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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 measurements (?2H and ?18O) 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 (TapGroundwater), river (TapRiver) and lake (TapLake). ?2H in tap water correlates strongly with values predicted for local precipitation across Canada with a stronger correlation for TapGroundwater and TapRiver than for TapLake. We then constructed water balance models to predict the ?2H of surface water across Canada, and validated it against Canadian river water ?2H data. ?2H in tap water correlates strongly with values predicted for surface water across Canada with a stronger correlation for TapRiver and TapLake than for TapGroundwater. TapGroundwater ?2H values reflect the ?2H of annually averaged precipitation, whereas TapRiver and TapLake ?2H values reflect post-precipitation processes. We used the ?2H residuals between the observed and predicted ?2H values to assess regional processes influencing tap water ?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. This study provides baselines for isotopic monitoring of tap water resources and forensic studies in Canada.

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Spatial Variations in Tap Water Isotopes Across Canada: Tracing Water from Precipitation to Distribution and Assess Regional Water Resources

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Key Points:

- Natural and anthropogenic processes cause significant evaporative water losses across Canada
- Glacier and snow melt from the Rockies are major contributors of tap water across Western Canada
- We present the first national map of tap water δ^2H across Canada with water management and human forensic implications

Abstract

With global warming and increasing water use, tap water resources need sustainable management. We used hydrogen and oxygen isotope measurements (δ^2H and δ^{18}O) 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 in tap water correlates 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 it against Canadian river water δ^2H data. δ^2H in tap water correlates strongly with values predicted for surface water across Canada with a stronger correlation for Tap_{River} and Tap_{Lake} than for $Tap_{Groundwater}$. $Tap_{Groundwater}$ δ^2H values reflect the δ^2H 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. This study provides baselines for isotopic monitoring of tap water resources and forensic studies in Canada.

Plain Language Summary

We present a geo-hydrological study of hydrogen and oxygen stable isotope in tap water across Canada to assess regional water resources vulnerability. To trace water cycling from precipitation to tap water supply, we compared tap water δ^2H values with those of local precipitation. To understand post-precipitation processes, we constructed water balance models to predict surface water δ^2H across Canada, and compared tap water δ^2H with those of local surface water. Tap water δ^2H exhibit strong correlations with both precipitation and surface water δ^2H, suggesting precipitation supplies most of Canadian tap water. The isotopic difference between tap water and precipitation and surface water demonstrate snow/glacier melt from the Rockies is an important source for groundwater recharge across Western Canada, and some rivers and lakes in Alberta and British Columbia. Tap waters are highly evaporated across Western Canada regardless of their source and the Great lakes region mainly those sourced from lakes. Many localities in East Coast regions rely on natural and human-made lakes, including storage of pumped groundwater on surface reservoirs, and are subjected to evaporation. We present the first national map of tap water δ^2H measurements, providing a baseline for tap water resources monitoring and forensic applications.

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 governance (Bakker & Cook, 2011; Medeiros et al., 2017). 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 (Government of Canada, 2017b). 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 (Government of Canada, 2017b). Warming, reduced snow cover and glacier retreat from the Rockies will continue to impact water availability and supply across the Prairies (Bakker, 2009). 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 (Jasechko et al., 2017). In addition to these natural threats to water availability, Canadian water management practices vary between localities. 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 require regional monitoring of tap water resources and an understanding of the impact of climate change on these tap water sources.

Stable hydrogen and oxygen delta measurements ($\delta_{VSMOW}{}^{2}$H and $\delta_{VSMOW}{}^{18}$O, which are herein expressed as δ^{2}H and δ^{18}O, respectively) are powerful tracers of water cycling processes. Global patterns in the isotopic composition of precipitation follow climatic and geographic patterns (e.g., meridional water transport, continentality, elevation, temperature and relative humidity variations; (Dansgaard, 1964; Feng et al., 2009; Gat, 1980; Hollins et al., 2018; Kendall & Coplen, 2001). Environmental water resources inherit their isotopic composition and spatiotemporal variations primarily from modern precipitation (Davisson et al., 1999; Dutton et al., 2005; Gat & Gonfiantini, 1981; Smith et al., 2002). 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 (Good et al., 2014; Landwehr et al., 2014; Tipple et al., 2017). 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 (Bowen et al., 2007, 2011; Du et al., 2019; Ehleringer et al., 2016; Wang et al., 2018).

