PLOS WATER

RESEARCH ARTICLE

Spatial variations in tap water isotopes across Canada: Tracing water from precipitation to distribution and assess regional water resources

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Abstract

With global warming and increasing water use, tap water resources need sustainable management. We used hydrogen and oxygen isotope analyses in tap water (i.e., δ^2H and $\delta^{18}O$ values) to identify issues associated with tap water resources in Canada. We analyzed 576 summer tap samples collected from across Canada and 76 tap samples from three cities during different seasons and years. We classified the samples based on their sources: groundwater ($Tap_{Groundwater}$), river (Tap_{River}) and lake (Tap_{Lake}). δ^2H values in tap water correlate strongly with values predicted for local precipitation across Canada with a stronger correlation for $Tap_{Groundwater}$ and Tap_{River} than for $Tap_{Lake.}$ We then constructed water balance models to predict the δ^2H of surface water across Canada, and validated them against Canadian stream δ^2H data. δ^2H values in tap water correlate strongly with values predicted for local surface water, however, the water balance models improved the predictability only for Tap_{River} and Tap_{Lake} and not for $Tap_{Groundwater}$. $Tap_{Groundwater}$ δ^2H values reflect the δ^2H values of annually averaged precipitation, whereas Tap_{River} and Tap_{Lake} δ^2H values reflect post-precipitation processes. We used the δ^2H residuals between the observed and predicted δ^2H values to assess regional processes influencing tap water δ^2H values across Canada. Regionally, snow/glacier melt contributes to all tap sources around the Rockies. Tap waters are highly evaporated across Western Canada, irrespective of their sources. In the Great Lakes and East Coast regions, tap waters are evaporated in many localities, particularly those using surface reservoirs and lakes. We propose the use of these isotopic baselines as a way forward for the monitoring of tap water resources at different scales. These isotopic baselines also have valuable applications in human forensic studies in Canada.

Citation: Bhuiyan SA, Jameel Y, Chartrand MMG, St-Jean G, Gibson J, Bataille CP (2023) Spatial variations in tap water isotopes across Canada: Tracing water from precipitation to distribution and assess regional water resources. PLOS Water 2(1): e0000068. https://doi.org/10.1371/journal.pwat.0000068

Editor: Rodolfo Nóbrega, University of Bristol, UNITED KINGDOM

Received: May 8, 2022

Accepted: December 19, 2022

Published: January 24, 2023

Data Availability Statement: All data to verify the conclusions of this work have been made available. Data A and B in S1 Text are available at https://doi.org/10.6084/m9.figshare.19243518. The data used for water balance modelling is open-access and available online at Waterisotopes.org (https://wateriso.utah.edu/waterisotopes/index.html), HydroSHEDS (https://www.hydrosheds.org/) and Physical Sciences Laboratory (https://psl.noaa.gov/data/gridded/data.narr.monolevel.html#plot) websites. Canadian streams isotope data that were used for models validations can be requested from Dr. John Gibson (jjgibson@uvic.ca) at University of Victoria.

Funding: CPB and SAB acknowledge funding from Canadian Security and Safety Program Targeted Investment (CSSP-2018-TI-2385). GSJ and MMGC acknowledge funding from the Chemical, Biological, Radiological and Nuclear Research & Technology Initiative (CRTI 08-0116RD). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

Competing interests: All authors declare that they have no conflicts of interest.

1. Introduction

Long term sustainability of water resources has become a concern in Canada due to the combination of rapid ongoing global climate change across the country and fragmented water governance [1, 2]. Although Canada is a water rich country, most of its freshwater flows north into the Arctic Ocean and is not accessible to the majority of Canadians who live in southern Canada [3]. Canada's climate and water abundance varies from region to region, for example, the coastal regions are wet throughout the year whereas the Prairies are vulnerable to droughts due to continental semi-arid conditions. Some Canadian regions, particularly the Prairies and southern Ontario, have already experienced serious water availability threats [3]. Warming, reduced snow cover and glacier retreat from the Rockies will continue to impact water availability and supply across the Prairies [4]. A recent study in the continental Nelson River basin (MB) suggested that aquifer recharge in this region is dependent on winter precipitation and snow melt, and is therefore vulnerable to regional changes in winter water balance [5]. Southern Ontario is experiencing freshwater availability threats due to contamination, overconsumption and climate change and this water stress is predicted to worsen without immediate actions to protect the freshwater resources (Streams, rivers, lakes, and groundwater) [6]. In addition to these natural threats to water availability, Canadian water management practices vary between localities and provinces. Some regions preferentially use and store water in lakes (e.g., large cities and Eastern Canada) whereas others pump water directly from large rivers and groundwater (e.g., Prairies). These different practices have different sensitivity to climate change and require a regional monitoring of tap water resources.

Stable hydrogen and oxygen delta measurements (herein expressed as δ^2H and δ^{18}O values, respectively) are powerful tracers of water cycling processes. Global patterns in the isotopic composition of precipitation follow climatic and geographic patterns including meridional water transport, continentality, elevation, temperature and relative humidity variations [7–11]. Environmental water resources inherit their isotopic composition and spatiotemporal variations primarily from modern precipitation [12–15]. However, water in human-managed distribution networks might not follow these natural variations, for example, due to evaporative loss while residing in reservoirs, mixing or switching between multiple water sources, and importation of non-local water [16–18]. Therefore, isotopic investigation of tap water is useful to identify water origin, risks at source level, water supply management issues and climatic vulnerability of critical water resources used for public water supply [19–23].

In a pioneering study, Bowen et al. [19] used δ^2H and δ^{18}O values of tap water to trace regional hydrological processes and to characterize regional water issues across the contiguous United States. Since then, tap water δ^2H and δ^{18}O analyses have been successfully applied to investigate tap water across the globe including for partitioning regional and seasonal reliance on surface water and groundwater, for identifying regions extracting fossil groundwater, or for estimating the quantity of imported water from inter-basin transfer [16, 21, 23, 24]. At the scale of a city (e.g., Western USA), Jameel et al. [25, 26] and Tipple et al. [18] used tap water isotopic composition to capture district level differences in water management practices, to provide independent validation of flow within the water distribution system, and to quantify water losses due to evaporation in urban water systems.

Here we present the first Canadian national-level isotopic analysis of tap water, based on samples collected from across Canada. We document the main supply sources of these tap water samples using publicly available records. First, we analyzed the tap water δ^2H, δ^{18}O and *d-excess* patterns over Canada. Then we compared tap water δ^2H values with predicted local precipitation δ^2H values [27]. Then we constructed a series of water balance models to predict δ^2H values in surface water across Canada and we validated these models by using an existing dataset of δ^2H values in streams [28], and compared the tap water δ^2H values with those of predicted local surface water. Finally, we analyzed the residual δ^2H values between tap water and local precipitation, and residual δ^2H values between tap water and local surface water. Using all the data analysis, we assessed regional hydrological processes, vulnerability to ongoing climate warming and potential water management issues. Our analysis offers a baseline for nationwide monitoring of water resources in Canada [19, 24, 29]. We also underline the value of these tap water and surface water databases for human forensic applications across Canada, as previously established for other regions [30].

2. Materials and methods

2.1 Tap water samples collection

We collected a total of 579 tap water summer samples from across Canada covering 425 cities and towns over a 4-year period (2008 to 2011) (Data A in S1 Text) and removed 3 samples prior to analysis due to accidental leakage of water. We selected tap water sites that were easily accessible within southern Canada and covering the most populous centres as well as agricultural regions where water demand is the greatest. We also sampled a few time-series collecting tap water seasonally at several sites of three major metropolitan areas for several years–Ottawa (27 samples, 2008–2012, 5 sites), Montreal (19 samples, 2008–2010, 7 sites) and Sudbury (30 samples, 2008–2011, 7 sites) (Data B in S1 Text). At each tap water sampling site, we recorded the latitude, longitude and altitude. Prior to sampling in a 50 mL centrifuge tube (Sarstedt, Montreal, Canada), the tap was run for 10 seconds, the tube was filled, then capped.

At each site, we recorded the main source of each of the tap water samples by asking the local residents and/or municipality, and based on this information, classified the sources as groundwater ($\text{Tap}_{\text{Groundwater}}$), river ($\text{Tap}_{\text{River}}$) and lake ($\text{Tap}_{\text{Lake}}$). $\text{Tap}_{\text{Groundwater}}$ is defined as tap water sourced from wells. $\text{Tap}_{\text{River}}$ is defined as tap water sourced from streams and rivers. Tap_{Lake} is defined as tap water sourced from small or large lakes, pounds and artificial reservoirs. When the information was available, we also recorded more details about the name of the rivers and the name of the lakes from which the tap water was sourced at each site. Data A and B in S1 Text including all the information related to this classification is available at https://doi.org/10.6084/m9.figshare.19243518.

