-71.5833	-57	-8.6	-50 ± 7	-49	-49	-45	-53	-4.5	-6.4	[40]
Houston, TX	29.7631	-95.3631	-16	-2.0	-14 ± 3	-12	-12	-12	-23	-0.5	-1.1	[37]
Houston, TX	29.7631	-95.3631	-16	-2.0	-13 ± 6	-12	-12	-12	-23	-0.5	-1.1	[21]
Jerico, VT	44.5039	-72.9981	-74	-10.6	-67 ± 7	-65	-64	-59	-66	-5.7	-8.0	[40]
Madison, WI	43.0731	-89.4011	-55	-7.7	-42 ± 5	-47	-46	-43	-50	-4.0	-5.6	[39]
Mt. Rainier, WA	46.800	-121.800	-113	-15.1	-86 ± 1	-107	-106	-98	-90	-8.4	-12.7	[41]
					Slope	1.09 ± 0.13	1.07 ± 0.13	0.99 ± 0.12	0.83 ± 0.06	0.74 ± 0.07	1.05 ± 0.12	
					Intercept	2.77 ± 7.0	2.54 ± 7.0	1.47 ± 6.7	-12.24 ± 3.1	-0.08 ± 0.4	-0.30 ± 0.75	
					$r^2$	0.92	0.92	0.91	0.97	0.96	0.94	
Location	Latitude	Longitude	Measured		Modelled $\delta^{18}\text{O}$ body water				Lon <sup>e</sup>	Kohn <sup>f</sup>	Source	
			$\delta^{18}\text{O}_{\text{bw}}$	V_1	V_2	V_3	V_4					
Albuquerque, NM	35.0844	-106.6506	NA	NA	NA	NA	NA	NA	NA	NA	NA	[1]
Chicago, IL	41.8500	-87.6500	-4.8 ± 0.6	-4.6	-4.5	-4.2	-5.5	-3.3	-4.7	[22]		
Harvard, MA	42.5000	-71.5833	-6.2 ± 1.4	-6.1	-6.1	-5.7	-6.9	-4.5	-6.4	[40]		
Houston, TX	29.7631	-95.3631	-1.4 ± 1.3	-1.1	-1.1	-0.9	-2.5	-0.5	-1.1	[37]		
Houston, TX	29.7631	-95.3631	0.5 ± 1.6	-1.1	-1.1	-0.9	-2.5	-0.5	-1.1	[21]		
Jerico, VT	44.5039	-72.9981	-8.2 ± 1.5	-7.8	-7.7	-7.3	-8.3	-5.7	-8.0	[40]		
Madison, WI	43.0731	-89.4011	-5.2 ± 0.7	-5.5	-5.4	-5.0	-6.1	-4.0	-5.6	[39]		
Mt. Rainier, WA	46.800	-121.800	-10.3 ± 0.1	-11.9	-11.8	-11.2	-9.8	-8.4	-12.7	[41]		
				Slope	0.99 ± 0.11	0.98 ± 0.11	0.95 ± 0.1	0.73 ± 0.05	0.74 ± 0.07	1.05 ± 0.12		
				Intercept	-0.40 ± 0.65	-0.40 ± 0.65	-0.22 ± 0.62	-2.23 ± 0.31	-0.08 ± 0.4	-0.30 ± 0.75		
				$r^2$	0.95	0.95	0.95	0.98	0.96	0.94		

Notes: The last column is the source for the measured  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values. Slope, intercept, and  $r^2$  values given at bottom of the columns ( $\pm 1$  SE) are the results obtained by comparing the predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values with the observed  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values.

<sup>a</sup>V\_1, model for an adult male created using the estimated  $\delta$  values of tap water.

<sup>b</sup>V\_2, model for an adult female created using the estimated  $\delta$  values of tap water.

<sup>c</sup>V\_3, model for an adult male created using the estimated  $\delta$  values of tap water and the  $\delta$  values of food held constant.

<sup>d</sup>V\_4, model for an adult male created using the estimated  $\delta$  values of precipitation.

<sup>e</sup>Lon, results for the Longinelli model ( $\delta^{18}\text{O}_{\text{bw}} = 0.60 \times \delta^{18}\text{O}_{\text{dw}} + 0.68$ ).

<sup>f</sup>Kohn, model from Kohn [7].

