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PAPER

GENERAL

Momoko Ueda, M.A.; and Lynne S. Bell, Ph.D.

A City-wide Investigation of the Isotopic Distribution and Source of Tap Waters for Forensic Human Geolocation Ground-truthing*

ABSTRACT: Human geolocation is prefaced on the accuracy of the geographic precision of mapped isotopic values for drinking water. As most people live in cities, it becomes important to understand city water supplies and how the isotopic values uniquely reflect that city. This study investigated the isotopic distribution of $\delta^2 H$ and $\delta^{18} O$ from sourced tap waters that were collected from across the Metro Vancouver (MV) area (n=135). The results revealed that the isotopic values reflect their water sources with a range of 5.3% for $\delta^{18} O_{tap}$ and 29.3% for $\delta^{2} H_{tap}$ for MV. The results indicate that individual cities need higher resolution studies to determine their tap water isotopic ranges, and a good understanding of the water supply network itself for human geolocation work. With an extended high-resolution understanding of each city, human tissue may be compared with more certainty for geolocation.

KEYWORDS: forensic science, stable isotopes, tap water, oxygen, hydrogen, human geolocation, city, water source

Stable isotope analysis of human tissues has been applied to the field of forensics primarily to aid in the process of human identification (1-3). The isotopic compositions of human tissues are reflective of the environment in which tissue formed, with drinking water being the major contributor for hydrogen and oxygen stable isotopes (1,4–10). Drinking water is derived from precipitation, which includes all forms of water-rain, ice, or snow—which are involved in the hydrological cycle. The relationship between hydrogen $(\delta^2 H_{ppt})$ and oxygen $(\delta^{18} O_{ppt})$ isotopes of precipitation water on the global scale is expressed as— $\delta^2 H_{ppt} = 8$ $\delta^{18} O_{ppt} + 10$ —and is known as the Global Meteoric Water Line (GMWL). Global $\delta^2 H_{ppt}$ and $\delta^{18} O_{ppt}$ follow a general negative distribution with increasing latitude (11– 14), and thus, the isotopic compositions of precipitation differ geographically across the globe. Furthermore, local physiographic factors can influence the isotopic values of local precipitation (15). Isotopic values of human tissues are observed to follow a similar trend to that of global precipitation (4,5,8,9,16) and can provide insight into the geographical history of an individual.

The isotopic compositions of human drinking water are often estimated through the use of the Online Isotopes of Precipitation Calculator (OIPC). The OIPC is a tool for estimating mean monthly and annual isotopic compositions of $\delta^{18}O_{ppt}$ and δ^2H_{ppt} in areas where no such data exist (17). The algorithm is largely based off Global Network of Isotopes in Precipitation (GNIP)

¹School of Criminology, Centre for Forensic Research, Simon Fraser University, 8888 University Dr. Burnaby, BC, V5A 1S6 Canada.

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data. GNIP is a global precipitation monitoring program that collects and analyzes oxygen and hydrogen isotopes and was put into operation by International Atomic Energy Agency (IAEA)'s water resources program and the World Meteorological Organization (WMO) in 1961. The OIPC is a user-friendly online calculator where the users can simply input the latitude, longitude, and elevation data of a geographical location to generate the estimates of $\delta^{18}O_{ppt}$ and δ^2H_{ppt} .

Although human drinking water is generally derived from precipitation water, it is important to gain an understanding of the isotopic compositions of tap water. Over half of the global human population rely on tap water systems as the primarily drinking water supply (18). The isotopic compositions of tap water, as opposed to precipitation water, have not been studied extensively across the globe, with the exception of an interpolated tap water data for the contiguous United States (19,20). Bowen et al. (19) generated a prediction map of oxygen $(\delta^{18}O_{tap})$ and hydrogen (δ^2H_{tap}) isotopes in tap water across the United States by comparing tap water and precipitation data. This study became the basis for subsequent studies investigating the isotopic relationships of tap water and human tissues. The relationships between both $\delta^2 H$ and $\delta^{18} O$ values of human hair and tap waters were reported by Ehleringer et al. (1), which further applied Bowen et al.'s tap water model to develop a prediction map of δ^2 H and δ^{18} O for human hair across the contiguous United States. Ehleringer et al.'s study holds significant forensic value as a pioneering project by directly mapping isotopic compositions of human tissue.

Tap water from cities across the United States have been collected and analyzed for its $\delta^2 H$ and $\delta^{18} O$ values (19,21); however, no study, to date, has looked at a single city in detail. It is important to study cities, as tap waters are distributed through piped systems, and the origin of the water source may come

from multiple sources or be at some distance from the tap itself (21). Tap water may be sourced from physiographically distinct and distant areas to that of which it supplies, and, in such a case, local precipitation data of $\delta^{18}O_{ppt}$ and $\tilde{\delta}^2H_{ppt}$ will not match that of tap water from the geographic location where water is consumed. The sources themselves may not only reflect a geography but also, depending on their origin, may incorporate a temporal component that may be at variance with local precipitation (15,21-26). The complex distribution systems of tap water for cities are generally known, as infrastructure is built specifically for the delivery of tap waters throughout a city. For example, tap water for Metro Vancouver (MV), located in British Columbia, Canada, is supplied by multiple water sources, and is delivered to each municipality through major water mains. The distribution of tap waters is controlled and monitored on an hourly basis via a sophisticated electronic control grid.

MV, previously known as the Greater Vancouver Regional District or GVRD, is a regional district that comprises of 21 municipalities, one electoral area and one treaty of First Nations (27). Three major watersheds that are closely monitored and operated by the Greater Vancouver Water District (GVWD)— Capilano, Seymour, and Coquitlam-supply the majority of MV's drinking water to businesses and residential areas (Fig. 1). The three watersheds are located on the northern mountains of MV. The Capilano watershed is located at latitude 49°22'47" and longitude 123°07' 29", Seymour watershed at latitude 49°27′12" and longitude 122°57′33", and Coquitlam watershed at latitude 49°24'15" and longitude 122°47'05" (28). These watersheds are comprised of a mixture of rainwater, snowmelt, and surface runoffs from creeks and streams from mountains surrounding the watersheds (29). The watersheds are composed of water collected and stored throughout the year with the months of September to March receiving much of the annual precipitation. Tap water from the watersheds is distributed by gravity through the main pipes that run down to MV municipalities. The three watersheds together supply 99% of the entire MV, and a complex network of groundwater aquifers supply the remaining

Whilst there is some understanding of the macro distribution of global precipitation and some country-wide understanding of tap water values within the United States, the importance of more highly resolved data has, to date, not been investigated at the city level. As stated, tap water values can differ across a single locale or large conurbation, and if drinking waters are outsourced, it could present potentially confounding values. The aim of this study was to understand how the known MV tap water source and distribution system would translate to the isotopic distribution of oxygen and hydrogen values derived from those tap waters, in order to provide a detailed isotopic map for MV. This would be important for human geolocation as it would provide an isotopic range that is specific to MV tap waters, which may differ from ranges observed in other cities, and can thus be compared to isotopic values obtained from human tissues.