2.2 Tap Water δ^2H and δ^{18}O analysis and traceability to the VSMOW scale

We analyzed all water samples at the Ján Veizer Stable Isotope Laboratory at the University of Ottawa. Prior to isotope analysis, we added a piece of Cu (to remove any S species) and a few grains of activated charcoal (to remove any organics) to the water sample vials at least 24 hours prior to isotopic analysis. For δ^{18}O analysis, we pipetted a 200 μL aliquot of the sample water into an exetainer vial and capped with a gas-tight cap. The headspace of the exetainer vial was flushed with 2% CO_2 in He for 4 minutes, then stored on the bench to equilibrate for 24 hours. We then placed the exetainers in a 25˚C heating block, allowed them to equilibrate, and the CO_2 gas was analyzed for δ^{18}O using a GasBench II (Thermofisher, Bremen, Germany) with a Delta$^+$XP isotope ratio mass spectrometry (IRMS; Thermofisher, Bremen, Germany). For δ^2H analysis, a piece of hokko platinum catalyst, along with 200 μL aliquot of the sample water, was added into the exetainer and capped. The headspace was flushed with 2% H_2 in He for 4 minutes, and left on the bench to equilibrate for at least 2 hours. The exetainers were then placed in a 25˚C heating block, allowed to equilibrate, and the H_2 gas was analyzed for δ^2H using the same GasBench II with a Delta+XP IRMS as for δ^{18}O. Several replicates of three internal water reference materials (RMs) were included in each analysis sequence: W-7 ($\delta^2\text{H} = -198.5 \pm 2.0$ ‰ and $\delta^{18}\text{O} = -24.55 \pm 0.2$ ‰), W-10 ($\delta^2\text{H} = -85.9 \pm 2.0$ ‰ and $\delta^{18}\text{O} = -11.84 \pm 0.2$ ‰) and

W-9 (δ^2H = +11.3 ± 2.0 ‰ and δ^{18}O = −5.06 ± 0.2 ‰). These internal water RMs are traceable to the VSMOW scale via calibration against VSMOW (δ^2H = 0 ‰, δ^{18}O = 0 ‰ [31]), GISP (δ^2H = −189.5 ± 1.2 ‰, δ^{18}O = −24.76 ± 0.09 ‰ [32]); and SLAP (δ^2H = −428 ‰, δ^{18}O = −55.5 ‰ [31]). Tap water δ^2H and δ^{18}O values were processed using the LIMS software [33]. A water QC material, W-20 (δ^2H = −5.9 ± 2.0 ‰ and δ^{18}O = −7.34 ± 0.2 ‰), was also included in every analysis sequence. The analytical precision (2σ) of the δ^2H and δ^{18}O analyses, based on long-term replicate measurements of W-20 at the University of Ottawa is better ± 2.0‰ and ± 0.2 ‰, respectively. All water samples were analyzed once, and 10% of the samples were analyzed in duplicate, with the standard deviation of δ^2H and δ^{18}O replicates less than ± 2.0‰ and ± 0.2 ‰, respectively.

2.3 Spatial patterns of tap water isotopes and comparison with predicted precipitation δ^2H values

To analyze the spatial variability of tap water isotopes across Canada, we used ESRI ArcGIS Pro to map the δ^2H and *d-excess* values (d, where d = δ^{18}O - 8* δ^2H; a more negative (i.e. lower) *d-excess* is an indicator for post-precipitation isotopic fractionation due to evaporative water loss [7]). Since tap water δ^2H and δ^{18}O values show very similar patterns (Figs 2 and 5 and Figs A, B in S1 Text), we focused our interpretations on δ^2H values and *d-excess* values. Using the latitude and longitude of each tap water site, seasonal and annual predicted precipitation δ^2H values were extracted from the map of Bowen [27]. Comparison of δ^2H values in tap water to those in precipitation can provide insights into how water is cycled from its local precipitation source to the consumer faucet (e.g., Bowen et al. [19]). One limitation, however, is that the precipitation isotopes models can sometimes be less accurate where sampling density of precipitation isotopes is low. For example, in North America, the predicted isotopic values in precipitation along the Pacific coast do not represent accurately the isotopic gradient from coast toward inland locations [19, 34].

2.4 Water balance modelling to predict surface water δ^2H values

In an effort to further understand water cycle processes along the water supply chain, we constructed a series of water balance models to predict δ^2H values in surface water across Canada. Unlike the precipitation model that only accounts for atmospheric controls of isotopic variability, water balance models integrate post-precipitation modifications associated with surface hydrology, for example, mixing of isotopically distinct waters from different sources (e.g., snowmelt or groundwater), evaporative losses and losses due to transpiration or infiltration.

Four datasets were used for the water balance modelling: 1) long-term monthly mean isotopic values for global precipitation [27]; 2) North American flow direction [35]; 3) long-term monthly mean of daily total precipitation [36] and 4) long-term monthly mean evapotranspiration [36]. We followed a similar approach to Bowen et al. [20] to predict the δ^2H variability in surface water across Canada. Briefly, we calculated discharge (Q) and isotopic flux associated with discharge (δQ) from each grid cell at 1km^2 resolution using the equations in Table 1 within the North America boundary defined by the HydroSHEDS dataset (Fig 1). We accumulated upstream accumulated Runoff Q and accumulated Runoff δQ using the digital topography map with drainage direction from HydroSHEDS and the "Flow Accumulation" tool (Spatial Analyst Toolbox; ESRI ArcGIS). The downstream surface water isotopic values were calculated as accumulated Runoff δQ divided by accumulated Runoff Q.

In addition to the annual water balance models by Bowen et al. [20], we built summer water balance models (Table 1) to match with our tap water collection season. We built the annual and summer models using two different approaches: (1) by propagating the δ^2H values in

Table 1. Equations used to calculate discharge and isotopic flux at 1km^2 grid cell to be accumulated downstream. P = precipitation; ET = evapotranspiration; δP = isotopic composition of precipitation; Q = discharge, and δQ = isotopic flux associated with discharge.

ID	Discharge	Isotopic flux (Discharge * δ^{2}H)
1. Monthly Weighted Annual Model	Q = Jan P +. . ..+ Dec P	δQ = (Jan P * Jan δP) +. . .. + (Dec P * Dec δP)
2. Monthly Weighted Summer Model	Q = May P +. . .. + Oct P	δQ = (May P * May δP) +. . .. + (Oct P * Oct δP)
3. Monthly Weighted Annual ET Model	Q = Jan (P—ET) +. . .. + Dec (P-ET)	δQ = (Jan (P—ET) * Jan δP) +. . .. + (Dec (P–ET) * Dec δP)
4. Monthly Weighted Summer ET Model	Q = May (P—ET) +. . .. + Oct (P-ET)	δQ = (May (P—ET) * May δP) +. . .. + (Oct (P-ET) * Oct δP)
5. Annual average Model	Q = Jan P +. . ..+ Dec P	δQ = total annual (P) * annual average δP
6. Annual average ET Model	Q = total annual P—total annual ET	δQ = (total annual P—total annual ET) * annual average δP

https://doi.org/10.1371/journal.pwat.0000068.t001

precipitation weighted by total precipitation (P), or (2) by propagating δ^2H values in precipitation weighted by effective precipitation (P-ET), which aimed to quantify whether accounting for spatial evapotranspiration (ET) variations improved the estimated predicted δ^2H values in surface water. In total we built six water balance models (Table 1).

2.5 Validation of the six water balance models: Comparison between observed δ^2H values in streams and predicted surface water δ^2H values

To validate our approach, we first compared our predicted local surface water isotopic values with a dataset of δ^2H values in Canadian streams [28]. However, the latitude and longitude of the streamflow samples collected in Gibson et al. [28] did not always line-up with the geometry of the HydroSHEDS river network. In other words, uncertainties of the HydroSHEDS river network often lead real sampling points to fall on pixels that are outside of the actual sampled stream. In order to resolve those spatial mismatches and to properly compare the predicted

