Assessing the Predictability of Existing Water-to-enamel Geolocation Models Against Known Human Teeth: Do We Need Tissue-Specific δ 18O Reference Maps?

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Title: Assessing the predictability of existing water-to-enamel geolocation models against known human teeth: Do we need tissue-specific $\delta^{18}\text{O}$ reference maps?

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Abstract

Stable isotope analysis of human tissues has become a valuable tool for mapping human geolocation. This study adds to the existing knowledge of the relationship between oxygen stable isotopes in human enamel ($\delta^{18}\text{O}_{\text{enam}}$) and drinking water ($\delta^{18}\text{O}_{\text{dw}}$) by presenting $\delta^{18}\text{O}_{\text{enam}}$ values in clinic-extracted human dental enamel with known provenance. The results from this study indicate that the theoretical isotopic relationship between $\delta^{18}\text{O}_{\text{enam}}$ and $\delta^{18}\text{O}_{\text{dw}}$ is valid although weak at the city and country-level. However, differences of up to 15 ‰ were observed between predicted $\delta^{18}\text{O}_{\text{dw}}$ values using existing models and observed values, highlighting the complexity of using enamel-to-water conversion equations. A city-specific range of $\delta^{18}\text{O}_{\text{enam}}$ values is now understood for Metro Vancouver [$\delta^{18}\text{O}_{\text{carbonate}} = -9.7\text{‰}$ to -7.2‰] and presents the possibility of both including within the city of Vancouver and excluding individuals, utilizing

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20 stable oxygen analysis as an exclusionary tool. Overall, this study's results support the
21 development of tissue-specific $^{18}\text{O}_{\text{enam}}$ geographical reference maps for human geolocation.
22

Introduction

Human tooth enamel stable oxygen isotope composition ($\delta^{18}\text{O}_{\text{enam}}$) has been used to localize region-of-origin of unknown human remains. $\delta^{18}\text{O}_{\text{enam}}$ ratios predominantly reflect stable oxygen isotope composition in drinking water ($\delta^{18}\text{O}_{\text{dw}}$) influenced by local precipitation water, which varies globally and can provide information on an individual's geographical history^{1,2}. Such data can help forensic investigations narrow down missing person searches by complementing other conventional identification methods such as fingerprint or DNA analysis. The current approach to human provenancing utilizes predictive models to estimate $\delta^{18}\text{O}_{\text{dw}}$ values from $\delta^{18}\text{O}_{\text{enam}}$ values. Individuals are subsequently placed on a geographical map, otherwise known as isoscapes.

Isoscapes for stable oxygen have been developed for precipitation and tap water, mainly at the country level^{1,3}, but also for more constrained geographical areas such as the state of Mississippi, U.S.⁴. There has also been an attempt to create an isoscape for $\delta^{18}\text{O}_{\text{enam}}$ values⁵. However, it is vital to recognize that isoscapes do not represent actual data spread but are instead extrapolated models, often based on limited measurements⁶. A suggested alternative empirical approach to human geolocation involves the direct mapping of the tissue isotopes themselves, thus eliminating any errors associated with the use of regression models⁷⁻⁹. However, mathematical models are important for predicting human origin in areas where either human or drinking water data do not exist. What is key to creating a reliable model for assisting with missing persons cases is to ensure that the baseline data and hence, $\delta^{18}\text{O}_{\text{dw}}$ to $\delta^{18}\text{O}_{\text{enam}}$ equations, accurately represent true relationships. Further, the predictability of such equations remains uncertain.

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Enamel is extremely resistant to post mortem alteration due to its high mineral composition (95%) in the form of calcium carbonate or phosphate hydroxyapatite ($\text{Ca}_{10}(\text{PO}_4\text{CO}_3)_6(\text{OH})_2$)^{10,11}. The stable oxygen isotope compositions in enamel hydroxyapatite can be determined by analyzing the more abundant phosphate or the structural carbonate substitute¹². $\delta^{18}\text{O}_{\text{enam}}$ values analyzed from both phosphate and structural carbonate are shown to have a direct positive relationship with $\delta^{18}\text{O}_{\text{ppt}}$ or $\delta^{18}\text{O}_{\text{dw}}$ values^{13–15}. Phosphate analysis has generally been the preferred analytical methodology for archaeological materials due to the high resistance to the environmental exchange of oxygen¹⁶. Conversely, structural carbonate is more susceptible to diagenetic alterations compared to phosphate but is more cost-effective and allows for robust analysis in terms of the amount of material required¹⁷. The susceptibility to diagenetic alterations is less of an issue with forensic samples due to the limited time in which enamel experiences environmental exposure. The relationship between carbonate and phosphate oxygen has been investigated for mammalian^{18,19} and human bioapatite^{20,21}.

Oxygen from body water is incorporated into enamel bioapatite at a constant temperature of 37 °C during tooth formation, preserving the isotopic record of body water oxygen within the enamel tissue. Three primary forms of oxygen input that influence the isotopic compositions of oxygen in body water ($\delta^{18}\text{O}_{\text{bw}}$) are drinking water ($\delta^{18}\text{O}_{\text{dw}}$), food water ($\delta^{18}\text{O}_{\text{fw}}$), and atmospheric oxygen ($\delta^{18}\text{O}_{\text{atm}}$). The main source of oxygen in body water has been shown to come from drinking water²². $\delta^{18}\text{O}_{\text{fw}}$ values also contribute to the overall $\delta^{18}\text{O}_{\text{bw}}$ values, although believed to be small for humans, unlike herbivores whose isotopic signature of oxygen

in the hydroxyapatite may be influenced by the relatively larger proportion of oxygen due to increased consumption of plants^{9,23}. Further, the contribution of $^{18}\text{O}_{\text{atm}}$ to $^{18}\text{O}_{\text{bw}}$ is approximately 20% and globally homogeneous at an average $\delta^{18}\text{O}_{\text{atm}}$ value of +23 ‰¹⁴. Oxygen isotopic compositions in human enamel are, therefore, strongly influenced by meteoric water.

The global geospatial isotopic pattern of meteoric water is dependent on local physiographic factors such as latitude, altitude, and the distance from the coast (Dansgaard, 1964; Rozanski, Araguás-Araguás, & Gonfiantini, 1993), with stable oxygen isotope values decreasing with increasing latitude (Bowen & Wilkinson, 2002). Global precipitation data is readily available through the Global Network of Isotopes in Precipitation, a monitoring network with collection stations located worldwide (IAEA/WMO, 2020). The Online Isotopes in Precipitation Calculator (OIPC) also provides a means to estimate $\delta^{18}\text{O}_{\text{ppt}}$ values for any geographical location from latitudinal, longitudinal, and elevational data (Bowen, 2018). Local precipitation water feeds into drinking water reservoirs, allowing $\delta^{18}\text{O}_{\text{ppt}}$ values to be good proxies for $\delta^{18}\text{O}_{\text{dw}}$ values.

While $^{18}\text{O}_{\text{ppt}}$ can influence $^{18}\text{O}_{\text{dw}}$ ratios, the degree to which one translates to another in modern society is dependent on several factors, such as the type of drinking water sources, climatic characteristics, and water infrastructure^{24,25}. There is a heightened reliance on complex water distribution systems for safe drinking water delivery and is notably essential for high density and expanding metropolitan regions^{26,27}. Tap water distribution systems often draw water from multiple water sources or geographically distant areas with varying physiography²⁸ to meet growing communities' demands. This ultimately leads to significant offsets in tap water oxygen

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($\delta^{18}\text{O}_{\text{tap}}$) values compared to that of local $\delta^{18}\text{O}_{\text{ppt}}$ values. It is becoming crucial to understand the source and distribution of drinking water to accurately determine the fractional contribution of $^{18}\text{O}_{\text{dw}}$ to $^{18}\text{O}_{\text{enam}}$, particularly with the modern population increasingly relying on piped water systems. It is especially vital because of the heavy influence of $^{18}\text{O}_{\text{dw}}$ on $^{18}\text{O}_{\text{bw}}$ and subsequently on $^{18}\text{O}_{\text{enam}}$ ratios.