Materials and Methods

Ethics Permission

In review of the research proposal outlining the details of the sample collection process and the interaction with participants, Simon Fraser University Office of Research Ethics had granted an exemption from the requirements of Research Ethics Review for this study.

Sample Acquisition

A total of 141 samples of tap water were collected for this study. A total of 135 samples were collected from across MV with the remaining six from Abbotsford (British Columbia, Canada), Whistler (British Columbia, Canada), Beaver Creek (Yukon, Canada), Whitehorse (Yukon, Canada), Point Roberts (Washington, USA), and Fairbanks (Alaska, USA), which were collected for comparison purposes (Fig. 2). MV sampling sites were selected to evenly cover the geographical area, and included 16 of the 21 MV municipalities. Samples were collected with the help of volunteers, who were personally known to the principal investigator, during the summer week of June 23 to June 29, 2013. Tap water samples were specifically collected during the summer, as it reflects annual precipitation water. MV precipitation falls during the winter months from September to March, which are collected and stored in the MV watersheds for distribution throughout the year. A total of 114 vials were distributed to volunteers, in which 94 of the vials were retrieved for analysis. The principal investigator collected the remaining 47 samples. Nineteen of the total number of samples were collected outside of the designated collection week.

All samples were collected in an airtight 8-dram short form style VWR borosilicate glass vial with phenolic screw on cap and polyvinyl-faced pulp liner to ensure no leakage of water during storage. Samples were stored in a refrigerator immediately after retrieval. The vials were checked for any signs of leakage or evaporation and presence of large air bubbles prior to storage.

MV Water Source Information

GVWD controls and monitors the distribution of tap waters to municipalities across MV. Source information on the sampled tap waters was obtained from GVWD according to its location of retrieval. The MV tap water distribution system is an active system with the main pipes emptying and refreshing its water source up to three times a day. The water source provided to a certain locale can differ from time to time, even changing by the hour, depending on demand and water quality that is often affected by elevated levels of turbidity. Thus, the transit time from the main pipes to its final destination can vary periodically. GVWD closely monitors these changes to tap water sources and thus were able to provide accurate source data.

While approximately 99% of MV tap water is sourced by the three major watersheds, the remaining areas are supplied by well water taken from a complex network of 18 aquifers that underlie the Township of Langley. Tap water samples were collected from these areas to cover the entire geographical area of MV; however, the samples could not be identified to a specific aquifer, and thus, the source is effectively unknown. Thus, all tap water samples that are sourced by an aquifer will be generally labeled as "well water".

Sample Analysis

 $\delta^2 H_{tap}$ and $\delta^{18} O_{tap}$ were measured using a Los Gatos Triple Liquid Water Isotope Analyzer (LG-TLWIA), model TLWIA-45-EP 2013, connected to CTC Analytics LEAP Technology PAL liquid auto-sampler. LG-TLWIA is an off-axis integrated cavity output spectroscopy (ICOS) laser system, which is a laser absorption technology where the water molecules are measured directly rather than through the conversion to gases (31). This

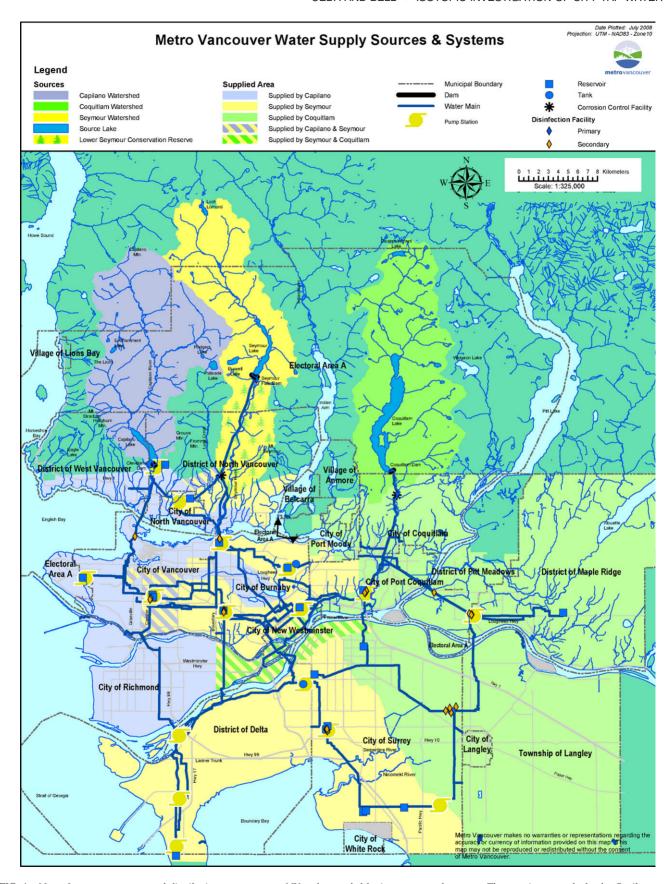


FIG. 1—Map of tap water source and distribution system across MV, colour-coded by its water supply source. Three major watersheds, the Capilano watershed, the Seymour watershed, and the Coquitlam watershed, lie north of MV. Transport of tap water is through main pipes that run from the watersheds and are driven by gravity. The colours indicate the areas supplied by each of the watersheds. Tap water for much of Township of Langley is sourced from groundwater aquifers. Used with permission (34). [Color figure can be viewed at wileyonlinelibrary.com]





FIG. 2—Map of sample collection sites. Sampling sites were selected to evenly cover the geographical area of MV. Tap water samples were collected from 16 of the 21 MV municipalities. Six samples were collected outside of MV for comparison purposes. [Color figure can be viewed at wileyonlinelibrary.com]

instrument is recommended by the International Atomic Energy Agency (IAEA) (32). The working standards were that of Los Gatos Standards 1A, 2A, 3A, 4A, and 5A. All values are reported in the δ -notation and normalized to VSMOW-SLAP and GISP standard.

The instrument was calibrated to ensure that the laser was centered to the absorption graph prior to running the analysis. A new septum was installed into the injection port before running every batch of analysis to ensure no leakage of atmospheric air into the equipment. The injection port was set to a temperature of 100°C. Four hundred microlitres of each standard and sample was pipetted into a 1.4-mL vial and capped with a Mini-UniPrep

syringeless nylon filter media with polypropylene housing and pore size of 0.45 μm to ensure removal of any foreign particulates. Each tray held a batch consisting of 54 vials. Each of the five standards was similarly prepared onto a separate tray. TLWIA postanalysis software was utilized for the data correction process. Statistical calculations were performed using SPSS Statistics 2, at the 99% level of confidence, and the generation of graphs was carried out in Microsoft Excel for Mac 2011.