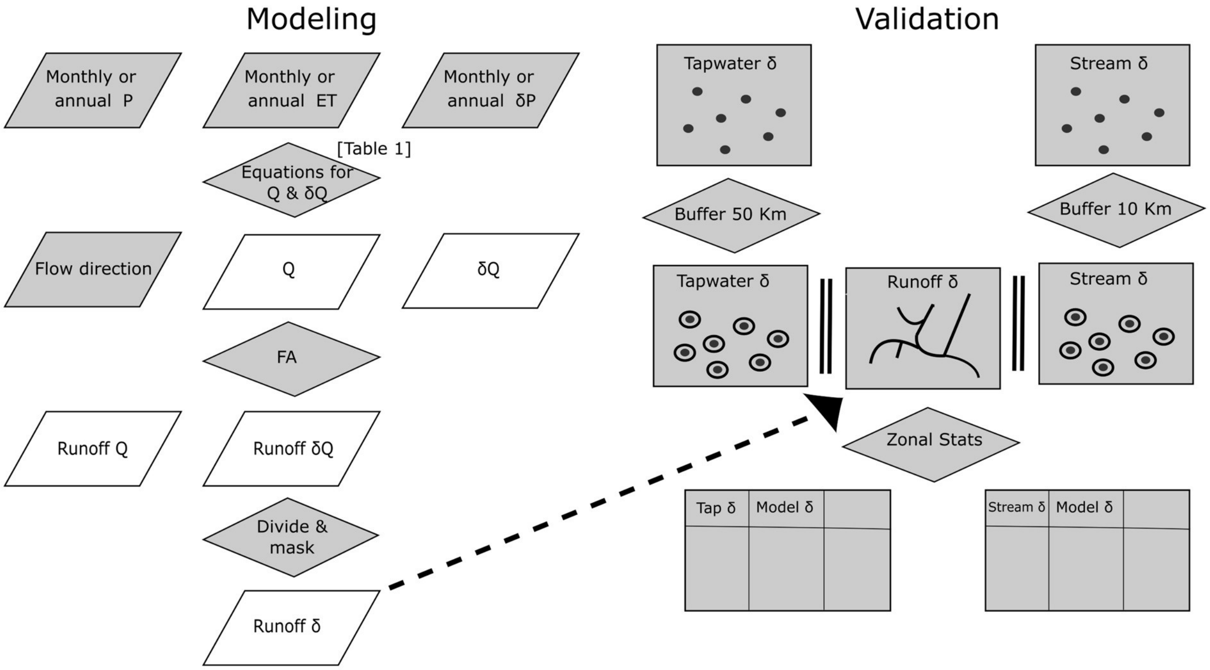

Fig 1. Workflow for GIS based water balance modeling and validation modified from Bowen et al. [20]. Diamond = operations and rectangular (shaded) = input raster data sets. P = precipitation; ET = evapotranspiration; δP = isotopic composition of precipitation; FA = flow accumulation; Q = discharge, and δQ = isotopic flux associated with discharge.

https://doi.org/10.1371/journal.pwat.0000068.g001

δ^2H values in surface water derived from our models with the observed δ^2H values in streams from Gibson et al. [28], we applied two GIS processing steps following Bowen et al. [20]. First, we masked all the pixels of our models for which the HydroSHEDS flow accumulation was lower than 9 km^2 to exclude small streams because only large streams were sampled in Gibson et al. [28] (Fig 1). We then extracted the predicted δ^2H values in local surface water from these masked models (i.e., with only large streams represented) at each stream sample site of Gibson et al. [28] by calculating the flux weighted average δ^2H values predicted by our models for a 10 km radius circular zone around each stream sample site (Fig 1). We then compared our annual models with the observed annual average δ^2H values in streams ($Stream_{Annual}$) δ^2H values at 262 sites and our summer models with the observed summer average δ^2H values in streams ($Stream_{Summer}$) δ^2H values at 241 sites [28]. All the models were validated using linear regression models between the observed δ^2H values in streams [28] and the predicted δ^2H values in local surface water from our models.

2.6 Comparison between observed tap water δ^2H values and predicted local surface water δ^2H values

We then compared the observed δ^2H values in tap water to the predicted δ^2H values in local surface water derived from our models. The approach was similar to that described in Section 2.5, but with a larger 50 km radius circular buffer around each of the tap water sampling sites (Fig 1). A larger radius was necessary as the exact source of tap water was not always easy to locate, and some large cities (e.g., Vancouver, Calgary) use more distant reservoirs as their main tap water sources. We evaluated the correlation between the observed δ^2H values in tap water and the predicted δ^2H values in local surface water from our models.

2.7 δ^2H residuals analysis

We extracted and mapped the residuals between the observed δ^2H values in tap water and predicted δ^2H values in local annual precipitation (monthly weighted). To explore if natural and anthropogenic processes impose potential threats to tap water sources at a regional level, we analyzed residuals between the observed δ^2H values in tap water and predicted δ^2H values in local annual surface water based on our Monthly Weighted Annual ET Model.

3. Results

3.1 Spatial patterns of δ^2H observations in Canadian tap water

δ^2H values in Canadian tap water range from -188‰ to -33‰ (Fig 2). There are strong spatial patterns of increasingly more negative δ^2H values from low latitude coastal regions towards high latitude and high-altitude inland regions (Fig 2). Generally, the most negative δ^2H values were observed in Western Canada (mountainous regions) and the most positive δ^2H values in the Eastern Canada's coastal and Great Lakes regions, irrespective of tap water sources (Fig 2). The *d-excess* values of tap water also show large spatial variability, ranging from -35.3‰ to +19.1‰ (Fig 3). The general patterns show more positive *d-excess* values dominate across the Prairies (Alberta, Saskatchewan and Manitoba) and British Columbia, whereas the East Coast regions (New Brunswick, Nova Scotia, Prince Edward Island and Newfoundland) are dominated by more negative *d-excess* values, irrespective of tap water sources (Fig 3). In contrast, the Great Lakes regions (Ontario and Quebec) show an interesting combination of high and low *d-excess* values, mainly for $Tap_{Groundwater}$ and Tap_{Lake} respectively (Fig 3).

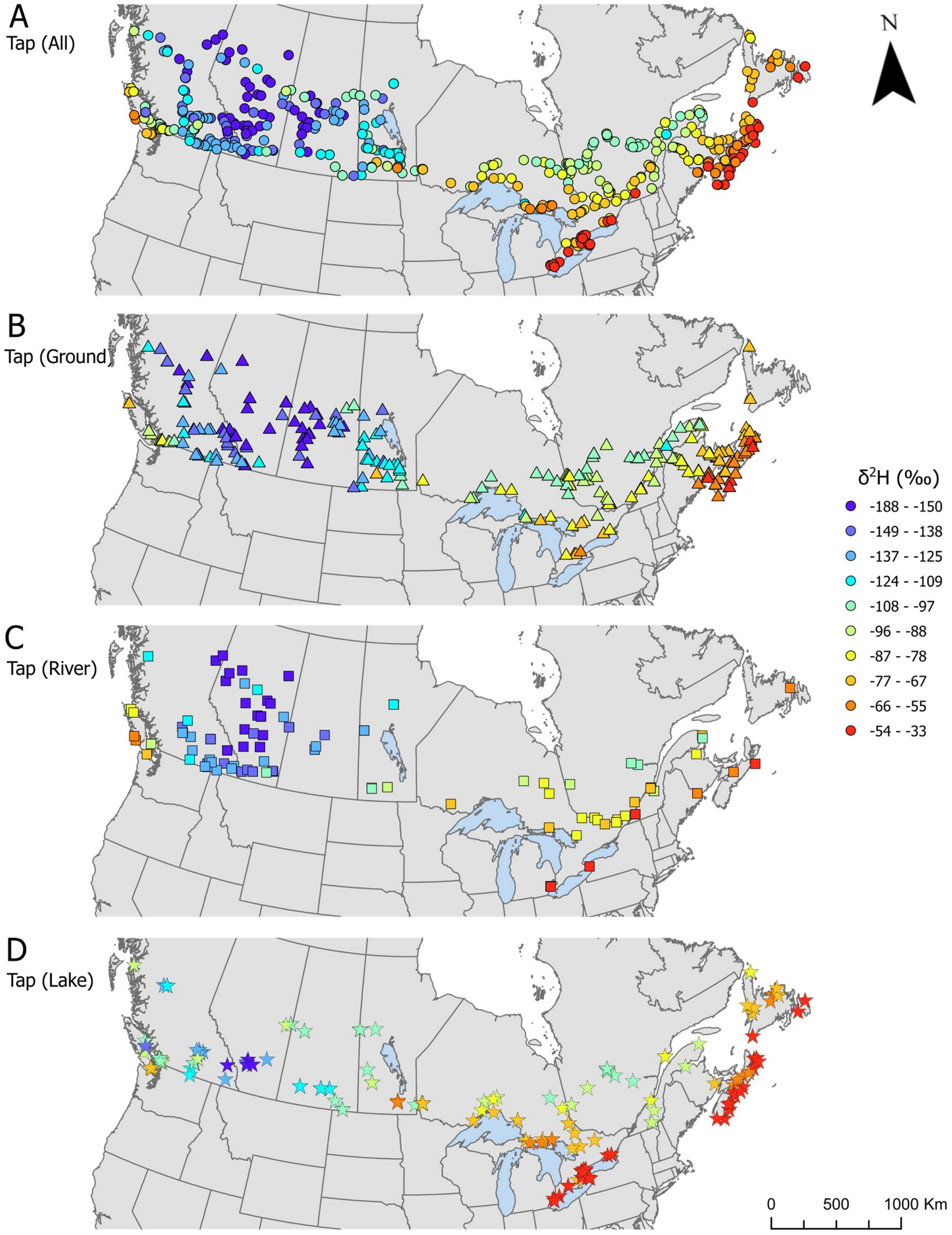

Fig 2. Spatial distribution of sample locations and δ^2H values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n = 281), c: tap water sourced from rivers (n = 118) and d: tap water sourced from lakes (n = 177). Classification between groundwater, river, and lake is detailed in the Methods. Administrative boundaries are from http://www.naturalearthdata.com/. This map was generated in ESRI ArcGIS Pro.