In a pioneering study, Bowen et al. (2007) used δ^{2}H and δ^{18}O of tap water to trace regional hydrological processes and to characterize regional water issues across the contiguous United States. Since then, tap water δ^{2}H and δ^{18}O analyses have been successfully applied to water investigations across the globe, including partitioning regional and seasonal reliance on surface and groundwater for supply, identifying regions extracting fossil groundwater, or importing water through inter-basin transfer (Du et al., 2019; Good et al., 2014; Wang et al., 2018; de Wet et al., 2020). At the scale of a city (e.g., Western USA), Jameel et al. (2016; 2018) and Tipple et al. (2017) 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, and based on the hypothesis that vulnerability to climatic change and water management can vary depending on the source type, we explore risks at the source level, (Wang et al., 2018; de Wet et al., 2020). First, we analyzed the tap water δ^{2}H isotopic patterns over Canada and compared them with predicted local precipitation isotopic values (Bowen, 2019). A series of water balance models to predict modifications expected in surface water isotopic values across Canada were constructed and validated via comparison to river water isotopic values collected from across Canada (Gibson et al., 2020), and compared the tap water isotopic values with those of predicted local surface water. Finally, we analyzed tap water isotopic values, including d-excess, residual isotopic values between tap water and local precipitation, and residual isotopic values between tap water and local surface water altogether to assess regional hydrological processes, vulnerability to ongoing climate warming and potential water management issues. Our analysis offers a baseline for nationwide monitoring of critical water resources (Bowen et al., 2007; de Wet et al., 2020; Zhao et al., 2017). 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 (e.g., Ehleringer et al., 2008).

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) (Dataset S1) 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) (Dataset S2). 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 ($Tap_{Groundwater}$), river (Tap_{River}) and lake (Tap_{Lake}). $Tap_{Groundwater}$ is defined as tap water sourced from wells. Tap_{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. We also recorded the name of the rivers and lakes sources at each site. Dataset S1 and S2 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 (δ^2H = −198.5 ± 2.0 ‰ and δ^{18}O = −24.55 ± 0.2 ‰), W-10 (δ^2H = −85.9 ± 2.0 ‰ and δ^{18}O = −11.84 ± 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 ‰ (Brand et al., 2014)), GISP (δ^2H = −189.5 ± 1.2 ‰, δ^{18}O = −24.76 ± 0.09 ‰ (IAEA, 2007))); and SLAP (δ^2H = −428 ‰, δ^{18}O = −55.5 ‰ (Brand et al., 2014)). Tap water δ^2H and δ^{18}O values were obtained using the LIMS for Light Stable Isotopes (Shrestha & Yesha, 2017). 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. The Ján Veizer Stable Isotope Laboratory also applies this uncertainty to the three internal water reference materials used for normalization, but the standard deviation of replicate measurements is typically better than these uncertainties.

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

To analyze the spatial variability of tap water isotopes across Canada, we mapped the δ^2H and *d-excess* (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 (Dansgaard, 1964)) in ESRI ArcGIS Pro. Since the tap water δ^2H and δ^{18}O show very similar patterns, we only interpreted the correlation between the observed δ^2H values of tap water and the predicted δ^2H values of local precipitation (Bowen, 2019). At each tap water site, seasonal and annual predicted precipitation δ^2H values were extracted. 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., 2007). 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 the isotopic gradient from coast toward inland locations accurately (Bowen et al., 2007; Gibson et al., 2018).

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 in surface water across Canada. As δ^2H and δ^{18}O show very similar patterns, we built the models to predict surface water δ^2H only. Unlike the precipitation model that only accounts for atmospheric controls of isotopic variability, water balance models incorporate isotopic variability and post-precipitation modifications associated with surface hydrology.

Four datasets were used for the water balance modelling: 1) long-term monthly mean isotopic values for global precipitation (Bowen, 2019) ; 2) North American flow direction (HydroSHEDS, 2020); 3) long-term monthly mean of daily total precipitation (PSL, 2000) and 4) long-term monthly mean evapotranspiration (PSL, 2000). We followed a similar approach to Bowen et al. (2011) 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 1sq-km resolution using the equations in Table 1 within the North America boundary defined by the HydroSHEDS dataset (Figure 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. (2011), we built seasonal water balance models (Table 1) to assess seasonal isotopic variability in surface water. We built the annual and seasonal models using two different approaches: (1) by propagating the δ^2H values in 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 eight water balance models (Table 1).

Table 1. Equations used to calculate discharge and isotopic flux at 1sq-km 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^2H)
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 Winter Model	Q = Nov P +….+ Apr P	δQ = (Nov P * Nov δP) +…. + (Apr P * Apr δP)
4. Monthly Weighted Annual ET Model	Q = Jan (P - ET) +…. + Dec (P-ET)	δQ = (Jan (P - ET) * Jan δP) +…. + (Dec (P - ET) * Dec δP)
5. Monthly Weighted Summer ET Model	Q = May (P - ET) +…. + Oct (P-ET)	δQ = (May (P - ET) * May δP) +…. + (Oct (P - ET) * Oct δP)
6. Monthly Weighted Winter ET Model	Q = Nov (P - ET) +…. + Apr (P-ET)	δQ = (Nov (P - ET) * Nov δP) +…. + (Apr (P - ET) * Apr δP)
7. Annual average Model	Q = Jan P +….+ Dec P	δQ = total annual (P) * annual average δP
8. Annual average ET Model	Q = total annual P - total annual ET	δQ = (total annual P - total annual ET) * annual average δP