This generation of 2013 LG-TLWIA model has improved precisions over previous models with published precisions of $\delta^2 H = \pm 0.3\%$ and $\delta^{18}O = \pm 0.1\%$ (31,33–35). The published precisions were checked against internal standards as well as

TABLE 1— $\delta^{18}O_{tap}$ and δ^2H_{tap} data for the all analyzed tap water samples.

| Sample | Municipality | Collection Date (DD/MM/YY) | Water Source | $\delta^{18}O_{tap}~(\%_o)~VSMOW$ | $\delta^2 H_{tap}$ (%) VSMOW |
|----------------------|--------------------------|-------------------------------|--------------------------------|-----------------------------------|------------------------------|
| 1 | Vancouver | 23/06/13 | Seymour | -13.9 | -98.3 |
| 2 | Vancouver | 23/06/13 | Seymour | -13.9 | -98.5 |
| 3 | Richmond | 25/06/13 | Capilano/Seymour | -13.7 | -96.9 |
| | | | mix | | |
| 4 | Burnaby | 23/06/13 | Seymour | -13.9 | -98.6 |
| 5 | Township of Langley | 24/06/13 | Coquitlam | -12.9 | -90.2 |
| 7 | Vancouver | 26/06/13 | Seymour | -13.7 | -98.5 |
| 9 | Vancouver | 27/06/13 | Capilano | -13.2 | -94.8 |
| 10 | Richmond | 23/06/13 | Seymour | -13.2 -13.6 | -94.8 -97.6 |
| | | 27/06/13 | | -13.0 -13.0 | -97.0 -93.5 |
| 11 | Surrey | | Seymour | | |
| 12 | Vancouver | 27/06/13 | Capilano | -13.4 | -96.7 |
| 13 | Vancouver | 27/06/13 | Capilano | -13.1 | -95.4 |
| 14 | Vancouver | 27/06/13 | Capilano | -13.5 | -96.2 |
| 15 | Surrey | 28/06/13 | Seymour | -13.8 | -98.0 |
| 18 | Burnaby | 24/06/13 | Seymour | -13.6 | -95.8 |
| 19 | Vancouver | 24/06/13 | Seymour | -13.5 | -97.5 |
| 20 | Richmond | 24/06/13 | Seymour | -13.2 | -94.8 |
| 22 | Coquitlam | 27/06/13 | Coquitlam | -12.7 | -89.9 |
| 23 | Coquitlam | 26/06/13 | Coquitlam | -12.5 | -90.2 |
| 25 | Township of Langley | 25/06/13 | Coquitlam | -12.7 | -90.4 |
| 26 | North Vancouver District | 24/06/13 | | -12.7 -13.4 | -90.4 -95.4 |
| | | | Capilano | | |
| 27 | Burnaby | 24/06/13 | Capilano/Seymour | -13.5 | -95.9 |
| | | | mix | | |
| 28 | Burnaby | 26/06/13 | Seymour | -13.6 | -96.8 |
| 32 | City of North Vancouver | 26/06/13 | Capilano | -13.7 | -96.1 |
| 34 | North Vancouver District | 23/06/13 | Seymour | -13.8 | -98.4 |
| 37 | Vancouver | 25/06/13 | Seymour | -13.2 | -95.4 |
| 38 | Surrey | 23/06/13 | Coquitlam | -13.1 | -90.6 |
| 39 | Maple Ridge | 24/06/13 | Coquitlam | -12.2 | -88.5 |
| 40 | Vancouver | 25/06/13 | Capilano | -13.5 | -95.8 |
| 41 | Vancouver | 28/06/13 | Capilano | -13.3 | -95.2 |
| 42 | Vancouver | 26/06/13 | | -13.5 -13.2 | -95.2 -95.2 |
| | | | Capilano | | |
| 43 | Richmond | 25/06/13 | Capilano/Seymour | -13.6 | -97.4 |
| 44 | Richmond | 28/06/13 | mix Capilano/Seymour mix | -13.3 | -97.1 |
| 15 | Vancouver | 29/06/13 | Capilano | -13.4 | -98.1 |
| 45 | | | | | |
| 46 | Vancouver | 27/06/13 | Capilano | -13.2 | -96.8 |
| 48 | Vancouver | 26/06/13 | Capilano | -12.5 | -95.3 |
| 50 | Port Moody | 23/06/13 | Coquitlam | -12.2 | -90.0 |
| 51 | Richmond | 27/06/13 | Capilano/Seymour | -13.5 | -97.6 |
| 52 | Richmond | 28/06/13 | mix Capilano/Seymour mix | -14.1 | -97.3 |
| 52 | Dolto | 24/06/12 | | 12.4 | 05.2 |
| 53 | Delta Delta | 24/06/13 | Well | -13.4 | -95.2 -98.9 |
| 54 | Delta | 24/06/13 | Seymour | -14.2 | |
| 55 | Delta | 24/06/13 | Well | -13.6 | -95.1 |
| 56 | Delta | 28/06/13 | Capilano/Seymour mix | -13.6 | -95.4 |
| 57 | West Vancouver | 24/06/13 | Capilano | -13.3 | -93.4 |
| 58 | Burnaby | 25/06/13 | Seymour | -14.0 | -98.7 |
| 60 | Vancouver | 25/06/13 | Capilano | -13.4 | -96.1 |
| 61 | Maple Ridge | 25/06/13 | Coquitlam | -12.7 | -90.3 |
| 62 | Maple Ridge | 24/06/13 | Well | -12.5 | -89.8 |
| 64 | Coquitlam | 26/06/13 | Coquitlam | -12.6 | -90.7 |
| 67 | Surrey | 26/06/13 | Seymour/Coquitlam mix | -12.0 | -93.4 |
| 69 | New Westminster | 27/06/13 | Coquitlam | -13.2 | -89.8 |
| 70 | Port Moody | 25/06/13 | Coquitlam | -13.0 | -89.8 |
| 72 | Richmond | 28/06/13 | Capilano/Seymour mix | -13.7 | -97.4 |
| 73 | Vancouver | 24/06/13 | Capilano | -12.9 | -95.0 |
| 74 | Township of Langley | 23/06/13 | Coquitlam | -12.4 | -90.3 |
| 7 4 75 | | | Well | -12.4 -11.0 | -90.5 -83.4 |
| | Township of Langley | 23/06/13 | | | |
| 76 77 | Township of Langley | 23/06/13 | Well | -10.8 | -81.6 |
| 77 7 2 | Township of Langley | 23/06/13 | Well | -10.7 | -80.5 |
| 78 | Township of Langley | 23/06/13 | Well | -10.9 | -80.5 |
| 79 | Abbotsford, BC | 23/06/13 | | -11.1 | -82.4 |
| 81 | Surrey | 23/06/13 | Coquitlam | -12.1 | -89.2 |
| 82 | Township of Langley | 23/06/13 | Well | -9.3 | -69.6 |