There is an increased understanding of the isotopic patterns of tap water across large geographical areas such as the conterminous United States^{29,30}, Mexico³¹, South Africa³², and China³³, as well as for defined metropolitan areas where studies have shown that isotopic values closely follow its distribution systems^{34,35}. Ueda and Bell³⁵ showed that $\delta^{18}\text{O}_{\text{tap}}$ values of Metro Vancouver (MV) uniquely follow the three primary water sources' distribution patterns—Capilano, Seymour, and Coquitlam watersheds. The regional district includes 21 municipalities, one electoral area, and one treaty of First Nations. Tap water is delivered to each municipality through a modern water distribution system closely monitored and operated by the Metro Vancouver Water Services³⁶. This highly detailed understanding of the source and distribution of tap waters across MV allows for a precise evaluation of the relationship between $\delta^{18}\text{O}_{\text{enam}}$ and $\delta^{18}\text{O}_{\text{dw}}$ values through the careful collection and the analysis of both modern tap water and enamel samples with known geographical information.

This study aims to validate the theoretical understanding of the drinking water-to-enamel relationship through a paired analysis of known modern $\delta^{18}\text{O}_{\text{c}}$ and $\delta^{18}\text{O}_{\text{dw}}$ values focusing on a single regional district of Metro Vancouver, British Columbia, Canada. Additionally, results

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from this study will be compared against existing predictive models to determine how well they perform against known modern samples.

Methods

Ethics Permission

This research project has been approved by the Associate Director, Simon Fraser University Office of Research Ethics, on behalf of the Research Ethics Board under the study number [2015s0125]. Informed consent was obtained from all subjects, and all experiments were performed in accordance with University Policy R20.01 (<http://www.sfu.ca/policies/gazette/research/r20-01.html>).

Collection and analysis of tooth samples

154 empty vials were sent out to dental clinics across Metro Vancouver during the years 2015 to 2017. 126 vials were returned with extracted human molar samples comprised of first, second, and third molars. Molars were placed in 20 mL scintillation vials labelled with sample identifiers following the extraction and returned to the laboratory for storage. Immense care was taken to obtain information from individuals on residence location and any relocations from birth to 25 years to ensure that known geographical information was acquired directly from individuals for the entire duration of tissue formation (**Table 1**). This information was vital for ensuring that $^{18}\text{O}_{\text{enam}}$ values were matched with appropriate $^{18}\text{O}_{\text{tap}}$ values from the location of tissue formation.

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Collected molars were cleansed with tap water and dried for at least 24 hours before drilling. Core enamel was extracted into powdered form (~1 mg) by gentle abrasion with a diamond-tipped hand-held drill (Dremel). The enamel's external surface was mechanically removed and discarded prior to sampling of core enamel to ensure sampled areas were devoid of dental calculus and overt caries. Care was taken to ensure no dentine was sampled, and the drill was cleaned of any enamel powder with compressed air between each sample. Enamel powder was prepared after Lee-Thorp et al.³⁷. The powder was soaked in 50 % sodium hypochlorite for 45 to 60 minutes, centrifuged, and rinsed with distilled water. The powder was then reacted with 0.1 M acetic acid for 5 to 15 minutes, thoroughly rinsed with distilled water then freeze-dried.

Samples were weighed into 12 ml borosilicate tubes (Labco, Ceredigion, U.K.) and closed with screw top lids containing a septum. The tubes were placed in a sampler tray temperature-controlled to 72 °C. Using the CTC Analytics A200S autosampler (Switzerland) the tubes were flushed with helium to remove any atmospheric air present within them. 5 to 7 drops (according to sample size) of warm (72 °C) acid (85 % orthophosphoric acid and phosphorus pentoxide, specific gravity of solution = 1.92) were then manually added to each sample tube through the septum using a 1 ml syringe. The samples were left to react for three hours before running the analysis. The gas evolved from each reaction was sampled by the autosampler and passed to a Thermo Finnigan (Germany) model II gasbench, where the sample gas was passed through a Nafion water removal unit. It then passed through the "Poraplot Q" G.C. column to separate the gas compounds released by the reaction, and then through a second Nafion water trap. The gas

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was then passed from the gasbench to a Delta Plus XP isotope ratio mass spectrometer (IRMS) (Thermo electron, Bremen, Germany) computer-controlled by Isodat software. The gasflow was controlled to give 8 sample peaks and 5 reference peaks. The CO₂ reference gas (99.995 % purity) was introduced into the mass spectrometer via the gasbench, which was also controlled by the Isodat software.

Oxygen analyses were carried out on the structural carbonate of tooth enamel. Isotopic results are reported in conventional ‰ - notation in units of per mil (‰) with reference to Vienna Pee Dee Belemnite (VPDB) carbonate and calculated according to the equation:

$$\delta^{18}O_{std} = \frac{R(^{18}O/^{16}O)_P}{R(^{18}O/^{16}O)_{std}} - 1$$

where R = ratio of the abundance of heavier ¹⁸O to lighter ¹⁶O isotopes in substance P and std = standards³⁸. Analytical precision was ± 0.2 ‰.

Collection and analysis of annual MV $\delta^{18}O_{tap}$ values

Two treated tap water samples were collected monthly at both the Seymour-Capilano Filtration Plant (SCFP) and the Coquitlam Water Treatment Plant (CWTP) from June 2017 to May 2018. Samples were collected in air-tight 8-dram borosilicate glass vials and stored in a refrigerator to prevent evaporation. 3 of the 48 collected tap water samples were discarded due to leakage during transport, leaving only single measurements for June 2017 and May 2018 at CWTP and

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for October 2017 at SCFP, for a total of 45 tap water samples available for analysis. The water samples were analyzed for oxygen through the Los Gatos Triple Liquid Water Isotope Analyzer (TLWIA-45-EP 2013) connected to CTC Analytics LEAP Technology PAL liquid auto-sampler and checked against Los Gatos Standards 1A, 2A, 3A, 4A and 5A. Instrumental precision was $\pm 0.1 \text{ ‰}$.

MV tap water is delivered to municipalities through interconnected water pipes originating from the three main MV watersheds. Several municipalities additionally draw tap water from wells³⁹. Although the water distribution system is well monitored, the source of tap water delivered to the municipalities through the interconnected water pipes can vary even by the hour. As a result, the exact fractional contributions of end-member water sources to the overall municipal tap water supply are unknown. The fractional contribution (f) of each water source (a, b, c, d) to the total water supply can be expressed as:

$$1 = f_{(a)} + f_{(b)} + f_{(c)} + f_{(d)}$$

Source information of tap water samples collected and reported in Ueda and Bell's³⁵ study was used to determine each water source's potential fractional contribution to MV municipality water supply. The fractional contributions of each water source were then utilized to calculate the mean $\delta^{18}\text{O}_{\text{tap}}$ values for each MV municipality:

$$\delta^{18}\text{O}_{(\text{mean})} = \delta^{18}\text{O}_{(a)} * f_{(a)} + \delta^{18}\text{O}_{(b)} * f_{(b)} + \delta^{18}\text{O}_{(c)} * f_{(c)} + \delta^{18}\text{O}_{(d)} * f_{(d)}$$

Drinking water from Capilano and Seymour watersheds was treated under two separate facilities until the Seymour-Capilano Twin Tunnel construction in 2015, which now connects water from

the Capilano watershed to the Seymour-Capilano Filtration Plan before distribution to the municipalities³⁹.