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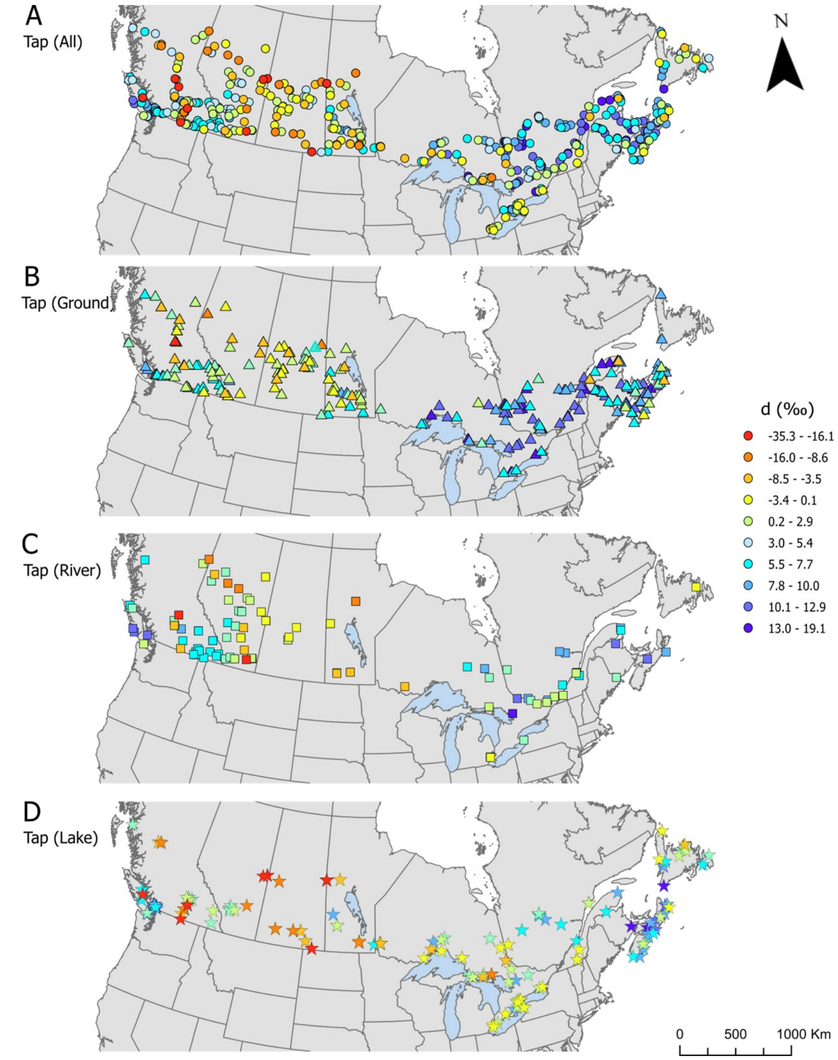

Fig 3. Spatial distribution of sample locations and *d-excess* (d) values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n = 281), c: tap water sourced from rivers (n = 118) and d: tap water sourced from lakes (n = 177). Classification between groundwater, river, and lake is detailed in the Methods. Administrative boundaries are from http://www.naturalearthdata.com/.

https://doi.org/10.1371/journal.pwat.0000068.g003

3.2 Relationship between tap water δ^2H values and predicted precipitation δ^2H values

The δ^2H and δ^{18}O values of tap water samples generally follows the Canadian Meteoric Water Line (CMWL) (Fig 4) [28]. However, ~27% of the samples fall below the CMWL, indicating isotopic fractionation from evaporation. There is a strong positive correlation between the observed δ^2H values in tap water and the predicted δ^2H values in local precipitation irrespective of tap water sources and seasonality of precipitation (Table 2 and Fig 5 and Fig C in S1 Text). Plotting the δ^2H values in tap water grouped by their pre-classified water sources shows δ^2H values of $\text{Tap}_{\text{Groundwater}}$ and $\text{Tap}_{\text{River}}$ have a much higher correlation with local precipitation than δ^2H values of Tap_{Lake}, both annually and seasonally (Table 2 and Fig 5 and Fig C in S1 Text). When accounting for seasonal precipitation, δ^2H values of Tap_{Lake} have a stronger correlation with summer precipitation, yet, they remain much less predictable relative to other

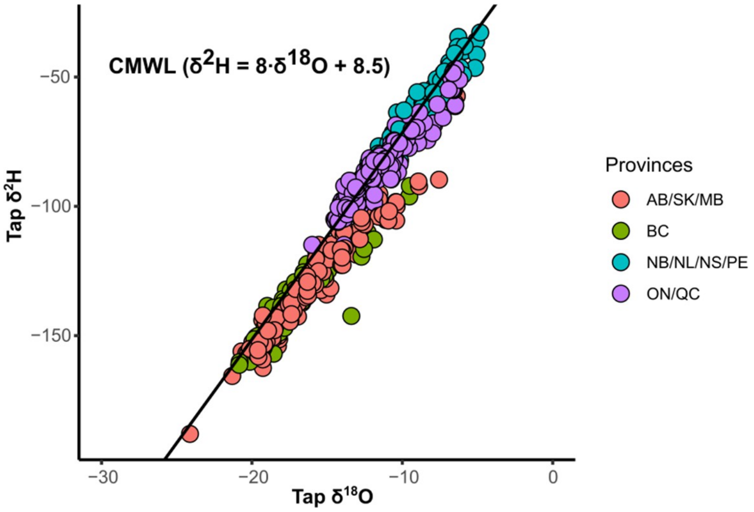

Fig 4. Covariation of tap water δ^2H and δ^{18}O values (n = 576) in relation to Canadian meteoric water line (CMWL) [28].

https://doi.org/10.1371/journal.pwat.0000068.g004

sources (groundwater and river). The correlation between δ^2H values of tap water and predicted winter precipitation is weaker than summer and annual precipitation, irrespective of the tap water source types (Table 2).

3.3 Validation of the six water balance models: Relationship between observed δ^2H values in streams and predicted surface water δ^2H values

There is a strong positive correlation between the observed δ^2H values in streams from Gibson et al. [28] (Stream$_{\text{Annual}}$ and Stream$_{\text{Summer}}$) and predicted δ^2H values in surface water, for both our annual and summer models (Fig F in S1 Text) validating our water balance modelling approach. With the exception of the semi-arid regions of Alberta, Saskatchewan and Manitoba (the Prairies), where the Stream$_{\text{Annual}}$ and Stream$_{\text{Summer}}$ are consistently more positive than the predicted δ^2H values (Fig F in S1 Text), the monthly weighted annual models perform better (i.e., closer to 1:1 line) than annual average models and summer models (Fig F in S1 Text).

3.4 Relationship between observed tap water δ^2H values and predicted local surface water δ^2H values

There is a strong positive correlation between the observed δ^2H values in tap water and predicted δ^2H values in local surface water, irrespective of tap water sources (Table 3 and Fig 6 and Fig D in S1 Text). Plotting the δ^2H values in tap water grouped by their pre-classified

Table 2. Results of linear correlation model between tap water δ^2H values and local precipitation δ^2H values.