TABLE 1—Continued.

| Sample | Municipality | Collection Date (DD/MM/YY) | Water Source | $\delta^{18}O_{tap}$ (%) VSMOW | $\delta^2 H_{tap}$ (%) VSMOW |
|------------|---|----------------------------|---------------------------------|--------------------------------|------------------------------|
| 83 | City of Langley | 23/06/13 | Coquitlam | -11.3 | -83.4 |
| 84 | Surrey | 23/06/13 | Coquitlam | -13.1 | -90.2 |
| 85 | Maple Ridge | 23/06/13 | Coquitlam | -12.9 | -89.3 |
| 86 | City of Langley | 23/06/13 | Coquitlam | -13.1 | -89.8 |
| 87 | Delta | 25/06/13 | Well | -14.3 | -98.4 |
| 88 | Port Coquitlam | 23/06/13 | Coquitlam | -12.9 | -89.5 |
| 89 | Surrey | 25/06/13 | Coquitlam | -12.7 | -89.1 |
| 90 | Surrey | 25/06/13 | Seymour | -13.6 | -98.0 |
| 91 | Richmond | 25/06/13 | Capilano/Seymour mix | -13.0 | -96.8 |
| 92 | Richmond | 25/06/13 | Capilano/Seymour mix | -13.3 | -97.6 |
| 93 | New Westminster | 25/06/13 | Coquitlam | -11.7 | -89.8 |
| 94 | Surrey | 25/06/13 | Seymour | -13.0 | -97.7 |
| 95 | Surrey | 25/06/13 | Coquitlam | -12.6 | -90.3 |
| 96 | Surrey | 25/06/13 | Seymour | -13.8 | -98.4 |
| 97 98 | Vancouver | 26/06/13 | Seymour | -13.5 -14.1 | -96.3 -97.8 |
| 100 | Richmond Vancouver | 25/06/13 28/06/13 | Capilano/Seymour mix Seymour | -14.1 -13.9 | -97.8 -98.1 |
| 100 | Vancouver | 28/06/13 | Seymour | -13.9 -14.1 | -98.1 -98.2 |
| 101 | Vancouver | 23/06/13 | Capilano | -14.1 -13.7 | -98.2 -95.4 |
| 103 | New Westminster | 28/06/13 | Capitalio | -13.7 -13.2 | -90.8 |
| 105 | New Westminster | 28/06/13 | Coquitlam | -13.2 -14.0 | -98.4 |
| 106 | Surrey | 27/06/13 | Coquitlam | -13.5 | -97.0 |
| 108 | Surrey | 27/06/13 | Seymour | -13.5 | -98.2 |
| 109 | Point Roberts, WA | 31/07/13 | Seymour | -12.9 | -95.6 |
| 111 | Burnaby | 29/06/13 | Seymour | -13.2 | -98.7 |
| 112 | Richmond | 01/08/13 | Capilano | -12.2 | -94.6 |
| 113 | Coquitlam | 28/06/13 | Coquitlam | -11.7 | -90.4 |
| 114 | Township of Langley | 24/06/13 | Well | -11.5 | -81.3 |
| 115 | Burnaby | 26/06/13 | Seymour | -13.8 | -98.7 |
| 116 | Pitt Meadows | 29/06/13 | Coquitlam | -12.8 | -91.2 |
| 117 | Burnaby | 24/06/13 | Seymour | -13.4 | -97.9 |
| 118 | North Vancouver District | 27/06/13 | Seymour | -13.3 | -97.8 |
| 121 | North Vancouver District | 27/06/13 | Capilano | -13.0 | -94.7 |
| 122 123 | Port Coquitlam North Vancouver District | 29/06/13 | Coquitlam | $-12.2 \\ -13.3$ | $-88.8 \\ -94.9$ |
| 123 | Surrey Surrey | 07/08/13 25/06/13 | Seymour Coquitlam | -13.5 -12.5 | -94.9 -88.4 |
| 124 | Vancouver | 26/06/13 | Seymour | -12.5 -14.6 | -86.4 -97.9 |
| 126 | Surrey | 25/06/13 | Seymour | -14.0 -14.1 | -97.9 -97.2 |
| 127 | Burnaby | 26/06/13 | Seymour | -13.8 | -98.6 |
| 130 | Delta | 25/06/13 | Seymour | -13.7 | -97.9 |
| 131 | Vancouver | 25/06/13 | Capilano/Seymour mix | -13.4 | -95.7 |
| 132 | City of North Vancouver | 27/06/13 | Capilano | -13.5 | -95.8 |
| 133 | Burnaby | 26/06/13 | Seymour | -13.7 | -98.1 |
| 134 | Vancouver | 26/06/13 | Capilano | -13.7 | -96.5 |
| 135 | New Westminster | 26/06/13 | Seymour | -13.8 | -98.2 |
| 136 | Vancouver | 26/06/13 | Capilano | -13.6 | -95.3 |
| 138 | North Vancouver District | 29/07/13 | Seymour | -13.6 | -96.4 |
| 143 | Whitehorse, YT | 04/08/13 | | -19.1 | -149.6 |
| 144 | Port Coquitlam | 31/07/13 | Coquitlam | -12.5 | -90.4 |
| 145 | Surrey | 28/07/13 | Coquitlam | -10.6 | -78.0 |
| 146 | Burnaby | 28/07/13 | Seymour | -13.6 | -96.5 |
| 147 | Maple Ridge | 31/07/13 | Coquitlam | -12.7 | -90.1 |
| 148 | Surrey Township of Langley | 28/07/13 | Coquitlam | -12.9 | -91.3 |
| 149 | Township of Langley | 28/07/13 | Coquitlam | -12.5 | -89.6 |
| 150 | Township of Langley | 28/07/13 | Well | -11.3 | -81.8 |
| 151 152 | Surrey Richmond | 28/07/13 28/07/13 | Conilana | -13.1 | -93.8 -95.1 |
| 152 | City of Langley | 28/07/13 28/07/13 | Capilano Coquitlam | -13.1 -12.7 | -95.1 -90.0 |
| 153 | Township of Langley | 28/07/13 | Well | -12.7 -11.3 | -90.0 -82.2 |
| 155 | Whistler, BC | 18/08/13 | **** | -11.5 -16.1 | -62.2 -119.3 |
| 160 | Fairbanks, AK | 19/08/13 | | -20.9 | -119.5 -164.5 |
| 161 | Beaver Creek, YT | -/08/13 | | -22.6 | -177.0 |

with the LG STD that are certified to be calibrated against international standards of VSMOW and VSLAP from the IAEA.