Data collection of $\delta^{18}\text{O}_{\text{dw}}$ values for individuals placed outside of MV

Isotopic data on $\delta^{18}\text{O}_{\text{dw}}$ values for cities outside of MV were retrieved from existing publications for comparisons with $\delta^{18}\text{O}_{\text{c}}$ values. The preferred drinking water type was tap water with known source water information. Tap water data without any indications of the actual source of tap water were also collected but with the understanding that it may represent mixed water sources if supplied by multiple water sources. The temporal representation of the data, whether annual or a one-time collection, was also noted. Isotopic data of actual or possible source water were taken as an alternative when tap water data did not exist. Average values were calculated if more than one $\delta^{18}\text{O}_{\text{tap}}$ value was reported for a given city. Information on tap water sources was collected through municipal or governmental websites and from published studies. The OIPC⁴⁰ and the Regionalized Cluster-based Water Isotope Prediction (RCWIP) database^{41–43} were utilized to estimate local $\delta^{18}\text{O}_{\text{OIPC}}$ and $\delta^{18}\text{O}_{\text{RCWIP}}$ values, respectively.

Statistical analysis

The pairwise t-test was used to test for differences between estimated $\delta^{18}\text{O}_{\text{ppt}}$ values retrieved from the two databases at the $\alpha=0.05$ level. The ordinary least squares (OLS) method was utilized to measure the correlation between $\delta^{18}\text{O}_{\text{c}}$ and $\delta^{18}\text{O}_{\text{dw}}$ values with adjusted r-squared ($\bar{R}^2$) as the goodness-of-fit measure⁴⁴. Coplen's⁴⁵ equation was utilized for any conversions from VPDB to Vienna Standard Mean Ocean Water (VSMOW). One-way analysis of variance

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(ANOVA) tests were performed to compare means of $\delta^{18}\text{O}_c$ between cities, water sources, and countries. When ANOVA assumptions were not met, the Kruskal-Wallis rank sum test⁴⁶ was performed as a non-parametric alternative to one-way ANOVA. Shapiro-Wilk's⁴⁷ method was used to test for normality of data. All mapping and statistical analyses were conducted on R version 3.2.3⁴⁸.

$\delta^{18}\text{O}_c$ to $\delta^{18}\text{O}_{dw}$ modelling and assessment of predictability

$\delta^{18}\text{O}_{c(\text{VSMOW})}$ values were converted to $\delta^{18}\text{O}_{dw}$ values when predictive models allowed for direct conversions. $\delta^{18}\text{O}_c$ to $\delta^{18}\text{O}_p$ conversion equations (Table 2) were utilized when conversions to $\delta^{18}\text{O}_p$ values were necessary for comparison purposes. Predicted $\delta^{18}\text{O}_{dw}$ values were regressed against actual $\delta^{18}\text{O}_{\text{tap}}$ values and $\delta^{18}\text{O}_{\text{OIPC}}$ values separately to evaluate the performance of each model or combination of models in cases where multiple conversions were needed. Predicted values were deliberately placed on the x-axis and observed values on the y-axis to ensure correct correlation estimates⁴⁹. The regression analyses were complemented by the reporting of the mean squared deviation (MSD)^{50,51} calculated as

$$\text{MSD} = \frac{1}{n} \sum_{k=1}^n (x_k - y_k)^2$$

which measures the degree of deviation from the equality line, where slope = 1 and intercept = 0. MSD was preferred over the more commonly used statistic of mean squared error that measures the mean deviation from the regression line rather than the equality line. MSD can further be partitioned into three separate components and is the sum of squared bias (SB),

$$\text{SB} = (\bar{x} - \bar{y})^2$$

non-unity slope (NU) where $b = \sum x_n y_n / \sum x_n^2$,

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$$NU = (1-b)^2 * (\sum x_n^2 * \frac{1}{n})$$

and lack of correlation (LC) where $r = (\sum x_n y_n) / \sqrt{\sum x_n^2 \sum y_n^2}$,

$$LC = (1 - r^2) * (\sum y_n^2 * \frac{1}{n})$$

The models or sets of models were then ranked according to MSD to determine the best performing models for predicting $\delta^{18}O_{\text{tap}}$ or $\delta^{18}O_{\text{OIPC}}$ values from observed $\delta^{18}O_c$ values.

Results

The collection of enamel samples from dental clinics across MV resulted in a globally represented dataset including 14 countries (Figure 1) reflecting the regional district's high fluidity in international migrations⁵². Of the collected tooth samples, enough enamel powder was generated from 103 samples for isotopic analysis. Two samples returned no oxygen isotopic signatures signifying the absence of organic matter within the enamel, which could be attributed to the presence of dental porcelain that is visually akin to dental enamel. City-level geographical information could not be retrieved from 21 of the analyzed samples due to either missing information or where individuals provided only the name of the country of residence. However, one sample came from the Fiji Islands. While the exact city was not identified, it was included in the study because of the country's relatively small size and as the only sample from Oceania. Lastly, five samples came from individuals who had relocated to multiple locations during the span of tissue formation and thus could not be associated with any single city, resulting in a total of 75 samples with reliable geographical associations.

A summary of $\delta^{18}\text{O}_c$ results for all analyzed samples is provided as Supplementary Table S1 online along with the corresponding mean $\delta^{18}\text{O}_{\text{dw}}$ values taken from other published studies for the city of residence, $\square^{18}\text{O}_{\text{OIPC}}$ and $\square^{18}\text{O}_{\text{RCWIP}}$ values. The estimated $\square^{18}\text{O}_{\text{OIPC}}$ and $\square^{18}\text{O}_{\text{RCWIP}}$ values were not statistically significantly different as per the pairwise t-test [$t(74) = -0.747$, $\text{df}=74$, $p=0.457$], and thus only the estimated values from the OIPC were used for subsequent analyses.

$\delta^{18}\text{O}_c$ and $\delta^{18}\text{O}_{\text{tap}}$ results for MV

24 individuals were of MV origin and covered eight municipalities—Burnaby, Coquitlam, Langley, Richmond, New Westminster, North Vancouver, Surrey, and Vancouver (Figure 2). $\delta^{18}\text{O}_{c(\text{VPDB})}$ values ranged from -11.0‰ (Burnaby) to -7.2‰ (Surrey) with a mean value of $-8.7\text{‰} \pm 0.8$ (Figure 3a). One-way ANOVA test showed no statistically significant inter-city differences between mean $\delta^{18}\text{O}_{c(\text{VPDB})}$ values [$F(7, 16) = 0.717$, $p = 0.659$]. Further, no statistically significant differences were observed for mean $\delta^{18}\text{O}_{c(\text{VPDB})}$ values when data was disaggregated by tap water source [$F(4,19) = 1.013$, $p = 0.426$] (Figure 3b); however, an outlier was identified for the sample with $\delta^{18}\text{O}_{c(\text{VPDB})}$ value of -11.0‰ . The MV $\delta^{18}\text{O}_{c(\text{VPDB})}$ range then becomes -9.7‰ to -7.2‰ when the outlier is excluded.