Tap sources	Monthly Weighted Annual precipitation	Monthly Weighted Summer precipitation	Monthly Weighted Winter precipitation
	R^2	R^2	R^2
All	0.79	0.81	0.69
Tap$_{\text{Groundwater}}$	0.86	0.87	0.79
Tap$_{\text{River}}$	0.86	0.88	0.76
Tap$_{\text{Lake}}$	0.62	0.67	0.45

* For all correlations, the p value is $<2.2^{*}e^{-16}$. P-values are calculated using the T-test

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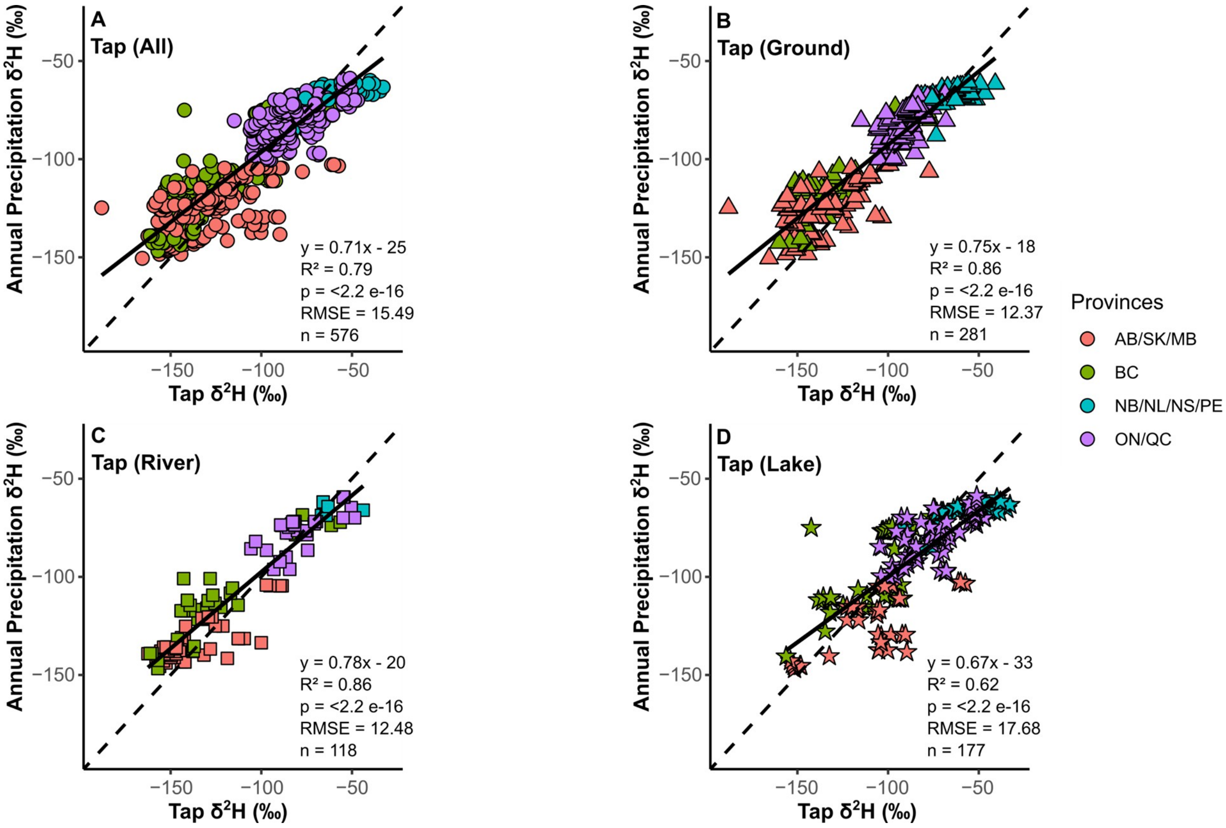

Fig 5. Correlation between tap water δ^2H values and monthly weighted local predicted annual precipitation δ^2H values. a: all the tap water samples combined, b: tap water sourced from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1 line. The black line represents the best fit linear model.

https://doi.org/10.1371/journal.pwat.0000068.g005

water sources shows the water balance models do not improve $\mathrm{Tap}_{\mathrm{Groundwater}}$ δ^2H prediction (Table 3 and Fig 6) relative to the precipitation-only model (Table 2 and Fig 5). Conversely, the water balance models do improve the prediction of $\mathrm{Tap}_{\mathrm{River}}$ δ^2H and $\mathrm{Tap}_{\mathrm{Lake}}$ δ^2H relative to the precipitation-only model. The monthly weighted annual models show the best prediction of δ^2H values in tap water (i.e., best metrics and closest to the 1:1 line).

3.5 δ^2H Residuals in Canadian tap water

We present the residual δ^2H values between tap water and local annual precipitation (monthly weighted) (Fig E in S1 Text), and the residual δ^2H values between tap water and

Table 3. Results of linear correlation model between tap water δ^2H values and predicted surface water δ^2H values.

Tap sources	Monthly Weighted Annual Model	Monthly Weighted Summer Model	Monthly Weighted Annual ET Model	Monthly Weighted Summer ET Model	Annual Average Model	Annual Average ET Model
	R^2	R^2	R^2	R^2	R^2	R^2
All	0.81	0.81	0.81	0.81	0.78	0.78
$\mathrm{Tap}_{\mathrm{Groundwater}}$	0.84	0.84	0.84	0.84	0.84	0.84
$\mathrm{Tap}_{\mathrm{River}}$	0.89	0.90	0.90	0.90	0.85	0.85
$\mathrm{Tap}_{\mathrm{Lake}}$	0.73	0.74	0.73	0.73	0.65	0.65

* For all correlations the p value is $<2.2^{*}e^{-16}$. P-values are calculated using the T test

https://doi.org/10.1371/journal.pwat.0000068.t003

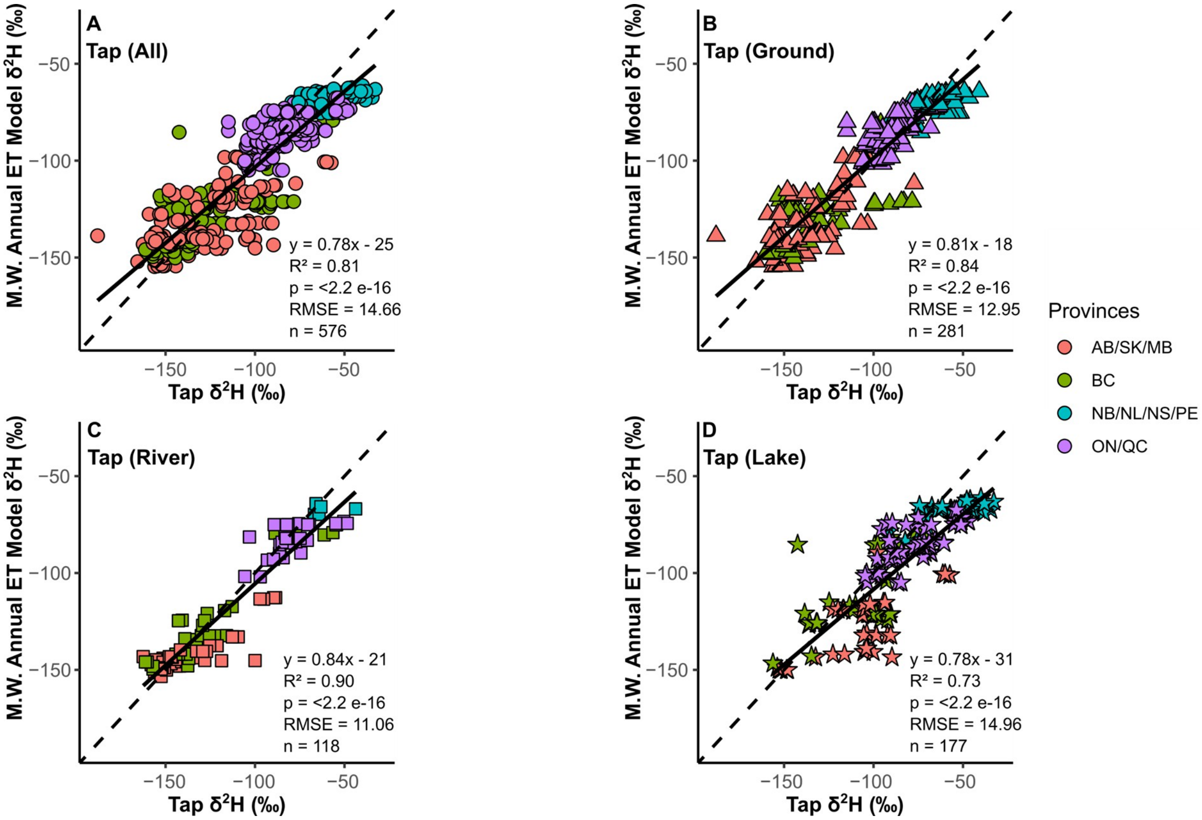

Fig 6. Correlation between tap water δ^2H values and local predicted surface water δ^2H values (based on the Monthly Weighted Annual ET Model). a: all the tap water samples combined, b: tap water sourced from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1 line. The black line represents the best fit linear model.

https://doi.org/10.1371/journal.pwat.0000068.g006

local annual surface water based on our Monthly Weighted Annual ET Model (Fig 7). We calculated the δ^2H residual values as predicted δ^2H values in precipitation or surface water minus the observed tap water δ^2H values. To differentiate these residuals, we name the residual δ^2H values between precipitation (predicted) and tap water (observed) as $R_{precipitation}$ values and the residual δ^2H values between surface water (predicted) and tap water (observed) as $R_{surface.}$values. When the predicted δ^2H value is more positive than the observed tap water δ^2H value, the residual will be positive; conversely when the observed tap water δ^2H value is more positive than the predicted δ^2H value, the residual will be negative. Across the Prairies and British Columbia, large scale $R_{precipitation}$ and $R_{surface}$ patterns show $Tap_{Groundwater}$ sources have more negative δ^2H values than those predicted in local precipitation or in local surface water (positive residuals, Fig E in S1 Text and Fig 7). Tap_{River} and Tap_{Lake} have more positive δ^2H values than the δ^2H values predicted in local precipitation or in local surface water (more negative residuals, Fig E in S1 Text and Fig 7) across Saskatchewan and Manitoba. However, Tap_{River} and Tap_{Lake} show both positive and negative δ^2H residuals with local precipitation and local surface water across Alberta and British Columbia. The Great Lakes and East Coast regions are dominated by negative $R_{precipitation}$ and $R_{surface}$ values for Tap_{River} and Tap_{Lake}, with Tap_{Lake} having the largest negative δ^2H residuals. Conversely, $Tap_{Groundwater}$ in the Great Lakes and East Coast regions have some small positive $R_{precipitation}$ and $R_{surface}$ values.