Approximately 1000 nL of water was injected into the heated septum port by the auto-sampler. All samples were prefiltered to $0.45 \mu m$, and each sample was injected eight times. The first two

injections were discarded as conditioning, and the remaining six injections were discarded as conditioning, and the remaining six injections were averaged for reporting. Duplicates were run at a minimum of every tenth sample. The precisions for the check measurements of $\delta^2 H$ and $\delta^{18} O$ that were conducted over a period of 8 months were conservative at 0.8% and 0.2%, respectively. **OIPC**

The OIPC is a popular tool for estimating the isotopic values of local precipitation for locations where no such data are available. Isotopic compositions of annual precipitation, with its 95% confidence interval, were estimated using OIPC for Vancouver, Township of Langley, and Point Roberts, WA, as well as for the three GVWD watersheds. These values were compared against mean MV tap water values that were obtained in this study. The watersheds are continuously supplied by water from a mixture of sources throughout the year, and winter precipitation contributes significantly to the watersheds, as MV receives much of its precipitation during the winter months. Thus, it is appropriate to compare OIPC values with MV tap water collected for this study.

Results

MV Tap Water

Isotopic variances of $\delta^{18}O_{tap}$ and δ^2H_{tap} were observed for tap waters collected across MV. Raw data on $\delta^{18}O_{tap}$ and δ^2H_{tap} values for the collected tap water samples are presented in Table 1, along with information on the municipality of sampling sites, date of sample collection, and water source information obtained from GVWD. The water sources for 18 of the MV tap water samples could not be determined and were excluded from the analysis. The negative isotopic values signify the depletion of the heavier isotope, whereas the positive values indicate the enrichment of the heavier isotope.

Mean values of $\delta^{18}O_{tap}$ and δ^2H_{tap} for each MV municipality are given in Table 2. Tap water collected from Delta had the most negative mean $\delta^{18}O_{tap}$ value of $-13.8\%_{o}\pm0.4$ (σ), and Township of Langley showed the most positive $\delta^{18}O_{tap}$ mean value of $-11.4\%_{o}\pm1.0$. Burnaby showed the lightest δ^2H_{tap} value of $-97.7\%_{o}\pm1.2$, and Township of Langley had the most positive mean δ^2H_{tap} value with $-83.5\%_{o}\pm6.0$. Mean $\delta^{18}O_{tap}$ and δ^2H_{tap} of all tap water collected across MV were $-13.1\%_{o}\pm0.9$ and $-93.5\%_{o}\pm5.3$, respectively. Non-MV tap water samples showed lighter values compared to MV tap water samples.

The isotopic values of collected tap waters were seen to cluster among those from the same water source (Fig. 3). Samples from the Seymour watershed showed the lightest isotopic values compared to samples from other water sources, and well water samples showed comparatively heavier values. Five samples from the Coquitlam watershed (samples 83, 105, 106, 145, and 151) and five samples from the well water source (samples 53, 55, 62, 82, and 87) showed significant deviations from their group and thus were noted as outliers. Outliers were defined as those samples with isotopic values that were significantly not consistent with other samples from the same water source.

The isotopic values of all tap water samples that were sourced by the GVWD watersheds and wells had a range of 5.3%, for $\delta^{18}O_{tap}$, with the most positive value of $-9.3\%_{o}$ to the most negative value of $-14.6\%_{o}$ (Table 3). $\delta^{2}H_{tap}$ of MV tap waters showed a 29.3%, range from $-69.5\%_{o}$ to $-98.8\%_{o}$. The tap water from Point Roberts, WA (sample 109), was included in this analysis as it was sourced by the Seymour watershed.

Mean isotopic values for MV tap waters from the Capilano watershed were $-13.3\%_o \pm 0.4$ for $\delta^{18}O_{tap}$ and $-95.6\%_o \pm 1.0$ for δ^2H_{tap} (Table 3). Mean isotopic values for tap waters that were sourced by the Seymour watershed were $-13.6\%_o \pm 0.4$

TABLE 2—Mean and standard deviations of $\delta^{18}O_{tap}$ and δ^2H_{tap} for each MV municipality.

| Municipality | n | δ ¹⁸ O _{tap} (‰) VSMOW Mean ± SD | $\begin{array}{c} \delta^2 H_{tap}~(\%)\\ VSMOW\\ Mean~\pm~SD \end{array}$ |
|------------------------|-----|--|--|
| Burnaby | 11 | -13.6 ± 0.2 | -97.7 ± 1.2 |
| City of Langley | 3 | -12.4 ± 0.9 | -87.7 ± 3.8 |
| City of North | 2 | -13.6 ± 0.1 | -96.0 ± 0.2 |
| Vancouver | | | |
| Coquitlam | 4 | -12.4 ± 0.5 | -90.3 ± 0.3 |
| Delta | 6 | -13.8 ± 0.4 | -96.8 ± 1.8 |
| Maple Ridge | 5 | -12.6 ± 0.3 | -89.6 ± 0.7 |
| New Westminster | 5 | -13.2 ± 0.9 | -93.4 ± 4.5 |
| North Vancouver | 6 | -13.4 ± 0.3 | -96.3 ± 1.5 |
| District | | | |
| Pitt Meadows | 1 | -12.8 | -91.2 |
| Port Coquitlam | 3 | -12.5 ± 0.3 | -89.6 ± 0.8 |
| Port Moody | 2 | -12.6 ± 0.6 | -89.9 ± 0.1 |
| Richmond | 13 | -13.4 ± 0.5 | -96.8 ± 1.1 |
| Surrey | 18 | -12.9 ± 0.8 | -92.9 ± 5.2 |
| Township of | 12 | -11.4 ± 1.0 | -83.5 ± 6.0 |
| Langley | | | |
| Vancouver | 25 | -13.5 ± 0.4 | -96.5 ± 1.3 |
| West Vancouver | 1 | -13.3 | -93.4 |
| All Metro Vancouver | 117 | -13.1 ± 0.9 | -93.5 ± 5.3 |

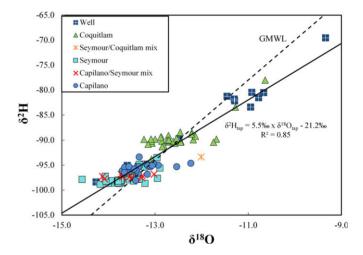


FIG. $3-\delta^{18}O$ versus δ^2H plot of all tap water samples collected across MV. The isotopic values of tap waters collected across MV are categorized by water source. The isotopic values of tap waters are seen to cluster according to the water source. Linear regression relationship between $\delta^{18}O_{tap}$ and δ^2H_{tap} in all tap water samples from MV is shown as a solid line ($\delta^2H_{tap}=5.5\%_0\times\delta^{18}O_{tap}=21.2\%_0$ R² = 0.85, n = 118). The global meteoric water line (GMWL) is shown as a dashed line for comparison. The more negative slope of MV tap water, when compared to GMWL, suggests the influence of local physiographic factors on water isotope values. [Color figure can be viewed at wileyonlinelibrary.com]