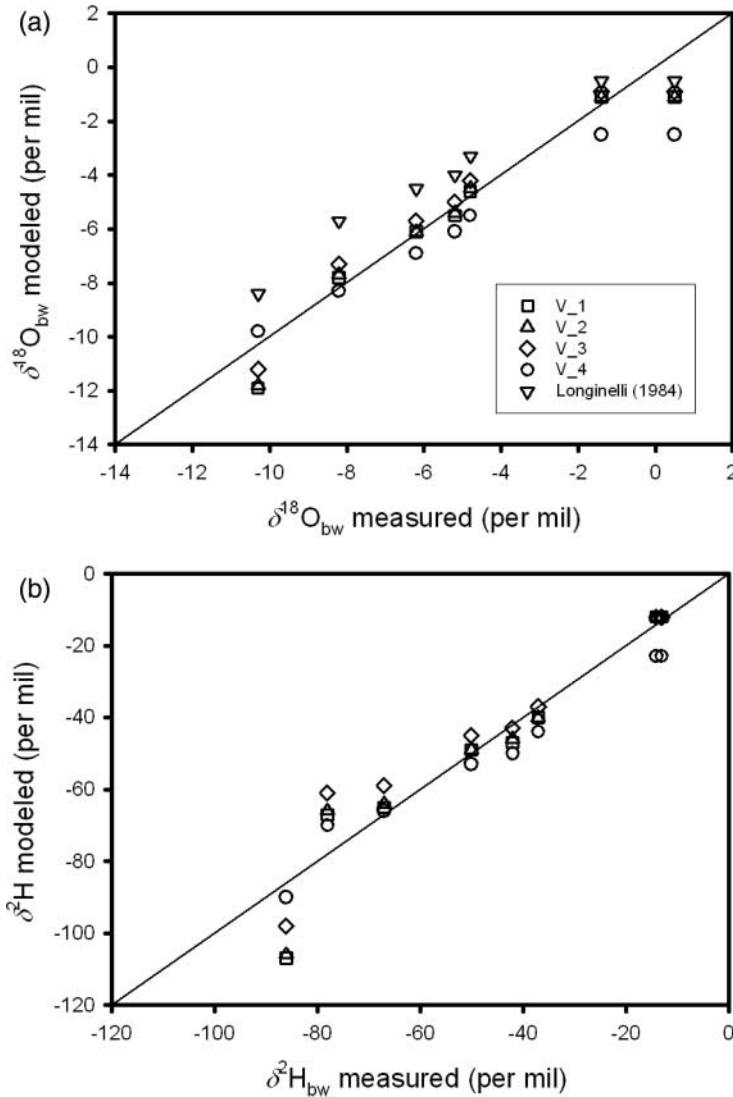


Figure 1. Comparisons between the modelled  $\delta^{18}\text{O}_{\text{bw}}$  (a) and  $\delta^2\text{H}_{\text{bw}}$  (b) values and the measured values for the four parameterisations of the body water model. V\_1: Model for an adult male created using the estimated  $\delta$  values of tap water. V\_2: Model for an adult female created using the estimated  $\delta$  values of tap water. V\_3: Model for an adult male created using the estimated  $\delta$  values of tap water and the  $\delta$  values of food held constant. V\_4: Model for an adult male created using the estimated  $\delta$  values of precipitation.

The parameterisation in which the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food were linked to tap water for an adult male (V\_1) estimated that the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values ranged from 4 to  $-147\text{‰}$  for H isotope ratios and from 1.4 to  $-16.3\text{‰}$  for O isotope ratios across the contiguous USA (Figure 2). The most positive  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values were found in interior Texas and along the Gulf Coast and the most negative values were found in the higher latitudes and elevations of the interior West (Figure 2). The predicted values for all locations, except for Mt. Rainier, were within  $10\text{‰}$  for H isotope ratios and, with the exception of Mt. Rainer and Houston, within  $0.7\text{‰}$  for O isotope ratios of published values (Table 4; Figure 2). The largest difference between the observed and predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values was for the parameterisation in which the estimated  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of precipitation were used as the values for drinking water (Table 4). The estimated