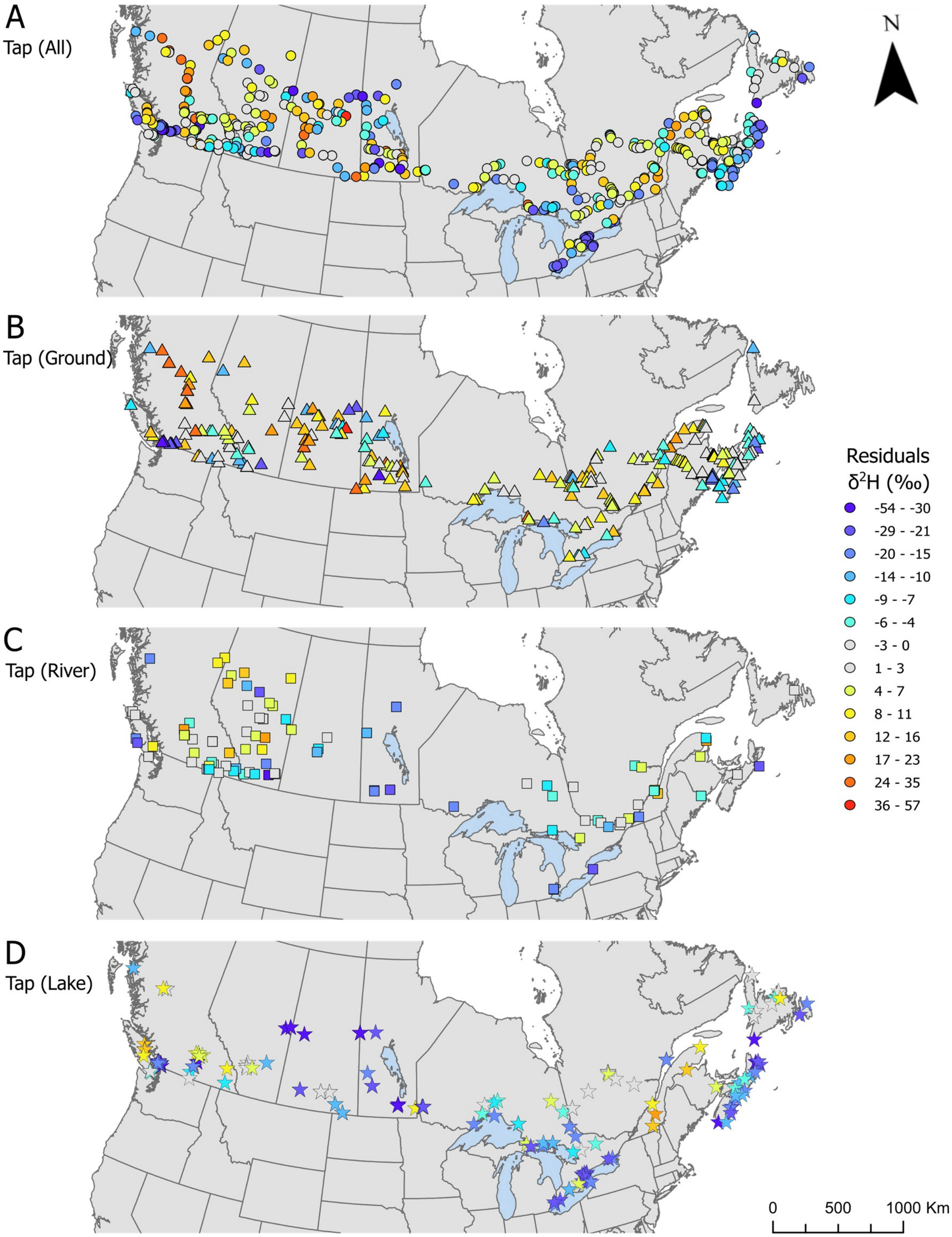

Fig 7. Residuals of δ^2H values between predicted local surface water (based on the Monthly Weighted Annual ET Model) and tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n = 281), c: tap water sourced from rivers (n = 118) and d: tap water sourced from lakes (n = 177). Classification between groundwater, river, and lake is detailed in the Methods. Administrative boundaries are from http://www.naturalearthdata.com/.

https://doi.org/10.1371/journal.pwat.0000068.g007

4. Discussion

4.1 General patterns of δ^2H observations in Canadian tap water and its relationship to δ^2H values in local precipitation and local surface water

As demonstrated in other studies [19, 20, 23, 37], the spatially coherent regional patterns of tap water δ^2H (Fig 2) and their strong correlation with local precipitation (annual/summer) (Fig 5 and Fig C in S1 Text and Table 2) indicate that precipitation is the primary control of tap water δ^2H composition in Canada. The annual and summer water balance models improve the predictability of δ^2H values of $\text{Tap}_{\text{River}}$ and Tap_{Lake}, but not $\text{Tap}_{\text{Groundwater}}$ (Fig 6 and Fig D in S1 Text and Table 3), providing insights into post precipitation processes. The water balance modeling approach described above does not account for isotopic fractionation due to evaporation, or for infiltration. As infiltration rates can vary seasonally, this might influence the predicted δ^2H values. In this study, we interpreted residual δ^2H values between our predicted local surface water and observed tap water (Fig 7) as reflecting either evaporative losses (for negative residuals) or other processes not accounted for in the water balance modeling [20].

4.2 Regional patterns in observed δ^2H values of tap water

4.2.1 East Coast regions (New Brunswick, Nova Scotia, Prince Edward Island and Newfoundland). In the East Coast regions, more positive δ^2H values and *d-excess* values in tap water (Figs 2 and 3) coincide with warm and humid summers and a year round rainy climate [38, 39]. This pattern is irrespective of the source of the tap water samples, and indicates modern precipitation is the primary source of tap water in these regions. We found some small positive $R_{\text{precipitation}}$ and R_{surface} values for $\text{Tap}_{\text{Groundwater}}$ (~38%) and $\text{Tap}_{\text{River}}$ (~33%) compared to Tap_{Lake} (~17%) (in red, Fig E in S1 Text and Fig 7). Similarly, Gibson et al. [28] observed positive δ^2H residuals between the predicted δ^2H values in precipitation and observed δ^2H values in eastern Canadian streams, suggesting evaporation into humid oceanic air masses can lead to isotopic enrichment of surface waters along high slope evaporation lines.

Many of the $\text{Tap}_{\text{Groundwater}}$ samples in the East Coast regions (~36%) have low (more negative) *d-excess* values (< 8.5 ‰) (Fig 3) indicating significant evaporative losses. Also ~62% of the total $\text{Tap}_{\text{Groundwater}}$ samples showed negative $R_{\text{precipitation}}$ and R_{surface} values (in blue, Fig E in S1 Text and Fig 7), which also supports evaporative losses in these waters. Comparatively, a recent study suggested that the Maritime regions exhibit some of the lowest evaporation related losses in Canada [40]. The lower d-excess values and large negative $R_{\text{precipitation}}$ and R_{surface} values likely reflect a combination of misclassification of municipal water sources and water management processes occurring during the storage and distribution of water to residents. For example, some $\text{Tap}_{\text{Groundwater}}$ samples in Nova Scotia originate from Middle Lake Road Wells where groundwater is stored on surface reservoirs (**Data A in S1 Text**, available at https://doi.org/10.6084/m9.figshare.19243518) [41]. Another example is, the City of Saint John in New Brunswick which uses the South Bay Treatment Facility and Loch Lomond Drinking Water Treatment Facility for water treatment and storage [42]. It is likely our $\text{Tap}_{\text{Groundwater}}$ samples from St John is indicative of evaporation losses during treatment and storage at those facilities, as water loss is common through this process [43]. We also explored satellite images by using the latitude and longitude of each of the anomalous $\text{Tap}_{\text{Groundwater}}$ samples with high evaporation signals and found out some of them directly fall close to lakes, pounds and surface reservoirs (**Data A in S1 Text**, available at https://doi.org/10.6084/m9.figshare.19243518). So it is likely some of the anomalous $\text{Tap}_{\text{Groundwater}}$ samples may have been misclassified by the municipalities or that these municipalities store groundwater on surface reservoirs. All these factors could have contributed to the evaporation signals found in a number of $\text{Tap}_{\text{Groundwater}}$

samples from the East Coast regions. In the future, as temperatures warm, such isotope signals would be practical to assess water management strategy and quantify losses of exploited groundwater. In dry regions, evaporative losses from reservoirs can run in excess of several million dollars for large cities [18, 25].