Monthly $\delta^{18}\text{O}_{\text{tap}}$ values ranged from -13.0‰ to -8.2‰ with an annual mean of -11.1‰ ($n=22$) and -13.2‰ to -8.5‰ with an annual mean of -11.7‰ ($n=23$) for MV tap waters treated at CWTP and SCFP, respectively (Figure 4). Fractional contributions of each major MV tap water

source to the overall municipal water supply were calculated for all municipalities represented by the enamel samples (Table 3).

The OLS regression for MV $\delta^{18}\text{O}_c$ and $\delta^{18}\text{O}_{\text{tap}}$ values yielded the result:

$$\delta^{18}\text{O}_{c(\text{VSMOW})} = 0.10 (\pm 0.75) \times \delta^{18}\text{O}_{\text{tap}(\text{VSMOW})} + 23.08 (\pm 8.58); [\text{df} = 22; \bar{R}^2 = -0.04] \quad (1)$$

The negative $\bar{R}^2$ value indicates that, statistically, the data cannot be explained by the model.

$\delta^{18}\text{O}_c$ results for Canada

In addition to the 24 individuals from MV, four individuals were from BC cities outside of MV, two from Manitoba (MB), seven from Ontario (ON), and two from Quebec (QC) (Figure 5). The mean $\delta^{18}\text{O}_{c(\text{VPDB})}$ value for BC was similar to MV at $-8.7\text{‰} \pm 0.8$ ($n = 29$). The mean $\delta^{18}\text{O}_{c(\text{VPDB})}$ value for all Canadian samples was also similar at $-8.7\text{‰} \pm 1.1$ ($n = 41$) with a range of -11.0‰ (Burnaby, BC) to -5.3‰ (Hamilton, ON). One-way ANOVA test showed no statistically significant differences between provinces [$F(3,35) = 0.825$, $p = 0.489$]. Cities supplied by Lake Ontario showed the largest range in $\delta^{18}\text{O}_{c(\text{VPDB})}$ values from -10.6‰ to -5.3‰ compared to other water sources from across Canada.

Source water information was retrieved for each represented city^{39,53–58}. No source water information could officially be retrieved for Chapeau, QC, but the most probable water source was identified to be Ottawa River⁵⁹. Modern $\delta^{18}\text{O}_{\text{tap}}$ data were found for Hamilton, St. Catharines, Toronto⁶⁰, and Winnipeg⁶¹ with given source information (see Supplementary Table S2 online). $\delta^{18}\text{O}_{\text{tap}}$ data for Ottawa was taken for both Chapeau and Morin Heights as no

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$^{18}\text{O}_{\text{tap}}$ data were found for the two cities. Ottawa River supplies both Chapeau and Morin Heights, and which also is the water source for Ottawa. Only surface water values were found for the drinking water sources of Kelowna⁶², Prince George, and Vanderhoof⁶³, and no $\square$ $^{18}\text{O}_{\text{dw}}$ values were associated with Williams Lake and Baysville.

$\delta^{18}\text{O}_{\text{c(VSMOW)}}$ values were regressed against three sets of $\delta^{18}\text{O}_{\text{dw}}$ values—(1) tap water values, (2) tap water and surface water or groundwater values ($\delta^{18}\text{O}_{\text{dw}_1}$), and (3) a combined set of tap water, surface water or groundwater and OIPC values ($\delta^{18}\text{O}_{\text{dw}_2}$) (Figure 6) because not all samples could be directly paired with modern $\square$ $^{18}\text{O}_{\text{tap}}$ values.

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.11 (\pm 0.09) \times \delta^{18}\text{O}_{\text{tap(VSMOW)}} + 23.29 (\pm 0.98); [\text{df} = 32; \bar{R}^2 = 0.01] \quad (2)$$

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.16 (\pm 0.08) \times \delta^{18}\text{O}_{\text{dw}_1(\text{VSMOW})} + 23.77 (\pm 0.82); [\text{df} = 35; \bar{R}^2 = 0.09] \quad (3)$$

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.15 (\pm 0.07) \times \delta^{18}\text{O}_{\text{dw}_2(\text{VSMOW})} + 23.65 (\pm 0.78); [\text{df} = 37; \bar{R}^2 = 0.08] \quad (4)$$

The Analysis of Covariance (ANCOVA) test was used to compare the three sets of linear regressions to determine whether they significantly differ from each other. Results showed no statistically significant differences between the three equations, $[F(2, 106)=0.010, p=0.990]$.

Equation (3) with the highest $\bar{R}^2$ value proved to be the most reliable equation for Canada.

Overall, all three regressions showed a positive but weak linear relationship between $\square$ $^{18}\text{O}_{\text{enam}}$ and $\square$ $^{18}\text{O}_{\text{dw}}$ values for the Canadian samples.

$\delta^{18}\text{O}_{\text{c}}$ results for the global dataset

The remaining 36 samples included individuals from 13 additional countries encompassing the latitudinal areas from 55.75124 (Moscow, Russia) to -17.71337 (Fiji Islands). The mean δ

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$^{18}\text{O}_{\text{c(VPDB)}}$ values were more negative in comparison to Canada for all countries except for the single enamel formed in Russia (Figure 7). There was a general latitudinal trend where $\delta^{18}\text{O}_{\text{c}}$ values increased with decreasing latitude, although with a large range at higher latitudes (Figure 8).

Tap water data were retrieved for China^{25,33,64}, Hong Kong^{33,64}, Iran, Kazakhstan, Kenya⁶⁴, Malaysia⁶⁵, Mexico, Russia, and Vietnam⁶⁴ (see Supplementary Table 2 online). While tap water data was available for New Delhi, India^{64,66}, only groundwater values could be retrieved for Kolkata, India⁶⁴. No tap water value was found for Kitwe, Zambia; however, surface water data of Kafue River, which is the drinking water source for Kitwe⁶⁷, was retrieved⁶⁸. No such data were found for Fiji, Japan, and for Yiyuan, Zibo in China.

The OLS regression for $\delta^{18}\text{O}_{\text{c}}$ and $\delta^{18}\text{O}_{\text{tap}}$ for the global dataset yielded the result (Figure 9):

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.41 (\pm 0.06) \times \delta^{18}\text{O}_{\text{tap(VSMOW)}} + 26.55 (\pm 0.55); [\text{df}=63, \bar{R}^2 = 0.41] \quad (5)$$

Similar to the Canadian dataset, $\delta^{18}\text{O}_{\text{c(VSMOW)}}$ values were regressed against $\delta^{18}\text{O}_{\text{dw}_1(\text{VSMOW})}$, and $\delta^{18}\text{O}_{\text{dw}_2(\text{VSMOW})}$ values, as well as against $\delta^{18}\text{O}_{\text{OIPC(VSMOW)}}$.

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.39 (\pm 0.05) \times \delta^{18}\text{O}_{\text{dw}_1(\text{VSMOW)}} + 26.41 (\pm 0.47); [\text{df}=68, \bar{R}^2 = 0.45] \quad (6)$$

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.40 (\pm 0.05) \times \delta^{18}\text{O}_{\text{dw}_2(\text{VSMOW)}} + 26.53 (\pm 0.45); [\text{df}=73, \bar{R}^2 = 0.47] \quad (7)$$

$$\delta^{18}\text{O}_{\text{c(VSMOW)}} = 0.43 (\pm 0.05) \times \delta^{18}\text{O}_{\text{OIPC(VSMOW)}} + 26.92 (\pm 0.46); [\text{df}=73, \bar{R}^2 = 0.51] \quad (8)$$

The ANCOVA test showed no statistically significant differences between the four equations [F(3, 280)=0.144, p=0.933]. Of the four equations, equation (8) had the highest $\bar{R}^2$ value and thus proved to be the most reliable equation for the global dataset.