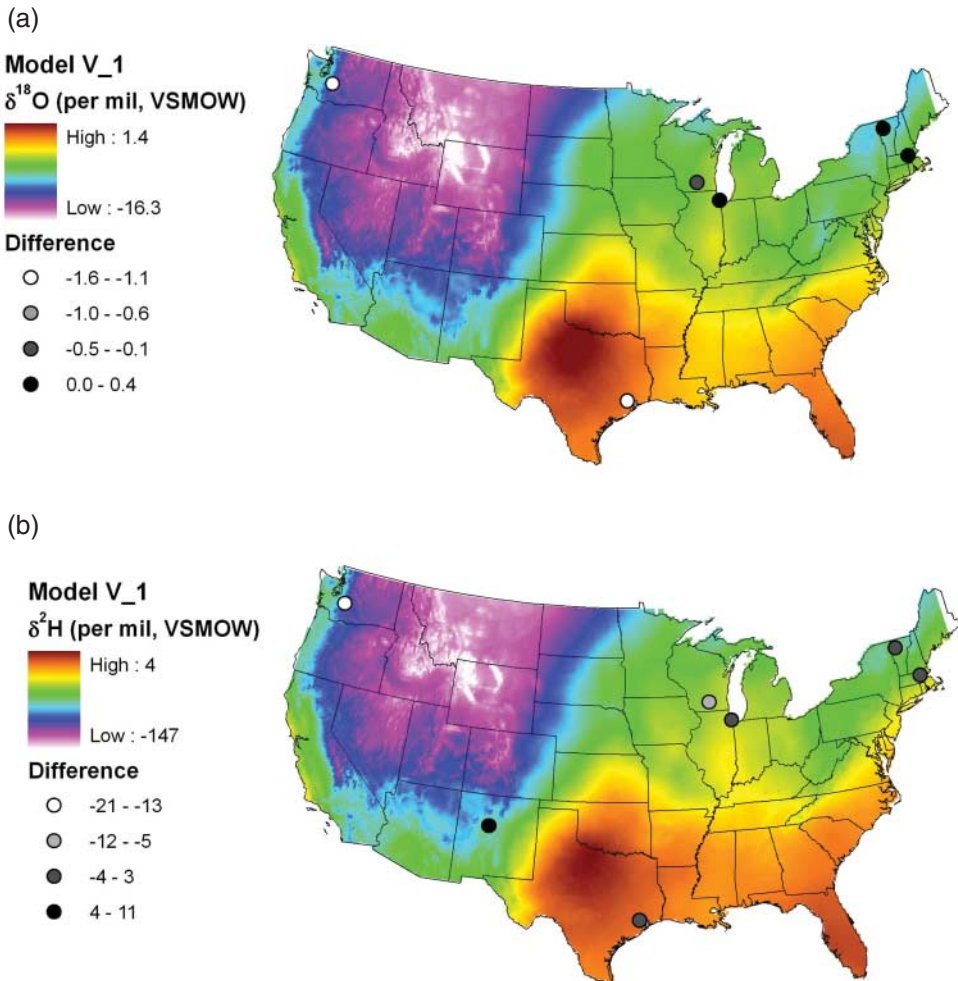


Figure 2. Comparisons between the modelled  $\delta^{18}\text{O}_{\text{bw}}$  (a) and  $\delta^2\text{H}_{\text{bw}}$  (b) values and the measured values for parameterisation V\_1 of the body water model. The round symbols indicate the locations where the measured values of body water have been analysed and the colour of the symbol reflects the difference between the measured value and the modelled value for that location. V\_1: Model for an adult male created using the estimated  $\delta$  values of tap water (spatial resolution:  $10' \times 10'$ )

$\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values from parameterisation V\_4 differed by  $>40\%$  for H isotope ratios and  $>5.5\%$  for O isotope ratios when compared with the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values estimated with V\_1 (Figure 3).

### 3.3. Application of the spatial body water model to distinguish locals from visitors

We used the  $\delta^2\text{H}_{\text{bw}}$  values estimated using parameterisation V\_1 for Houston, Chicago, and Salt Lake City combined with the estimated range in body water turnover for an adult male to predict the length of time required for the body water of an individual starting off at equilibrium with water and food in one city and then moving to become isotopically indistinguishable from a resident in a different city (Table 5). A non-resident of Chicago was indistinguishable from a resident 14–18 days after arrival, whereas a visitor to Salt Lake City was indistinguishable from residents 25–32 days after arrival (Table 5).