Approximately 58% and 83% of the total $\mathrm{Tap}_{\mathrm{Lake}}$ samples display low (more negative) *d-excess* (<8.5 ‰) and negative $R_{\mathrm{precipitation}}$ and R_{surface} values respectively. These negative *d-excess* and negative residuals are found mainly in Newfoundland and Nova Scotia suggesting significant evaporative losses from these coastal lakes. Most of these samples originate from small lakes or artificial pounds such as Lake George, Little Lake, Sand Lake, Landrie Lake, Lake Major and Rodney Lake, for which higher evaporative losses is expected (**Data A in S1 Text**, available at https://doi.org/10.6084/m9.figshare.19243518). Many of these lakes are used to supply water to small towns or communities. These regions could benefit from isotopic monitoring to assess the long-term losses of water due to natural and local water management strategies and to improve the sustainability of their water management practises.

4.2.2 The Great Lakes regions (Ontario and Quebec). In the Great Lakes regions, more positive δ^2H values dominate for tap water, similar to what is observed in precipitation for this region [44]. However, these tap waters show an interesting combination of positive and negative *d-excess* values for $\mathrm{Tap}_{\mathrm{Groundwater}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ respectively. $\mathrm{Tap}_{\mathrm{Groundwater}}$ samples have *d-excess* similar to those found in precipitation in these regions, suggesting limited evaporative losses [28]. The more positive *d-excess* of the $\mathrm{Tap}_{\mathrm{Groundwater}}$ reflects the amount of recycled water fluxes ('lake-effect' precipitation events) in the Great Lakes regions, as suggested by earlier studies [45, 46]. Aquifers that recharge near the lakes have more positive *d-excess* values than areas that are further away from these lakes [47]. Conversely, $\mathrm{Tap}_{\mathrm{Lake}}$ have more negative *d-excess* values, and negative $R_{\mathrm{precipitation}}$ and R_{surface} values, suggesting they have undergone more evaporative losses with its associated fractionation [12]. Bowen et al. [19] showed similar patterns of "low *d-excess* regions" around the Great Lakes in the United States, however in Bowen et al. [19], the sources for those tap water samples were not known. Here, we show that $\mathrm{Tap}_{\mathrm{Lake}}$ can undergo significant evaporation in these regions [48]. Such high evaporative losses could be partially due to tap water management related issues as recent study suggests this region to have very limited evaporative losses [40]. Except for a few small lakes such as Aspey Lake, Lauzon Lake, Lake Sassagianga and Lake Wawa, most of the $\mathrm{Tap}_{\mathrm{Lake}}$ samples in these regions are sourced from the Great Lakes (**Data A in S1 Text**, available at https://doi.org/10.6084/m9.figshare.19243518). The risks and issues associated with these water resources with respect to climate change occurs over longer timescales and requires a good understanding of the long-term water balance of the Great Lakes [48–50]. Long-term seasonal and multi-annual isotopic monitoring of tap waters could be used to identify the long-term effect of climate or water management practices on tap water supplied by lake waters in the Great Lakes region.

4.2.3 The prairies (Alberta, Saskatchewan and Manitoba) and British Columbia regions. In the Prairies and British Columbia, the δ^2H values of tap water shift to more negative values, and are generally associated with more negative *d-excess* values (Figs 2 and 3), as expected from the progressive rainout principle and the semi-arid continental climate conditions (e.g., less rainfall and low relative humidity) driving evaporative losses [29, 38]. The glacier and snow covered Rockies receive substantial orographic rainfall (mountain effects) [7, 39, 51], and have the most negative δ^2H values in our dataset. These mountainous regions also display quite negative *d-excess* values, suggesting substantial evaporative losses as expected with continental and seasonal climate patterns [52, 53]. These δ^2H patterns are consistent with earlier findings in precipitation and surface waters in these regions [28, 44]. The overall evaporative losses patterns follow the natural evaporative losses found in these regions [40]. However, the lack of stations in the Pacific coast can limit the accuracy of the predicted isotopic values in precipitation in the BC regions [19, 34]. This could influence $R_{precipitation}$ and $R_{surface}$ values seen in the BC regions. However, we found that the predicted δ^2H values in precipitation follow similar patterns as tap δ^2H values and does not appear to influence the residuals in any clear direction.

4.2.3.1 $Tap_{Groundwater}$ in the Prairies and British Columbia. Although we generally presume groundwater sources to be more sheltered from evaporation, ~96% and 92% of the total $Tap_{Groundwater}$ samples have more negative (low) *d-excess* in the Prairies and British Columbia, respectively (Fig 3). Also, 62% of $Tap_{Groundwater}$ samples in both the Prairies and British Columbia have positive $R_{precipitation}$ and $R_{surface}$ values (in red, Fig E in S1 Text and Fig 7). The more negative δ^2H values in $Tap_{Groundwater}$ suggest that winter precipitation and snow/glacier melt runoff are important sources of water recharge to these aquifers in these regions. Strong water contribution from mountains is well-established across the semi-arid regions of North America [19, 54]. In Canada, the more negative δ^2H data in $Tap_{Groundwater}$ also reinforces the importance of winter precipitation and snowmelt in recharging Prairies aquifers, even those distant from mountain zones [5, 55]. Groundwater aquifers in British Columbia are also dependent on precipitation in the Rockies for recharge (snow/glacier melt). However, the more negative *d-excess* in these regions suggests that those snow/glacier melt runoff are highly evaporated. Snow/glacier melt runoff from mountainous regions is often stored in natural and artificial lakes and wetlands along their path, facilitating high evaporation rates in arid regions [28, 56].

4.2.3.2 Tap_{River} and Tap_{Lake} in Alberta and British Columbia. Tap_{River} and Tap_{Lake} of Alberta and British Columbia display a mix of positive (53% and 37%, respectively) and negative $R_{precipitation}$ and $R_{surface}$ values (47% and 63%, respectively) (Fig E in S1 Text and Fig 7). The majority (~83%) of the total Tap_{River} and Tap_{Lake} samples in these regions also have very negative *d-excess*. The positive $R_{precipitation}$ and $R_{surface}$ values combined with more negative *d-excess* in Alberta and British Columbia is similar to what was observed for the $Tap_{Groundwater}$ across the Prairies and British Columbia, and is attributed to snow and glacier melt contribution and evaporative processes along river paths [9, 19, 28]. The negative $R_{precipitation}$ and $R_{surface}$ values in these regions also suggest evaporative losses in the majority of these rivers and lakes sources. In British Columbia, out of 41 Tap_{Lake} samples at least 19 samples are sourced from human-made reservoirs (**Data A in S1 Text**, available at https://doi.org/10.6084/m9.figshare.19243518). British Columbia is also sourcing tap water from some small natural lakes such as Commox Lake, Kalamalka Lake, Osoyoos Lake and Tchesinkut Lake which show some of the highest evaporative losses in our dataset (*d-excess* ranging from -35 to -11 ‰). Gibson et al. [34] suggests that many of the smaller low elevation lakes in British Columbia are disconnected from the regional river drainage networks and therefore more susceptible to evaporation. The only samples with more positive *d-excess* values were collected in British Colombia (20 samples and mainly river and reservoirs), and likely reflect the higher relative humidity in coastal setting. Isotopic measurements would be useful to track the vulnerability of some water resources (e.g., mountainous lakes) through time and assess the long-term impact of climate change on the availability of different water resources for tap water consumption.

4.2.3.3 Tap_{River} and Tap_{Lake} in manitoba and saskatchewan. Tap_{River} and Tap_{Lake} samples from Manitoba and Saskatchewan show only negative $R_{precipitation}$ and $R_{surface}$ values suggesting significant evaporative losses [28]. Such high evaporative losses from rivers and lakes are common in the eastern Prairies [57, 58] making these regions highly dependent of large rivers originating from the Rockies and/or winter recharge. High evaporative losses occur along the path of large rivers throughout the Prairies (e.g., Athabasca River) or from the slow circulation of waters from open surface reservoirs such as lakes (e.g., Cold Lake, Douglas lake, Meadow Lake, Nickel Lake and Shoal Lake), man-made reservoirs or peatlands [59]. These evaporation mechanisms in the uplands or valleys lead to evaporated δ^2H signatures and more negative *d-excess* for all water sources in these regions [28]. Small changes in winter precipitation in these regions can have a significant impact on availability of the water resources [55]. Long-term monitoring of δ^2H in those tap waters would again help assess water source vulnerabilities to climate or water management practices (e.g., open reservoir storage) to extract water resources more sustainably through the year and limit evaporation [25].