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Predictability of existing models

The observed MV $\delta^{18}\text{O}_{\text{tap}}$ values were used to estimate the range of MV $\delta^{18}\text{O}_{\text{c}}$ values through existing predictive models and all equations derived in this study. Fars, Iran (MV007) and Khuzestan, Iran (MV097) were identified to be from hot arid climates as specified by the updated Koppen climate classification system⁶⁹. They were converted to $\delta^{18}\text{O}_{\text{p}}$ values using the equation given by Chenery et al.²⁰ [$\delta^{18}\text{O}_{\text{p}} = 1.122 \delta^{18}\text{O}_{\text{c}} - 13.73$]. The minimum MV tap water value of -11.7‰ was taken from annualized data of SCWT, and the maximum value of -9.3 was reported in Ueda and Bell³⁵ with a groundwater source. The range of values estimated by Dotsika's⁷⁰ equation provided the most accurate estimate where 91.7% of MV individuals were correctly identified to have resided in MV during their childhood years. 15 combinations of models failed to identify any MV individuals accurately. The observed ranges of $\delta^{18}\text{O}_{\text{c}}$ values for MV were generally more negative than predicted ranges from existing models except for ranges predicted with Posey's⁷¹ equation (Figure 10).

The prediction of $\square^{18}\text{O}_{\text{dw}}$ values from $\square^{18}\text{O}_{\text{enam}}$ values with existing models or a combination of existing models (see Supplementary Table S3 online) resulted in a wide range of $\square^{18}\text{O}_{\text{dw}}$ values for each observed $\square^{18}\text{O}_{\text{c}}$ measurement (Figure 11). The offset between predicted and observed values ranged from -15.3 to $+5.5\text{‰}$ for tap water (Figure 12a) and -12.5 to $+6.5\text{‰}$ for OIPC (Figure 12b). The use of the equation given by Dotsika⁷⁰ produced the least MSD between predicted and observed drinking water values for both tap water measurements (MSD=4.00; NU=0.00; LC=3.45; SB=0.55) (Figure 13a) and OIPC measurements (MSD=3.72; NU=0.07;

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LC=3.42; SB=0.23) (Figure 13b; see Supplementary Table S4 online). Equation (1) gave the highest MSD for both tap water (MSD=278.21; NU=210.61; LC=3.45; SB=64.15) and OIPC measurements (MSD=297.08; NU=223.42; LC=3.41; SB=70.24).

Discussion

Results from MV enamel samples revealed no statistically significant differences in mean $\delta^{18}\text{O}_c$ values across the regional district regardless of the municipal tap water source differences. This may be explained by the similarity in the annualized $\delta^{18}\text{O}_{\text{tap}}$ values of tap water treated at the two main MV tap water treatment facilities—SCTF and CWTP—that treats approximately 95% of all tap water delivered to MV municipalities³⁹. The sampled enamels were formed during the period when an independent treatment facility had treated Capilano water. However, Capilano $\delta^{18}\text{O}_{\text{tap}}$ values had shown significant overlap with Seymour $\delta^{18}\text{O}_{\text{tap}}$ values before the construction of the twin tunnels³⁵. The isotopically homogeneous results across MV municipalities indicate that human geolocation, based solely on $\delta^{18}\text{O}_c$ values, is difficult at the city-level for MV but that a defined range of values exists for the entire regional district, which is now known to span from -9.7‰ to -7.2‰ . Notably, the understanding of such a defined range in values for MV signifies the possibility of oxygen isotope analysis to be used as an exclusionary tool to identify non-MV residents who fall outside of this defined range.

The range of annualized $\delta^{18}\text{O}_{\text{tap}}$ values for MV (-11.7‰ to -9.3‰) includes groundwater values presented in Ueda and Bell's³⁵ study that are assumed to represent annualized water although from a single sample. Given that the fractional contribution of $^{18}\text{O}_{\text{dw}}$ to $^{18}\text{O}_{\text{enam}}$ is <1 ,

MV $\delta^{18}\text{O}_\text{c}$ values can be expected to have a smaller range than that of $\delta^{18}\text{O}_\text{dw}$ values. However, the exact estimation of this range is dependent on which combination of existing predictive models are applied. The predicted MV $\delta^{18}\text{O}_\text{enam}$ values generally were positively biased except for the values predicted from Dotsika's⁷⁰ equation which showed very good predictability. Dotsika's equation was developed based on a country-level analysis of modern human samples from Greece with known $\delta^{18}\text{O}_\text{dw}$ values.

Sampling across MV enabled the collection of a globally represented set of samples due to the regional district's high fluidity in both national and international migration⁵². Notably, $\delta^{18}\text{O}_\text{c}$ values in known modern samples from the entire MV region showed a distinct separation from other B.C. cities (Prince George and Vanderhoof) located at higher latitudes and signifies the possibility of utilizing oxygen isotope analysis to distinguish MV from northern B.C. cities. However, overlaps in $\delta^{18}\text{O}_\text{c}$ values were observed between other Canadian cities regardless of the latitudinal differences (Figure 6). A general latitudinal trend can, however, be observed when looking at the global enamel dataset. Mean Canadian $\delta^{18}\text{O}_\text{c}$ values were slightly more negative compared to $\delta^{18}\text{O}_\text{c}$ values from individuals residing in other parts of the world during the time of enamel formation, although with increasing overlap for the more positive $\delta^{18}\text{O}_\text{c}$ values. This again underscores this oxygen-isotope tool's potential to be used as an exclusionary method for identifying individuals with a non-Canadian origin when $\delta^{18}\text{O}_\text{c}$ values fall outside the Canadian range.

Equation (8) showed the highest $\bar{R}^2$, where $\delta^{18}\text{O}_{\text{dw}}$ values were calculated with the OIPC for the global dataset. In comparison, equation (1) had the lowest $\bar{R}^2$ but with the highest geographical resolution. The model performed better as the geographical resolution decreased from city to country to a global dataset. This indicates that a positive relationship between $^{18}\text{O}_{\text{enam}}$ and $^{18}\text{O}_{\text{dw}}$ exists at the global level, with precipitation most heavily influencing $\delta^{18}\text{O}_{\text{c}}$ values. Perhaps such an observation can be explained by the influence of precipitation on the overall drinking water systems. The lower $\bar{R}^2$ from MV and Canadian samples may also be due to the complexity in determining the fractional contributions of each water source to consumed water. The interconnectedness of MV water pipes reduces community dependence on a single water source. It enables water distribution from alternative sources during water shortage or contamination, which can change by the hour.

If it is true that a single line can represent the $^{18}\text{O}_{\text{c}}$ and $^{18}\text{O}_{\text{dw}}$ relationship, then the degree to which one relates to the other should be independent of geographical resolution. Globally constructed predictive models should then be applied with caution and with the understanding that there is a possibility of individuals deviating from the predicted regression line, especially when attempting to geolocate individuals on a smaller geographical scale. Additionally, the $^{18}\text{O}_{\text{c}}$ and $^{18}\text{O}_{\text{dw}}$ relationship simplification can incorrectly lead to the assumption that regional areas with a common $\delta^{18}\text{O}_{\text{ppt}}$ or $\delta^{18}\text{O}_{\text{tap}}$ value translates to a single $\delta^{18}\text{O}_{\text{c}}$ value. Like MV, enamel samples formed in Hong Kong showed a wide range of $\delta^{18}\text{O}_{\text{c}}$ values, which only became evident with the concentrated number of individuals within our sample set that had resided in

Hong Kong during the time of tissue formation. Other cities may show such a spread in $\delta^{18}\text{O}_c$ values, which can only be understood with a high-resolution study.