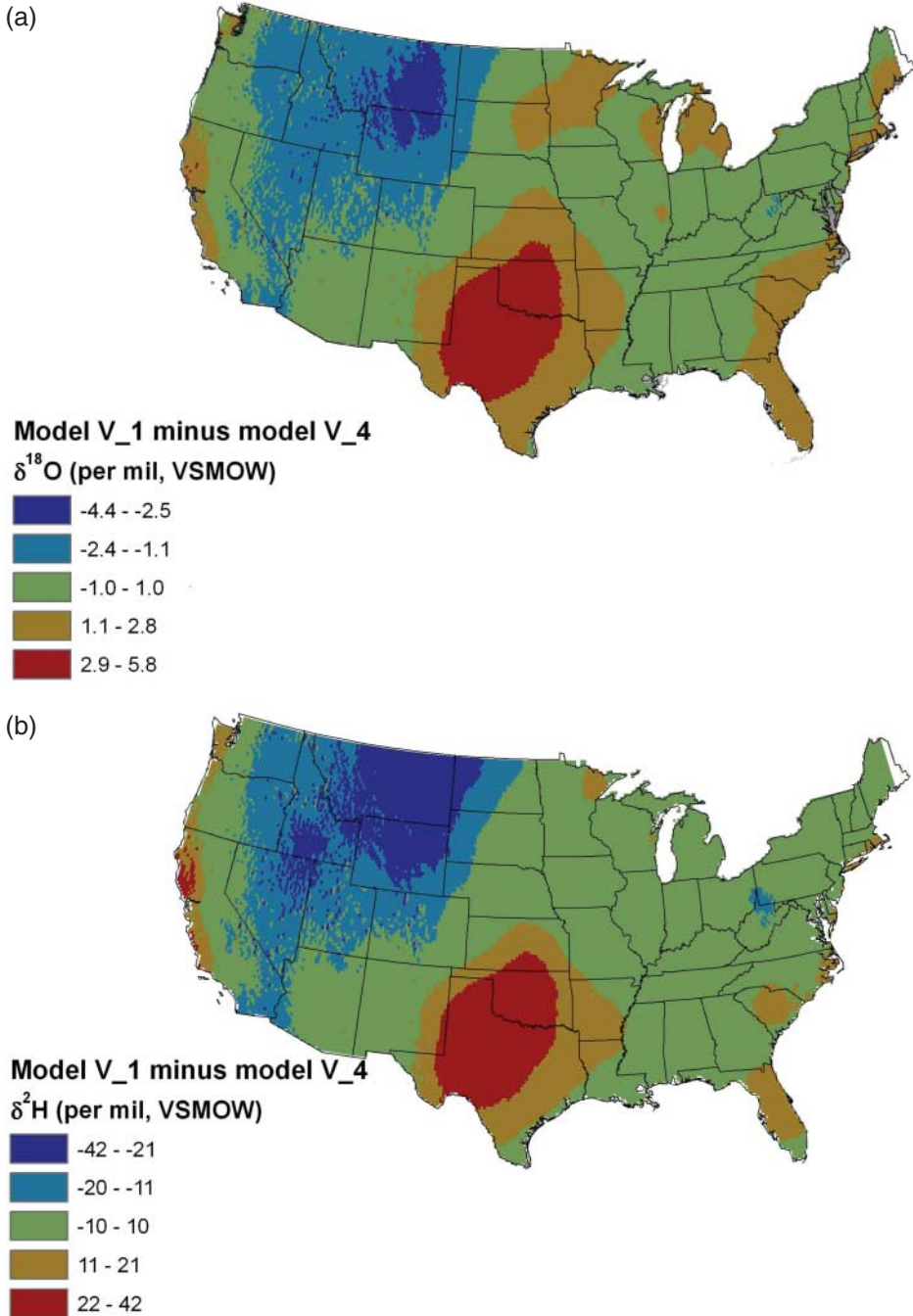


Figure 3. Differences between the modelled  $\delta^{18}\text{O}_{\text{bw}}$  (a) and  $\delta^2\text{H}_{\text{bw}}$  values (b) estimated using the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of tap water (parameterisation V\_1) and the modelled  $\delta^{18}\text{O}_{\text{bw}}$  and  $\delta^2\text{H}_{\text{bw}}$  values estimated using the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of precipitation (parameterisation V\_4). V\_1: Model for an adult male created using the estimated  $\delta$  values of tap water. V\_4: Model for an adult male created using the estimated  $\delta$  values of precipitation (spatial resolution:  $10' \times 10'$ ).