for $\delta^{18}O_{tap}$ and $-97.5\%_{o} \pm 1.3$ for $\delta^{2}H_{tap}$. The mean and standard deviation of $\delta^{18}O_{tap}$ values for the Capilano watershed and Seymour watershed show similar values and thus to assess whether isotopic values of tap water from the Capilano watershed and the Seymour watershed were statistically significantly different from each other, independent *t*-tests were performed on the mean values of $\delta^{18}O_{tap}$. A comparison of isotopic values between the watersheds showed that there was a significant difference in mean $\delta^{18}O_{tap}$ value of Capilano water and Seymour; t (56) = 3.931, p < 0.01. Furthermore, to test for any significant

| TABLE 3—Range and mean \pm standard deviation of δ^{I} | ^{3}O and $\delta^{2}H$ of tap water samples for each water source. |
|---|---|
|---|---|

| | N | $\delta^{18}O_{tap}~(\%_{oo})$ | VSMOW | $\delta^2 H_{tap}$ (%) VSMOW | |
|-----------------------|-----|--------------------------------|-----------------|------------------------------|-----------------|
| Water Source | | Range | Mean ± SD | Range | Mean ± SD |
| Capilano watershed | 22 | −13.7 to −12.2 | -13.3 ± 0.4 | −98.1 to −93.4 | -95.6 ± 1.0 |
| Seymour watershed | 36 | -14.6 to -12.9 | -13.6 ± 0.4 | -98.8 to -93.5 | -97.5 ± 1.3 |
| Coquitlam watershed | 35 | -14.0 to -10.6 | -12.6 ± 0.6 | -98.4 to -78.0 | -90.0 ± 3.1 |
| *excluding outliers | 30 | -13.2 to -11.7 | -12.6 ± 0.4 | -91.3 to -88.4 | -90.0 ± 0.7 |
| Capilano/Seymour mix | 12 | -14.1 to -13.0 | -13.6 ± 0.3 | -97.8 to -95.4 | -96.9 ± 0.8 |
| Seymour/Coquitlam mix | 1 | -12.0 | -12.0 | -93.4 | -93.4 |
| Well | 12 | -14.3 to -9.3 | -11.7 ± 1.4 | -98.4 to -69.5 | -84.9 ± 8.2 |
| *excluding outliers | 7 | -11.5 to -10.7 | -11.1 ± 0.3 | -83.4 to -80.5 | -81.6 ± 1.0 |
| All watersheds | 106 | −14.6 to −10.6 | -13.2 ± 0.6 | −98.8 to −78.0 | -94.5 ± 3.8 |
| All MV water sources | 118 | -14.6 to -9.3 | -13.0 ± 0.9 | -98.8 to -69.5 | -93.5 ± 5.3 |

^{*}Outliers were defined as those samples with isotopic values that were not consistent with other samples from the same water source.

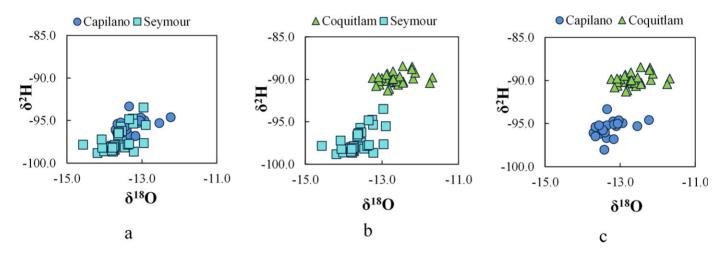


FIG 4—Comparison of $\delta^{18}O$ versus δ^2H of tap water samples from the three GVWD watersheds. The isotopic values of tap water samples that were sourced by the Capilano and Seymour watersheds show overlaps in its isotopic values (a), but are distinguishable from the tap water from the Coquitlam watershed (b, c). Wilk's λ test showed that the mean isotopic values between the major water sources were statistically significantly different from each other. Outliers have been excluded from this graph. [Color figure can be viewed at wileyonlinelibrary.com]

differences in mean isotopic values between the major water sources, Wilk's λ test (36) was conducted on mean values for both $\delta^{18}O_{tap}$ and δ^2H_{tap} . Results indicated that the combined $\delta^{18}O_{tap}$ and δ^2H_{tap} analysis of tap waters from each of the major sources—Capilano watershed, Seymour watershed, Coquitlam watersheds, well waters, Capilano/Seymour mixed, and Seymour/Coquitlam mixed were significantly different, F (10, 202), p<0.01; Wilk's $\lambda=0.041$, partial $\eta^2=0.80$.

Tap water samples sourced by the Coquitlam watershed were isotopically heavier than those samples sourced by the Seymour and Capilano watersheds. Mean values for the tap water samples from the Coquitlam watershed were $-12.6\%_{o}\pm0.6$ for $\delta^{18}O_{tap}$ and $-90.0\%_{o}\pm3.1$ for $\delta^{2}H_{tap}$ (Table 3). Mean values stayed unchanged for the Coquitlam watershed when the calculation was undertaken by excluding the five outlier samples; however, decreased the standard deviation to 0.4 for $\delta^{18}O_{tap}$ and 0.7 for $\delta^2 H_{tap}$. MV tap water samples from well water sources were the isotopically heavier than any of the tap water samples supplied by the MV watersheds and showed mean values of -11.7% ± 1.4 for $\delta^{18}O_{tap}$ and $-84.9\%_{oo} \pm 8.2$ for $\delta^{2}H_{tap}$. When the mean and standard deviation for tap waters sourced by well water were calculated with the exclusion of the five outlier samples, the values were -11.1% \pm 0.3 for $\delta^{18}O_{tap}$ and -81.6% \pm 1.0 for $\delta^{2}H_{tap}$. A graphical comparison of the isotopic data from the three major GVWD watersheds is shown in Fig. 4.

The Jenk's natural breaks method (37) was set to four classes and applied to generate the gradient maps for both $\delta^{18}O_{tap}$ and δ^2H_{tap} (Figs 5 and 6). The statistically optimal classification method was ideal as the number of classes could be defined to reflect the number of major water sources that supply MV. The natural breaks occurred at $-13.6\%_{\rm o}$, $-13.0\%_{\rm o}$, $-12.1\%_{\rm o}$, and $-9.3\%_{\rm o}$ for $\delta^{18}O_{tap}$ (Fig. 5) and at $-96.7\%_{\rm o}$, $-93.4\%_{\rm o}$, $-87.7\%_{\rm o}$, and $-69.5\%_{\rm o}$ for δ^2H_{tap} (Fig. 6).