Overall, the equations derived from this study reveal a smaller fractional contribution of $^{18}\text{O}_{\text{dw}}$ to $^{18}\text{O}_{\text{enam}}$ compared to existing water to enamel models. The equations from the global dataset (equations 5 to 8) were more comparable to other predictive models than those derived from higher geographical resolutions (equations 1 to 4). Equation (8) with the highest $\bar{R}^2$ was most comparable to Levinson et al's¹³ equation.

Dotsika's⁷⁰ equation proved to best predict $\delta^{18}\text{O}_{\text{dw}}$ from $\delta^{18}\text{O}_c$ values with the lowest mean standard deviation of 4.00 for tap water and 3.72 for OIPC. Again, this could be attributed to the care taken in the sampling of known data from across Greece with the pairing of known drinking water data. The equations that showed the least accuracies were those from this current study (equations 1 to 4) with large NU components. NU is a direct measure of the slope where $\text{NU} > 0$ when the slope $\neq 1$ ⁵¹, it may be deduced to reflect observed differences in the measured fractional contributions of $^{18}\text{O}_{\text{dw}}$ to $^{18}\text{O}_c$. Importantly, however, is to note that individuals from a constrained geographical region can show wide ranges in $\delta^{18}\text{O}_c$ values, which may lead to low correlation measures with drinking water samples, such as was the case for MV. This range of $\delta^{18}\text{O}_c$ values specific to its region can only be understood through concentrated sampling and analysis of a large number of individuals from that same region. Therefore, large NU measures do not necessarily translate to differences in measures of $^{18}\text{O}_{\text{dw}}$ to $^{18}\text{O}_c$ fractional contributions

but rather that the true range of $\delta^{18}\text{O}_c$ values became known and that perhaps it is not possible to measure the direct relationships simply by linear regression.

Daux et al.'s⁹ equation paired with Iacumin et al.'s¹⁹ equation (D5I) showed the largest MSD amongst the existing predictive models and scored high on SB (Figure 13). $\text{SB} > 0$ when intercept $\neq 0$ and thus reflects the vertical shifting of a function. High SB measures could be a reflection of inter-laboratory differences. Inter-laboratory differences between $\delta^{18}\text{O}_p$ values, as discussed by Chenery et al.⁷² are generally related to bio-phosphate preparation with either BiPO_4 or Ag_3PO_4 prior to oxygen extraction. Chenery et al. concluded that a correction of 1.4 to Levinson et al.'s¹³ equation is required to best predict local $\delta^{18}\text{O}_{\text{dw}}$ values as measured through the U.K. drinking water isoscapes. However, MSD calculations for this study showed contrary results to Levinson et al.'s original equation, which performed better for known modern samples measured for carbonate oxygen than that of Chenery et al.'s corrected Levinson's equation. SB measures were generally lower for equations derived from carbonate oxygen analysis and those derived from phosphate oxygen prepared with BiPO_4 . In contrast, equations derived from phosphate oxygen prepared with Ag_3PO_4 had larger SB measures, as were the case for Daux et al.'s equations.

Differences in the predicted and observed $\delta^{18}\text{O}_{\text{dw}}$ values were as high as 15 ‰, where differences increased with decreasing $\delta^{18}\text{O}_c$ values (Figure 12). Whereas the differences between predicted and observed $\delta^{18}\text{O}_{\text{dw}}$ values generally fell within ± 5 ‰ for $\delta^{18}\text{O}_c$ values of 25 to 28 ‰, greater differences were observed for lower $\delta^{18}\text{O}_c$ values, with the highest difference observed for a sample with $\delta^{18}\text{O}_c$ value of -20 ‰. Inter-laboratory differences between $\delta^{18}\text{O}_c$ values measured

at three separate laboratories using different methodologies and instruments were <1‰ for carbonate in tooth enamel¹⁷. Chesson et al.⁷³ further tested for interlaboratory differences of enamel oxygen from the same set of samples analyzed a few years apart. The differences were mainly attributed to sample preparation and concluded that isotopic differences >1.6 ‰ should be considered meaningful. The potential bias associated with inter-laboratory differences in sample preparation, methodology, and the choice of instruments, will further be compounded in the calculations of $\delta^{18}\text{O}_{\text{dw}}$ values using $\delta^{18}\text{O}_{\text{c}}$ to $\delta^{18}\text{O}_{\text{dw}}$ conversion equations. The compounding factor is dependent on the slope of the applied $\delta^{18}\text{O}_{\text{c}}$ to $\delta^{18}\text{O}_{\text{dw}}$ equation. For example, Ehleringer's⁵ equation with a slope of 1.12 would translate to a maximum of 1.8 ‰ difference in $\delta^{18}\text{O}_{\text{dw}}$ values that may be associated with inter-laboratory differences. Ehleringer's equation showed high offsets between observed $\delta^{18}\text{O}_{\text{dw}}$ values and predicted $\delta^{18}\text{O}_{\text{dw}}$ values for individuals with relatively more negative $\delta^{18}\text{O}_{\text{c}}$ values. Such large differences beyond what can be explained by the possible inter-laboratory differences can be concluded to be important.

Differences in slope and intercept of existing equations may be attributed to various methodological approaches, such as in the types of sampled tissues in which the equations were based upon, the period of tissue formation, the spatial resolution of collection sites^{5,7,9,13–15,72} (Table 4). For example, one of the early models given by Longinelli¹⁵ was based on measurements of human bone bioapatite rather than on human enamel. Some studies lacked information on the specific types of sampled tooth^{13,14} while others studied archaeological or forensic samples with inferred geographical information where drinking water sources may be unknown. □ $^{18}\text{O}_{\text{enam}}$ values can also be represented through the analysis of carbonate or

phosphate oxygen. Another key notable difference is in the definition of the term “drinking water”. $\delta^{18}\text{O}_{\text{dw}}$ values were measured from various sources, including meteoric water, predicted precipitation water, well water, tap water, or a mixture of the above. Thus, inter-laboratory differences cannot fully explain the observed differences between estimated drinking water values.

The reliability of predictive models is premised on the degree to which the human tissues can be assigned to a geographical locale and the understanding of the true source of drinking water in which the individual had consumed during the time of tissue formation. Models should then be based on the oxygen analysis of modern human enamel with known geographical information for the period in which tissue formed for the pairing with appropriate $\delta^{18}\text{O}_{\text{dw}}$ data. The preferred study is of known modern samples where geographical information can be obtained directly from the living donor over unknown or archaeological samples where inference of these types of information is required. Drinking water data should also be thoroughly examined as tap water with known source information can best provide the most accurate drinking water data.

Large offsets in predicted values can lead to significant errors when inferring an individual’s geographical movement for identification purposes. The results support the recommendation to standardize the methodology for the development of $\delta^{18}\text{O}_{\text{dw}}$ to $\delta^{18}\text{O}_{\text{enam}}$ equations, as stated in other papers^{17,73}. There is undeniably a much-needed refinement in the current predictive models and a need for consensus across the forensic community as to which predictive model is most appropriate for human provenancing. The alternative empirical approach to using predictive

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models for human provenancing recommended by several studies may be the more appropriate methodology for mapping human origin through stable oxygen isotopes. This approach involves the direct mapping of tissue isotopes themselves^{7,8} and limits the introduction of errors associated with predictive models. The development of such tissue-specific isotope maps will allow for a direct comparison of samples to reliable reference datasets.