Table 5. Locations,  $\delta^2\text{H}_{\text{bw}}$  at equilibrium, total change in  $\delta^2\text{H}_{\text{bw}}$ , and the estimated length of time for  $\delta^2\text{H}_{\text{bw}}$  values of a person who travels from Houston to Chicago or to Salt Lake City to be indistinguishable from those of residents.

Location	$\delta^2\text{H}_{\text{bw}}$ ‰	Change in $\delta^2\text{H}_{\text{bw}}$ ‰	Half-lives	
			7.5 days	5.9 days
Houston, TX	-12	0.0	0	0
Chicago, IL	-40	28	18	14
Salt Lake City, UT	-102	90	32	25

Note: The length of time is calculated using two half-lives (7.5 and 5.9 days) to determine the expected range in time.

## 4. Discussion

### 4.1. Steady-state body water model predictions and projections

The mechanistic body water model predicted the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for adult males and females within an expected range of variation of 10‰ for H isotope ratios and of 0.7‰ for O isotope ratios when compared with the published values (Table 4). There were limited differences between the estimated body water values for men and women; these differences were <1 and <0.1‰ for H and O isotope ratios, respectively. These differences are well within the expected variation [22,39] and are useful because not all the published studies of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values include the gender of the individual participants.

There were also limited differences between the predictions for males and females when the mechanistic model was input into ArcGIS 9.1 ModelBuilder<sup>®</sup> to produce spatial projections of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  for the contiguous USA (Table 4). Overall, parameterisation V\_1 for men and parameterisation V\_2 for women closely matched the published  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values (Table 4; Figure 2). The body water model predictions were within  $\pm 10\%$  for H and within  $\pm 0.7\%$  for O for all locations except for Mt. Rainier (H and O) and Houston (O) (Table 4; Figure 2). These spatial representations of  $\delta^{18}\text{O}_{\text{bw}}$  values more closely matched the measured values than the relationship between  $\delta^{18}\text{O}_{\text{bw}}$  and  $\delta^{18}\text{O}$  of precipitation developed by Longinelli [20] (Figure 1 and Table 4). The relationship produced by Longinelli was created using the estimated precipitation values and did not include the estimates for the influence of climate, diet, and metabolism. There were limited differences between the  $\delta^{18}\text{O}_{\text{bw}}$  values calculated with our version of the Kohn model and the original model published in 1996 since we used the Kohn model as our template (Tables 1, 2, and 4). There were slight differences between our version and the original version in the equations we used to predict evaporative water loss. However, these differences were not expected to, nor did they, overly change the predicted  $\delta^{18}\text{O}_{\text{bw}}$  values (Table 4). We added to the original Kohn model by also using the equations to predict  $\delta^2\text{H}_{\text{bw}}$  values since the H isotope ratio of organic tissues is also used in the study of human and animal ecology and palaeoecology. We imported the model into ArcGIS 9.1 ModelBuilder<sup>®</sup> to produce spatial predictions of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$ . These isoscapes of human body water can be used to produce spatial projections of the isotopic composition of hair, bones, and teeth.

The most positive  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values were found within interior Texas and along the Gulf Coast and the most negative values were found in the higher latitudes and elevations of the interior West (Figure 2). These projections closely matched the predicted  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of tap water [23]. Drinking water supplies >55% of the H atoms and >40% of the O atoms in the body water, and as a result, the isotope composition of the body water reflects the isotope composition of the tap water. However, the correlation between the isotope composition of drinking water and body water is not 1 due to the influence of climate through evaporation, the incorporation of

food H and O, and the incorporation of atmospheric O<sub>2</sub> into the body water. Atmospheric O<sub>2</sub> is responsible for >20% of the O atoms in the body water. The accurate spatial predictions of  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of body water require detailed process models that incorporate all influxes and effluxes of H and O.