70.6% of the group with the lightest $\delta^{18}O_{tap}$ values ($-14.6\%_{o}$ to $-13.6\%_{o}$) were supplied by the Seymour watershed, 11.8% were from the Capilano watershed, and the remaining 17.6% were from a mixture of Capilano and Seymour watersheds (Table 4). Similarly for δ^2H_{tap} values of the lightest group ($-98.8\%_{o}$ to $-96.7\%_{o}$), 69.2% of the samples came from the Seymour watershed, 7.7% from the Capilano watershed, and 23.1% from the mixture of both watersheds. Of the heaviest isotopic groups of $\delta^{18}O_{tap}$ ($-12.0\%_{o}$ to $-9.3\%_{o}$) and δ^2H_{tap} ($-87.6\%_{o}$ to $-69.5\%_{o}$), 70% and 100% were from a well water source, respectively.

OIPC

The mean isotopic values of tap water collected in Vancouver, Township of Langley, and Point Roberts, WA, and for the three major MV watersheds were compared with the



δ¹⁸O Samples

0 -14.6 - -13.6

-13.5 - -13.0

-12.9 - -12.1

-12.0 - -9.3

FIG. 5—Gradient map of $\delta^{18}O$ values of MV tap water samples. The isotopic values of oxygen were grouped according to the Jenk's natural breaks method, set to four classes. The isotopic values reflect the MV tap water distribution system (Fig. 1). [Color figure can be viewed at wileyonlinelibrary.com]

isotopic compositions of annual precipitation that were estimated using OIPC for each location. Figure 7 shows large offsets in isotopic values between OIPC values and actual tap water values. Mean tap water values for each location were more negative than OIPC values with Vancouver having the greatest difference of 2.6% for $\delta^{18}O$ and 15.6% for $\delta^{2}H$. This is mainly due to the OIPC generating its values through information pertaining to latitude, longitude, and elevation (17), without the consideration of source water providing to each area.

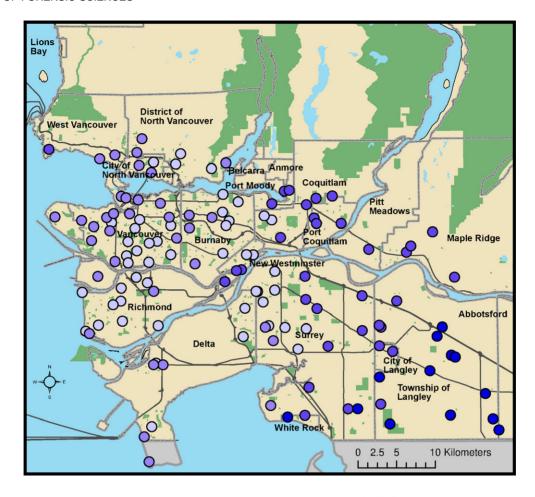
Discussion

This study successfully showed that isotopic variances exist within a single regional district of MV. MV tap water showed variances of 5.3% for $\delta^{18}O_{tap}$ and 29.3% for $\delta^{2}H_{tap}$, which is comparable to the spread observed for precipitation values across the entire UK at 4% for $\delta^{18}O_{ppt}$ and 30% for $\delta^{2}H_{ppt}$ (38,39). This confirms the importance of evaluating drinking water values with greater geographical resolutions. Furthermore, the

pattern of the isotopic values of both $\delta^{18}O_{tap}$ and δ^2H_{tap} of MV tap water samples closely followed the pattern of the tap water distribution system (Table 4).

A simple linear regression through the isotopic data for all collected MV tap water samples (including Point Roberts, WA) yielded a relationship of δ^2 H_{tap} = 5.5%, δ^{18} O_{tap} – 21.2%, R^2 = 0.85, n = 118 (Fig. 3). It is evident that the slope of the regression lines for MV tap water is more negative compared to the GMWL, suggesting the influence of local physiographic factors on water isotope values.

Non-MV tap water samples showed values that were more negative than the MV tap water samples. The lighter values are as expected for locations at higher latitudes (Table 1). The tap water sample from Abbotsford showed isotopic values that were similar to MV samples. The sample was taken from a regional park that is located geographically adjacent to the Township of Langley, and therefore, it is highly probable that the source of tap water was one of the aquifers underlying the Township of Langley. The isotopic values of tap water from Point Roberts, WA, were also similar to those of MV tap water samples.



δ²H Samples

- 98.8 -96.7
- 96.6 -93.4
- 93.3 -87.7
- -87.6 -69.5

FIG. 6—Gradient map of $\delta^2 H$ values of MV tap water samples. The isotopic values of hydrogen were grouped according to the Jenk's natural breaks method, set to four classes. The isotopic values reflect the MV tap water distribution system (Fig. 1). [Color figure can be viewed at wileyonlinelibrary.com]

According to the water source data given by GVWD, the tap water sample from Point Roberts, WA, was supplied by Seymour and thus explains the similarity in its value to MV samples.

According to OIPC, the three GVWD watersheds were expected to show similar isotopic values (Fig. 7). However, tap water samples sourced from these watersheds showed differences in their isotopic values. The observed variance in mean isotopic values of tap waters from the three GVWD watersheds may be due to differences in elevation. The Seymour watershed is located at approximately 218 m compared to that of Capilano at 145 m and Coquitlam at 152 m. However, elevation alone cannot explain the variance observed between the three watersheds, as Coquitlam showed the most positive values. There is generally a 0.1–0.5% depletion in δ^{18} O for every 100 m rise in altitude and a depletion of approximately 1–4% for δ^2 H (40,41). The differences between the isotopic values of tap water from Capilano and Seymour watersheds were 0.3% for δ^{18} Otap and 2.1% for δ^2 H_{tap},

which could be accounted for by the difference in elevation. However, mean isotopic values of tap water samples from Coquitlam showed a greater difference from Seymour values, pointing to an existence of an influential factor other than the elevation of the watersheds. Furthermore, mountain ranges surrounding the watersheds are similar across the three. Another possible explanation to the unexpected pattern of the isotopic values of MV tap water may be the effect of weather systems entering MV. The general direction of weather systems entering MV is from a southwesterly direction. Several physiographic factors can influence the isotopic compositions of water that is stored and collected in watersheds, which is then transported as tap water for consumption. The isotopic compositions of tap water are reflective of its source water, and thus, it is essential to gain an understanding of the isotopic compositions of that source water.