Conclusion

This study aimed to assess the relationships between known bio-geographical enamel data to known city-wide drinking water data. Results indicate that although weak, there is a positive correlation between $\delta^{18}\text{O}_c$ and $\delta^{18}\text{O}_{\text{tap}}$ values at the city and country-level. Mean standard deviations measurements were carried out to assess the predictability of existing conversion models. The equation provided by Dotsika⁷⁰ proved to be the best model for this current set of enamel data. However, large differences between actual and predicted $\delta^{18}\text{O}_{\text{dw}}$ values were noted depending on the conversion equation or combination of conversion equations published in the literature. Thus, this study supports the development of *tissue-specific* $\delta^{18}\text{O}_c$ geographical reference maps. Standardization of methodology for isotopic analysis is also vital for the collaborative collection of globally represented data. The development of international $\delta^{18}\text{O}_{\text{enam}}$ databases would ultimately render conversion equations unnecessary and eliminate any associated errors. A meaningful approach would be to define boundaries confined by the tap water distribution systems rather than by way of political borders, which can be made possible with the detailed understanding of tap water supply and distribution systems for any given geographical area. The development of a reliable database would greatly benefit the forensic

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community by providing accurate guidance for unidentified individuals' possible region-of-origin determination. The understanding of the city-specific range of $\delta^{18}\text{O}_\text{c}$ values for Metro Vancouver (-9.7‰ to -7.2‰) now allows for oxygen isotope analysis to be used as an exclusionary tool where individuals with $\delta^{18}\text{O}_\text{c}$ values that exceed threshold values can be identified as non-local. Such a tool is vital for assisting the forensic community in missing persons investigations.

Data availability

All data analysed during the current study are available from the corresponding authors on reasonable request.

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Author contributions

MU wrote the main manuscript text and prepared all figures. MU and LSB conceived research design, analysed and interpreted results.

Figure legends

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23.65 (± 0.78); [df = 37; $\bar{R}^2 = 0.08$]. The solid grey line represents the regression line between $\delta^{18}\text{O}_c$ and $\delta^{18}\text{O}_{\text{OIPC}}$ values; Eq (8): $\delta^{18}\text{O}_{c(\text{VSMOW})} = 0.43 (\pm 0.05) \times \delta^{18}\text{O}_{\text{OIPC}(\text{VSMOW})} + 26.92 (\pm 0.46)$; [df=73, $\bar{R}^2 = 0.51$]. Generally, positive but weak correlations were observed between the two values.

Figure 10. Range of $\delta^{18}\text{O}_c$ values as predicted by an equation or a combination of equations from the actual MV $\delta^{18}\text{O}_{\text{tap}}$ range (-11.7 to -9.3 ‰). Equations include all existing predictive models and equations from the current study (U1, U2, U3, U4, U5, U6, U7, UOIPC). Predicted ranges were generally more positive than the actual range for MV. 15 of the 44 predicted ranges fell outside the actual MV range. The best performing predictive model was Dotsika⁴⁰ (Do), which accurately identified 91.7% of the MV individuals residing in MV during tissue formation.

Figure 11. $\delta^{18}\text{O}_{\text{dw}}$ values were calculated from all $\delta^{18}\text{O}_{c(\text{VSMOW})}$ values (n=75) using an existing predictive model or a set of models. For those equations giving the relationship between O_p to O_{dw} , the values were first converted to O_p from O_c using an equation that shows the relationship between carbonate and phosphate oxygen. Shapes denote carbonate to phosphate oxygen conversions. The OIPC line represents the relationship between $\delta^{18}\text{O}_{c(\text{VMOSW})}$ and $\delta^{18}\text{O}_{\text{OIPC}}$ values.

Figure 12. Differences between predicted and observed $\delta^{18}\text{O}_{\text{dw}}$ values for (a) tap water and (b) OIPC for all samples from the current study (n=75). $\delta^{18}\text{O}_{\text{dw}}$ values were predicted from $\delta^{18}\text{O}_c$ values with existing predictive models or a combination of models. $\delta^{18}\text{O}_c$ values were initially

converted to $\delta^{18}\text{O}_\text{p}$ values before applying the equations that represent the relationship between $\delta^{18}\text{O}_\text{p}$ and $\delta^{18}\text{O}_\text{dw}$ values. Shapes denote carbonate to phosphate oxygen conversions, and colors represent the enamel to drinking water conversion equations.

Figure 13. A direct comparison of the mean standard deviation (MSD) between predicted and observed $\delta^{18}\text{O}_\text{dw}$ values for (a) tap water and (b) OIPC. $\delta^{18}\text{O}_\text{dw}$ values were predicted from $\delta^{18}\text{O}_\text{c}$ values of all samples (n=75) from the current study with existing predictive models or a combination of models. MSD components are lack of correlation (LC), non-unity slope (NU) and squared bias (SB). Low MSD indicates greater similarity between modelled and measured values.

Tables

Table 1. Summary of molar formation⁷⁴.

	Tooth	Formation of tissue	Crown completion	Root completion
Permanent maxillary teeth	First	Birth	2.5 – 3 years	9 – 10 years
	Second	2.5 – 3 years	7 – 8 years	14 – 16 years
	Third	7 – 9 years	12 – 16 years	18 – 25 years
Permanent mandibular teeth	First	Birth	2.5 – 3 years	9 – 10 years
	Second	2.5 – 3 years	7 – 8 years	14 – 15 years
	Third	8 – 10 years	12 – 16 years	18 – 25 years

Table 2. Existing conversion equations.

Conversion	Study	Equation	n	r ²
VPDB to VSMOW	Coplen (1988)	$\delta^{18}\text{O}_\text{x/VSMOW} = 1.03091 \delta^{18}\text{O}_\text{x/VPDB} + 30.91$		
		$\delta^{18}\text{O}_\text{x/VPDB} = 0.97001 \delta^{18}\text{O}_\text{x/VSMOW} - 29.99$		
$\delta^{18}\text{O}_\text{p}$ to $\delta^{18}\text{O}_\text{c}$	Iacumin et al. ¹⁹	$\delta^{18}\text{O}_\text{p(SMOW)} = 0.98 \delta^{18}\text{O}_\text{c(SMOW)} - 8.5$	31	0.98
	Bryant et al. ¹⁸	$\delta^{18}\text{O}_\text{c} (\pm 1.3) = 1.02 (\pm 0.04) \delta^{18}\text{O}_\text{p} + 8.3 (\pm 0.7)$	42	0.99
	Wright et al. ²¹	$\delta^{18}\text{O}_\text{p(SMOW)} = 0.899 \delta^{18}\text{O}_\text{c(PDB)} + 21.397$		