4.3 Seasonal and inter-annual variation in tap water isotopes

Tap waters collected at multiple sites across the Ottawa and Montreal regions show little seasonal or inter-annual variability (Fig I in S1 Text). In those regions, most sites source their water almost exclusively from the Ottawa River and the Saint Lawrence River, respectively. Both of those large rivers integrate buffered isotopic signals coming from large catchments that show small seasonal fluctuations with more negative δ^2H values during snowmelt and more positive δ^2H values during the summer [60, 61]. Tap water δ^2H values of those large municipalities show similar seasonal trends, but because large cities pump and store water all year long, isotopic fluctuations are attenuated. Conversely, Sudbury municipality source tap water from multiple lakes, groundwater wells and small rivers. δ^2H values in tap water across the municipality of Sudbury show a much larger range and more abrupt variations (Fig I in S1 Text). These variations likely reflect a switch in water sources by water management companies from surface water to groundwater (Fig I in S1 Text). Isotopic measurements of tap water are not only useful to quantify the impact of climate and evaporation on the water resources [21, 23, 29] but also provide a tool to track urban water supply system dynamics [26]. This small seasonal and inter-annual dataset supports the need for long-term monitoring of isotopes in tap water to quantify climatic and human-management impact on the water resource of Canada.

4.4 Climate change and tap water resources sustainability

With ongoing global warming, water balance changes will continue across Canada influencing the supply of tap water to Canadians. Changes in rainfall patterns and a reduction in snow and ice cover will alter the water balance of many watersheds [2]. The earlier and reduced runoff volume observed in many rivers across Canada can affect adequate water storage and threaten late-summer water availability [62], particularly in semi-arid regions. Winter discharge is predicted to increase with warmer winter and earlier snowmelt whereas reduced snowpack, and loss of glaciers will result in smaller river discharge in the summer [63]. Regionally, reduced snow and glacier melt from the Rockies will affect the recharge of important aquifers and rivers, impacting downstream communities that depend on these water sources [4]. Evapotranspiration related water losses will also accelerate in the upcoming decades with increased warming [63] further modifying the water balance of rivers and lakes that are often critical for human water supply throughout Canada. As new water management infrastructures are developed, reducing evaporation, tracing water provenance, and managing water sources are key priorities, particularly in regions where the water resources are scarce and vulnerable (e.g., Prairies). Water management plans should integrate regional water balance considerations in their water management. However, such regional considerations are often limited by the fragmented and localized water governance [1]. As seen in other countries, poor water management practices might exacerbate water losses in semi-arid regions (e.g., the Prairies) [64]. It is therefore critical to take into account the specific regional and long-term impacts of water management practices on Canadian water resources [64, 65]. Isotopic monitoring is an easy and cost-effective approach to trace water provenance, quantify evaporation, or identify early climatic and hydrologic changes to the water resources at the regional scale. Our models and databases contribute to this aim by providing an isotopic baseline in Canadian tap water for long-term monitoring of climatic and anthropogenic threats to the Canadian tap water resources.

4.5 Recommendations to water managers

Based on the findings of this study, we argue that a long-term network based monthly collection of tap water isotopes data across Canada could provide vital information to water managers for a more sustainable management of Canadian tap water resources. Our study shows that isotopes found in tap water are particularly useful to trace the dynamic changes in water sources used by municipalities and to quantify evaporative losses. In this study, we showed abrupt δ^2H variations across tap water samples collected in the Sudbury municipality over several years likely reflecting a switch in water sources between surface water and groundwater (Fig I in S1 Text). Such isotopic shift in tap water can trace the dynamic water supply in complex and poorly documented water distribution systems. Jameel et al. [26] demonstrated that isotopes data could unravel the physical structure of complex municipal supply systems and map the geographic distribution of water sources. As many water supply systems are poorly documented, monthly isotopes data in tap water offers an inexpensive solution to trace sources through time and space to validate the provenance of water along the distribution network, to monitor water budgeting and distribution and to trace the source or transmission of a contamination along the water supply system.

At municipal scale, our study also showed that isotopes could help quantify water evaporation losses. We showed that isotopes could trace how the storage of groundwater in small artificial lakes and pounds in the East Coast likely led to high evaporative losses. We argue that a series of spatiotemporal tap water isotopes studies in targeted cities with high evaporation rate (e.g., Prairies or East Coast) will help disentangle the contribution of natural and anthropogenic evaporative losses across the coupled natural-human systems [25]. Partitioning natural and anthropogenic evaporations would be critical to improve water management strategies by limiting excessive water losses, for example through concepts of smart location and geoengineering [66]. Although there are existing physical methods used in evaporation estimation, such as the pan evaporation method, the Dalton's method, the eddy covariance technique, or the Bowen ratio energy balance, these methods can be unreliable, particularly at increasingly large scale, because they are influenced by hidden physical drivers, time-scale-dependent feedbacks, and complex heterogeneities that govern reservoir evaporation rates [66]. In comparison, tap water isotopes data is cost-effective without requiring expensive field deployment of instruments and is less influenced by these confounding factors and provides a reliable and independent method to estimate evaporative losses along the tap water cycle [25]. However, some challenges remain with isotope-based evaporation estimates, as it is heavily dependent on the quality of existing geospatial data available including those on wind, humidity and temperature.

Isotopic monitoring offers advantages at both small scale and large scale. In particular, the long-term isotopic monitoring of tap water would be useful to track the effect of accelerating warming on the tap water resources. Temporal isotopic monitoring can provide estimate of the proportion of water coming through different natural sources through time as well as estimate of evaporation rate along the entire coupled natural-human systems. In this study, we have demonstrated that the Prairies and some regions of British Columbia are heavily reliant on water coming from the Canadian Rockies and from winter precipitation (Fig 6, discussed in section 4.2.3) echoing earlier findings [5, 19]. With climate warming, early and rapid spring flow, reduction in snow cover and glacier melt from the Rockies will impact water availability and supply across the downstream communities [4]. We also demonstrated that tap water in the Prairies and British Columbia are highly evaporated. Isotopic monitoring of tap water would provide a mean to characterize the rate at which these tap water resources are changing in terms of supply sources and evaporation losses at the regional scale. Those studies would give regional and local water planners data to collaborate on water use plan by identifying future vulnerabilities of the water supply and by providing metrics to characterize the effectiveness of current water management strategies. The isotopes maps and data generated in such monitoring effort would also facilitate discussion with decision-makers to invest in more sustainable water supply systems/alternatives (e.g., switching from surface to groundwater and 'conservation at source' concepts).

4.6 Forensic application

In addition to its potential use in water resource monitoring, our database is also a valuable tool in forensic studies. Local tap water is incorporated into many manufactured products (e.g., drugs, explosives) and organic tissues (e.g., food, human tissues). The isotopic signatures of tap water are usually reflected in these materials, providing an "isotopic fingerprint" to trace their origin. For example, a strong relationship exists between the δ^2H and δ^{18}O composition of local tap water and human hair providing a geolocation tool in determining origin and geographic movement of humans of interests and tracing the mobility or origin of individuals in cold cases [67, 68]. The dataset generated in this study provides a baseline to track forensically relevant materials across Canada. Recent studies in Canada have already demonstrated how this database could provide key information to investigate cold cases [69, 70] and reconstruct individual travel history [71].

5. Conclusions

Our study suggests that precipitation is the primary source of tap water across Canada. However, many natural and anthropogenic processes also contribute to δ^2H variability in tap water across Canada. The tap water resources in Western Canada are heavily dependent upon glacial melt from the Rockies and on winter precipitation recharging aquifers across the Prairies and British Columbia. In Eastern Canada, even if water is abundant, tap water is often supplied by large rivers and lakes that are dependent on regional water balance. The Canadian water resources are vulnerable to climate change and the threats are often worsened by water management practises. As natural tracer of hydrological processes in watershed, tap water isotopes show promises as an effective tool for water managers.

At the municipal scale, tap water isotopes monitoring can trace water sources along the distribution system and quantify evaporation of natural and anthropogenic reservoirs. At the regional scale, tap water isotopes monitoring would help quantifying the effect of climate change on the natural and anthropogenic processes influencing the tap water sources. As warming accelerates across Canada, effective regional water supply management strategies need to be implemented to limit evaporative water losses from the regionally important water resources. Our isotopic observations of tap water from across Canada provide a baseline, and establish a foundation to develop long term isotopic monitoring as a tool to better manage the water resources from source to tap by accounting for local and regional vulnerabilities.

Supporting information

S1 Text.
(DOCX)

Acknowledgments

We thank all the volunteers who participated in tap water samples collection from across Canada and the staff of Ján Veizer Stable Isotope Laboratory (Patricia Wickham, Wendy Abdi and Paul Middlestead) at University of Ottawa for assisting with laboratory analysis.

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