We also evaluated parameterisations in which the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food were held constant. This would be equivalent to having all food originate from a single location. The modern diet of many countries is regional to global in nature; that is, the 'food footprint' is large and, as a result, much of the food consumed by individuals may not be of local origin [28,42]. If this is indeed the case, then parameterisations that do not link the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food to local water values may produce more reliable estimates of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$ , especially for the USA. There were only slight differences between the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values predicted in which the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food were linked to local values of drinking water and those in which the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values were held constant (Table 4). The differences were minimal and were still within the expected range of values for a location. These results suggest that while reliable estimates of the isotopic composition of food may be necessary for modelling the isotopic composition of proteinaceous tissues, this information may not be so critical for modelling body water. This is because the amount of H and O that is organically bound in food is relatively small when compared with the vast inputs of H and O from drinking water to the body water pool; thus, reliable estimates of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  can be produced without detailed estimates of the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of food.

We evaluated a last parameterisation in which the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values were estimated using the predicted precipitation values for the contiguous USA (V\_4) to determine how differences in the predicted drinking values influence the predicted body water values. There were large differences in the  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values depending on the drinking water source layer used. The  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values calculated with the predicted tap water values (V\_1) differed by >40 and >5.5‰ for H and O isotope ratios, respectively, from body water values calculated using the predicted precipitation values (Figure 3). These large differences were related to the differences between the estimated tap water  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values and the estimated precipitation values [23]. In areas where tap water was more enriched in the heavier isotopes than local precipitation, such as parts of Texas, the predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values were more positive, and likewise, in areas where local tap water was more depleted in the heavier isotopes than local precipitation, the predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values were more negative. The difference between V\_1 and V\_4 is not a constant value across the contiguous USA due to body water being influenced by differences in climate, the incorporation of food H and O, and the incorporation of atmospheric O<sub>2</sub> into the body water. This comparison illustrates that there can be large differences in the predicted  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values depending on the source used for drinking water values. In addition to the source of drinking water values, it may also be important to consider the seasonal and interannual variation that may occur in the isotope values of tap water [23], as a consequence of climatological factors and/or source switching (e.g. California). The impact of variation in water input on the correct geographical assignment of organisms has been extensively covered in studies of migratory birds [43–46], and this work warns of using static averages to generate predictions. More research into the spatial and temporal variability of tap water isotope values and food isotope values is needed to further characterise the impact this may have on the accuracy of human provenancing. As more information about the seasonal variation in  $\delta_{\text{dw}}$  and food becomes available, this model can be modified to include such data layers. The data used to test the four parameterisations of this model do have some limitations due to the limited numbers of measured  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values available and a lack of personal information about each participant. We used mean  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values for each location to control for individual differences in diet, drinking water sources, and differences in metabolic expenditure. As this model contains many variables such as metabolic expenditure and uses numerous equations to estimate specific

variables, issues with model uncertainties are large. For a truly robust model of  $\delta_{\text{bw}}$ , a sampling of individuals across a geographical gradient is required. Such a sampling scheme will allow future researchers to better calibrate the model and to assess the inherent uncertainties in this type of modelling exercise.

These data also do not allow us to evaluate if differences in the consumption of large amounts of bottled water, imported fruits, vegetables, and meats would greatly modify  $\delta_{\text{bw}}$  for individuals from the same locale. The consumption of bottled water may or may not cause differences between individuals from the same locale. Much of the bottled water sold in urban areas is bottled locally from municipal water sources and will have  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values close to those of local tap water [23]. The consumption of one or two bottles of imported water also will not significantly modify  $\delta_{\text{bw}}$  due to the large size of the body water pool. However, individuals who consume large amounts of imported waters and foods may have significantly different body water values compared with individuals who primarily consume tap water. These questions can be answered with the collection of a body water sample and a dietary survey from multiple individuals from the same local. Body water can be sampled through the collection of blood, urine, or breath. If differences in the amounts of imported foods and waters consumed are important for predicting equilibrium  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values at large spatial scales, this model can be modified to include such information.

#### 4.2. Applications of steady-state body water models

The isotopic compositions of hair, bones, and teeth are influenced by  $\delta_{\text{bw}}$  [9,20,47,48], and as a result, hair, bones, and teeth have been used to study the migration patterns of animals and humans [3–5]. The oxygen in tooth enamel and bone is directly related to the body water and researchers have used the  $\delta^{18}\text{O}$  of tooth enamel and bone to estimate the location of origin for modern and ancient samples and to reconstruct climate [20,47,49]. The identification of the region of origin and climate reconstruction will be enhanced with spatial projections of  $\delta^{18}\text{O}_{\text{bw}}$  values. For example, a spatial layer of the expected  $\delta^{18}\text{O}$  values of tooth enamel could be constructed based on reliable spatial estimates of  $\delta^{18}\text{O}$  values of drinking water and body water. It is important to note that spatial estimates addressing migration patterns in ecological and/or archaeological studies would likely benefit from the utilisation of body water modelling based on precipitation values, as these values may be more accurate estimates of drinking water for wildlife or ancient humans.