	Chenery et al. ²⁰ (cold, temperate and warm humid climates)	$\delta^{18}\text{O}_p = 1.0322 (\pm 0.008) \delta^{18}\text{O}_c - 9.6849 (\pm 0.187)$	51	0.98
	Chenery et al. ²⁰ (hot-arid climates)	$\delta^{18}\text{O}_p = 1.122 \delta^{18}\text{O}_c - 13.73$	38	0.91
$\delta^{18}\text{O}_p$ to $\delta^{18}\text{O}_{dw}$	Longinelli ¹⁵	$\delta^{18}\text{O}_p = 0.64 \delta^{18}\text{O}_w + 22.37$	10	0.98
	Luz et al. ¹⁴	$\delta^{18}\text{O}_p = 0.78 \delta^{18}\text{O}_{dw} + 22.7$	6	0.97
	Levinson et al. ¹³	$\delta^{18}\text{O}_p = 0.46 \delta^{18}\text{O}_{dw} + 19.4$	40	0.93
	Daux et al. ⁹ (Eq. 4)	$\delta^{18}\text{O}_{dw} = 1.73 (\pm 0.21) \delta^{18}\text{O}_p - 37.25 (\pm 3.55)$	12	0.87
	Daux et al. (2008) (Eq. 5)	$\delta^{18}\text{O}_{dw} = 1.70 (\pm 0.22) \delta^{18}\text{O}_p - 39.28 (\pm 3.68)$	12	0.88
	Daux et al. (2008) (Eq. 6)	$\delta^{18}\text{O}_{dw} = 1.54 (\pm 0.09) \delta^{18}\text{O}_p - 33.72 (\pm 1.51)$	42	0.87
	*combined equation			
	Chenery et al. (2010) *corrected	$\delta^{18}\text{O}_p = 0.46 \delta^{18}\text{O}_{dw} + 20.8$	40	
	Levinson et al. (1987)			
	Pollard et al. ⁷ *superset	$\delta^{18}\text{O}_p = 0.531 \delta^{18}\text{O}_{dw} + 20.52$	94	0.907
	Hermann et al. (2015)	$\delta^{18}\text{O}_p = 0.62 \delta^{18}\text{O}_{dw} + 21.74$	45	0.63
	Warner et al. (2018)	$\delta^{18}\text{O}_{dw} = 1.64 \delta^{18}\text{O}_p - 31.35$	27	0.54
$\delta^{18}\text{O}_c$ to $\delta^{18}\text{O}_{dw}$	Ehleringer et al. ⁵	$\delta^{18}\text{O}_{c(\text{VSMOW})} = 0.350 \delta^{18}\text{O}_{dw} + 27.6$	5	0.85
	Posey (2011)	$\delta^{18}\text{O}_{c(\text{VSMOW})} = 0.77 \delta^{18}\text{O}_{dw} + 28.1$	58	0.70
	Keller et al. (2016)	$\delta^{18}\text{O}_{c(\text{VSMOW})} = 0.396 \delta^{18}\text{O}_{dw} + 26.9$	156	0.81
	Dotsika (2020)	$\delta^{18}\text{O}_{dw} = 1.020 \delta^{18}\text{O}_c - 32.941$	16	0.87

837 Table 3. Fractional contributions of water sources to MV municipal tap water supply.

	Capilano	Seymour	Coquitlam	Well	
Mean annual $\delta^{18}\text{O}_{\text{tap}}$	-11.6	-11.6	-11.1	-11.1	
Municipality	Fractional contributions (f)				$\delta^{18}\text{O}_{(\text{mean})}$
Burnaby	0.045	0.955	0	0	-11.7
Coquitlam	0	0	1.00	0	-11.1
Langley and Township of Langley	0	0	0.47	0.53	-11.1
New Westminster	0	0.20	0.80	0	-11.2
North Vancouver	0.50	0.50	0.00	0.00	-11.7
Richmond	0.495	0.495	0.00	0.00	-11.6
Surrey	0.00	0.41	0.59	0.00	-11.3
Vancouver	0.62	0.38	0.00	0.00	-11.7

838 Table 4. Summary of studies on predictive models.

Study	Time period	Sampled Material	Geographical scale	Definition of drinking water	Oxygen analysis
Longinelli (1984)	End of 19 th century	Human bone	Global	Meteoric water	Phosphate (BiPO ₄)

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Luz et al. (1984)	unspecified	Human tooth	Global	Estimated precipitation and tap water	Phosphate (BiPO_4)
Levinson et al. (1987)	Modern	Human tooth	Global	Unspecified	Phosphate (BiPO_4)
Daux et al. (2008) Eq. 4	Modern and historical (Greenland)	Human 2 nd and 3 rd molars	Global	Tap water (OPIC for Disko Bay)	Phosphate (Ag_3PO_4)
Daux et al. (2008) Eq. 5	Modern and historical (Greenland)	Human 2 nd and 3 rd molars	Global	OIPC precipitation estimate	Phosphate (Ag_3PO_4)
Ehleringer et al. ⁵	Modern	Human tooth	Country (U.S.)	Tap water	Carbonate
Posey (2011)	Modern	Human tooth (incisors, canines, premolars, molars)	Region (Middle East)	Tap water	Carbonate
Hermann et al. (2015)	Modern	Human tooth	Country (U.S.)	Meteoric water	Phosphate (Ag_3PO_4)
Keller et al. (2016)	Modern	Human 3 rd molars	Country (U.S.)	River water	Carbonate
Warner et al. (2018)	Modern	Human 3 rd molars	State (Mississippi, U.S.)	Tap water	Carbonate
Dotsika (2020)	Modern	Human 2 nd molars	Country (Greece)	Spring water	Carbonate

Figures

Figure 1

Locations of residence during tissue formation with the corresponding $\delta^{18}\text{O}_{\text{C(VPDB)}}$ values for all collected samples. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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Figure 3

Boxplots of MV $\delta^{18}\text{O}_\text{C}$ values as represented by a) city and b) tap water source. The boxes represent 25th and 75th percentiles, and whiskers represent the maximum and minimum observations. When outliers are present, the whiskers extend to 1.5 times the interquartile range. The middle line denotes the median or 50th percentile. One-way ANOVA tests showed no statistically significant inter-city differences

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Monthly MV tap water $\delta^{18}\text{O}$ data for samples taken between June 2017 to May 2018 at Coquitlam Water Treatment Plant (CWTP) and Seymour-Capilano Filtration Plant (SCFP). Mean annual MV tap water $\delta^{18}\text{O}$ values were -11.1‰ and -11.7‰ at CWTP and SCFP, respectively.

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Figure 7

A boxplot of $\delta^{18}\text{O}_C$ values for the globally aggregated data represented by country ($n=75$). The boxes represent 25th and 75th percentiles, and whiskers represent the maximum and minimum observations. When outliers are present, the whiskers extend to 1.5 times the interquartile range. The middle line denotes the median or 50th percentile.

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Figure 12

Differences between predicted and observed $\delta^{18}\text{O}_{\text{dw}}$ values for (a) tap water and (b) OIPC for all samples from the current study ($n=75$). $\delta^{18}\text{O}_{\text{dw}}$ values were predicted from $\delta^{18}\text{O}_{\text{C}}$ values with existing predictive models or a combination of models. $\delta^{18}\text{O}_{\text{C}}$ values were initially converted to $\delta^{18}\text{O}_{\text{P}}$ values before applying the equations that represent the relationship between $\delta^{18}\text{O}_{\text{P}}$ and $\delta^{18}\text{O}_{\text{dw}}$ values.

Shapes denote carbonate to phosphate oxygen conversions, and colors represent the enamel to drinking water conversion equations.

Figure 13

A direct comparison of the mean standard deviation (MSD) between predicted and observed $\delta^{18}\text{O}_{\text{dw}}$ values for (a) tap water and (b) OIPC. $\delta^{18}\text{O}_{\text{dw}}$ values were predicted from $\delta^{18}\text{O}_{\text{c}}$ values of all samples ($n=75$) from the current study with existing predictive models or a combination of models. MSD components are lack of correlation (LC), non-unity slope (NU) and squared bias (SB). Low MSD indicates greater similarity between modelled and measured values.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SupplementaryTableS1.xlsx](#)
- [SupplementaryTableS2.xlsx](#)
- [SupplementaryTableS3.xlsx](#)
- [SupplementaryTableS4.xlsx](#)