The comparison of MV tap water values to those estimated from OIPC showed large offsets for the three watersheds as well as for Vancouver and Point Roberts, WA (Fig. 7). The offset

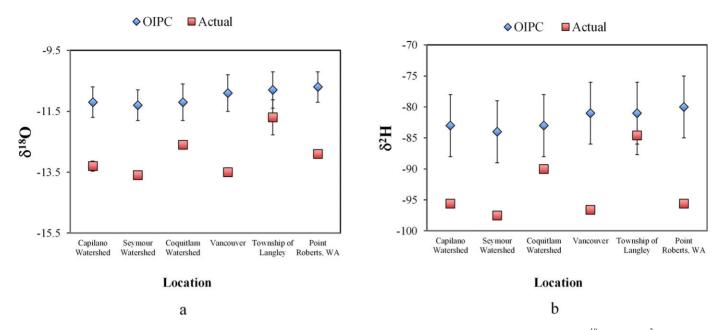


FIG. 7—Comparison of isotopic values of local precipitation and measured isotopic values of tap water. The comparison of (a) $\delta^{18}O$ and (b) $\delta^{2}H$ of isotopic values of local precipitation that were calculated from OIPC and mean tap water values from collected MV tap water samples, with 95% confidence intervals. Large offsets are observed for each location and suggest the need to better understand the isotopic compositions of tap water for each location at a high geographical resolution. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 4—The proportion of tap water samples supplied by each water source for the classes defined by the Jenk's natural breaks method.

| | $\delta^{18}O_{tap}$ (%) VSMOW | | | $\delta^2 H_{tap}$ (%) VSMOW | | | | |
|--------------------------|--|---|-------------------------------|--|--|-----------------------------|---------------------------------|-------------------------------|
| Water Source | $ \begin{array}{r} -14.6\%_{oo} \\ \text{to } -13.6\%_{oo} \\ (n = 34) \end{array} $ | $-13.5\%_{00}$ to $-13.0\%_{00}$ ($n = 38$) | -12.9% to $-12.1%$ $(n = 26)$ | $-12.0\%_{oo}$ to $-9.3\%_{oo}$ ($n = 10$) | $ \begin{array}{r} -98.8\%_{oo} \\ \text{to } -96.7\%_{oo} \\ (n = 34) \end{array} $ | -96.6% to $-93.4%$ (n = 38) | -93.3% to $-87.7%$ ($n = 26$) | -87.6% to $-69.5%$ $(n = 10)$ |
| Capilano | 0.118 | 0.395 | 0.115 | 0 | 0.077 | 0.613 | 0 | 0 |
| Capilano/Seymour mix | 0.176 | 0.158 | 0 | 0 | 0.231 | 0.097 | 0 | 0 |
| Seymour | 0.706 | 0.289 | 0.038 | 0 | 0.692 | 0.290 | 0 | 0 |
| Seymour/Coquitlam mix | 0 | 0 | 0 | 0.1 | 0 | 0 | 0.032 | 0 |
| Coquitlam | 0 | 0.158 | 0.846 | 0.2 | 0 | 0 | 0.968 | 0 |
| Well | 0 | 0 | 0 | 0.7 | 0 | 0 | 0 | 1 |
| Total | 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 |

was at 2.6‰ for $\delta^{18}O$ and 15.6‰ for $\delta^{2}H$ for Vancouver. This offset may be explained by tap water being sourced from watersheds located north of the municipality. Mean isotopic values of Township of Langley tap water showed the least difference with OIPC values at 0.9‰ for $\delta^{18}O$ and 3.6‰ for $\delta^{2}H$ and fell within the OIPC 95% confidence interval. This may be due to the underlying aquifers supplying much of the Township of Langley tap water. The GVWD watersheds also showed offsets from OIPC values.

While the OIPC can provide a good estimate of the isotopic compositions of local precipitation, it is necessary to investigate and understand the isotopic compositions of water sources that are supplying tap water to each city. For example, OIPC gives Point Roberts, WA, values of $-10.8\%_{\rm o} \pm 0.5\%_{\rm o}$ for $\delta^{18}O_{\rm ppt}$ and $-81\%_{\rm o} \pm 5\%_{\rm o}$ for $\delta^{2}H_{\rm ppt}$. The tap water sampled from this area showed $-12.9\%_{\rm o}$ and $-95.6\%_{\rm o}$ for $\delta^{18}O_{\rm tap}$ and $\delta^{2}H_{\rm tap}$, respectively. If human tissue samples were compared against OIPC values for geolocation purposes, the offset could lead to erroneous conclusions on the origin of the individual in question,

assuming the conversion equations (4,5,8,9,16), in themselves, are valid (42–47).

It is important to study the isotopic values of tap waters on a city scale as tap waters that are provided to cities are related to, but not identical to, local precipitation values. This is because tap water is collected and stored in reservoirs for distribution. Tap water may be distributed to places distantly located from those reservoirs in which tap water is drawn from, as drinking water is distributed via a network of pipes carrying tap water to residents in different communities. With the expansion of cities, drinking water must be strategically brought into a city to satisfy the growing need. It is also possible for geographically distinct communities to share water from the same reservoir. Offsets in isotopic composition between tap water and local precipitation water can exist, and therefore, the cities do not lend themselves to the OIPC.

Tap waters that are provided to cities can also be supplied by a single reservoir and can be composed of a single isotopic value. While a regional variance in isotopic values will allow tap water to be sourced to specific areas within a regional district, a single regional value can also be of significance as that value can be assigned to an entire regional district. Furthermore, although bottled water consumption has also risen in the recent decade (48–52) tap water is still the preferred and more accessible source of human drinking water. This study demonstrated that isotopic differences of tap water are observed across a regional district that is supplied by multiple water sources. There can be a significant difference in isotopic values of tap water across a geographical area, and therefore, a higher geographical resolution study than that of Bowen et al. (19) and Kennedy et al. (20) will allow for a better understanding of the isotopic variance within a single city.

Conclusion

While tap water can be linked to local precipitation water, the isotopic values can and do differ from each other, especially in areas where tap water is drawn from numerous water sources. These tap waters can be sourced from physiographically distant areas in which it supplies. When investigating the link between the isotopic compositions of human tissues and drinking water, it is vital to gain an understanding of the isotopic compositions of tap water on a city scale.

MV tap water is supplied by multiple water sources with three major GVWD watersheds supplying much of the regional district and a smaller percentage supplied by groundwater aquifers. $\delta^{18}O$ and δ^2H in tap water varied across MV with a range of 5.3‰ for $\delta^{18}O_{tap}$ and 29.3‰ for δ^2H_{tap} . It is thus not appropriate to assume that low-resolution sampling will provide an accurate assessment of a single city's isotopic range. It is evident from this study that increased sampling of "known source" city waters is important to establish the "actual" range for that city, and by extension, for human geolocation.

The isotopic compositions of tap water are influenced by local precipitation; however, offsets are observed between the estimated precipitation values and true tap water values on a small geographical scale. Thus, regardless of the geographical locale of a city or district, the focus should be on understanding the tap water distribution systems and determining the isotopic compositions of the water sources. It is vital to first gain an understanding of the isotopic patterns of $\delta^{18}O$ and δ^2H in tap waters on a city scale to establish the characteristic range for that single city and further to expand this approach to other cities so that a city-to-city isotopic map can be established. With this type of information, more precise geolocation of human tissues may be made.

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Additional information and reprint requests:

Lynne S. Bell School of Criminology Simon Fraser University 8888 University Dr. Burnaby, BC V5A 1S6 Canada

E-mail: lynneb@sfu.ca