Recently, Ehleringer *et al.* [5] created spatial maps of the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of hair for the contiguous USA. They also proposed a multiple-pool body water model for predicting the  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values of hair based on local drinking water values. Efforts such as these will also be enhanced with reliable spatial layers of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values.

#### 4.3. Creation of a transient body water model

One application for spatial models of  $\delta_{\text{bw}}$  is to distinguish non-residents from residents in a population. One limitation to using tissues such as hair to reconstruct movement patterns is that hair will not identify changes in location during the most recent past, that is, less than one week. In contrast,  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values may identify travel or migration during the recent past if the individual travels from one isotopically distinct location to another [36]. In general, turnover of the body water occurs within 2–3 weeks. A body water sample collected from an individual soon after arrival may be used to identify a likely region of origin for a non-resident. Samples such as blood, urine, and breath  $\text{CO}_2$  can be collected and analysed to determine the  $\delta_{\text{bw}}$  pool. Blood water and urine are isotopically the same as the body water pool, and breath  $\text{CO}_2$  is in isotopic

equilibrium with the body water pool [22,37,50]. Combining estimates of turnover and variability with a reliable spatial map of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values will produce an estimate for the length of time for an individual to be isotopically indistinguishable from locals.

#### 4.4. Applications of a transient body water model

We created two scenarios to demonstrate the utility of the transient body water model. First, we determined the length of time for  $\delta^2\text{H}_{\text{bw}}$  values of a resident of Houston travelling to Chicago to be indistinguishable from those of a resident of Chicago. Second, we determined the length of time for  $\delta^2\text{H}_{\text{bw}}$  values of a resident of Houston travelling to Salt Lake City to be indistinguishable from those of a resident of Salt Lake City. We used parameterisation V\_1 for an adult male to estimate the equilibrium  $\delta^2\text{H}_{\text{bw}}$  values for Houston, Chicago, and Salt Lake City (Table 5). We combined these equilibrium values with the expected half-life of body water for young adult males [38] and with an estimate of the expected variability in  $\delta^2\text{H}_{\text{bw}}$  values for a local population [39] to predict the length of time required for a non-resident visiting the area to become indistinguishable from residents.

We estimated that a visitor from Houston to Chicago would become indistinguishable from residents within 14–18 days (Table 5). A visitor to Salt Lake City from Houston would become indistinguishable from a resident within 25–32 days (Table 5). The difference in the length of time for the two scenarios is a consequence of the magnitude of the difference in the  $\delta^2\text{H}_{\text{bw}}$  values among the three locations. The  $\delta^2\text{H}_{\text{bw}}$  value of a native of Chicago is 28‰ more negative than that of a native of Houston and the  $\delta^2\text{H}_{\text{bw}}$  value of a native of Salt Lake City is 90‰ more negative than that of a native of Houston (Table 5). As a result, the visitor to Chicago has within 4.5‰ of equilibrium values ~1 week before the visitor to Salt Lake City.

Overall, this exercise demonstrates the utility of combining spatial predictions of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  with the estimates for the half-life of body water to estimate the length of time necessary for an individual to equilibrate isotopically with the local diet and drinking water. The largest limitation to using  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values to identify visitors occurs if visitors travel within isotopically homogenous areas. Visitors who travel to isotopically distinct areas such as from Houston to Chicago or Salt Lake City are identifiable for limited periods of time. However, a visitor from Houston to Florida would be indistinguishable from a native because there are limited differences in steady-state  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  values (Figure 2). Overall, this exercise demonstrates the utility of combining spatial predictions of  $\delta^2\text{H}_{\text{bw}}$  and  $\delta^{18}\text{O}_{\text{bw}}$  with the estimates for the half-life of body water to estimate the length of time necessary for a visitor to equilibrate isotopically with the local diet and drinking water.

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