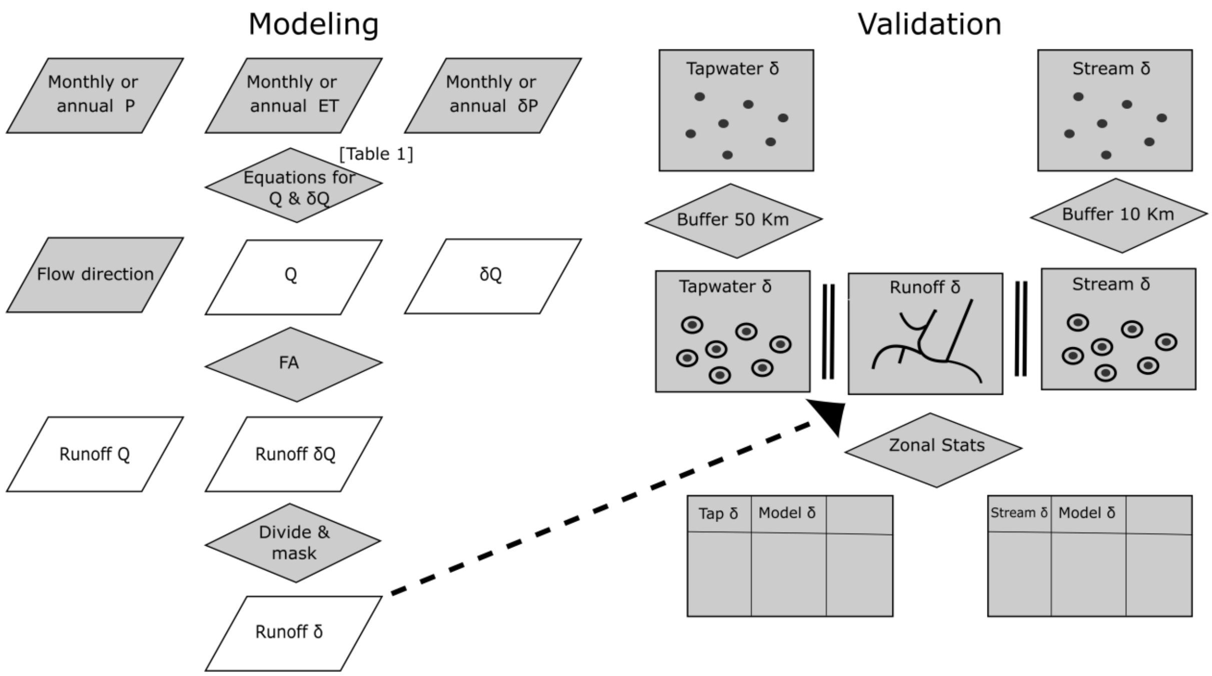

Figure 1. Workflow for GIS based water balance modeling and validation modified from (Bowen et al., 2011). Diamond = operations and rectangular (shaded) = input raster data sets. P = precipitation; ET = evapotranspiration; δP = isotopic composition of precipitation; FA = flow accumulation; Q = discharge, and δQ = isotopic flux associated with discharge.

2.5 Validation of the eight water balance models: comparison between predicted surface water δ^2H values and observed stream water δ^2H measurements

To validate our approach, we first compared our predicted local surface water isotopic values with an observed Canadian stream water δ^2H dataset (Gibson et al., 2020). However, the latitude and longitude of the river water samples collected in the Canadian stream water δ^2H dataset did not always line-up with the Hydroshed. In other words, if we extracted the δ^2H value of the water balance models for the pixel located at the collection site, this value might not correspond to the exact river. In order to compare the predicted δ^2H values in local surface water derived from our models with the observed δ^2H values in stream water, we masked all the pixels (from our models) with total drainage areas <9 km^2 to exclude the small streams (Figure 1) following Bowen et al (2011). We then extracted the predicted δ^2H values in local surface water at each stream sample site by: 1) using a 10 km radius around each stream sampling point and 2) calculating the flux weighted average δ^2H value within this area. We then compared the annual models with observed annual average stream water δ^2H values at 262 sites, the summer models with observed summer average stream water δ^2H values at 241 sites, and the winter models with observed winter average stream water δ^2H values at 217 sites. All the models were validated based on the significantly positive linear correlation between the observed δ^2H values in stream water and the predicted δ^2H values in local surface water from our models.

2.6 Comparison between 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 (Figure 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 measurements in Canadian tap water

δ^2H values in Canadian tap water range from -188‰ to -33‰ (Figure 2). There are strong spatial patterns of increasingly more negative δ^2Hvalues from low latitude coastal regions towards high latitude and high-altitude inland regions (Figure 2). Generally, the most negative δ^2H values were measured 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 (Figure 2). The *d-excess* values of tap water also show large spatial variability, ranging from -35.3‰ to +19.1‰ (Figure 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 (Figure 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 (Figure 3).

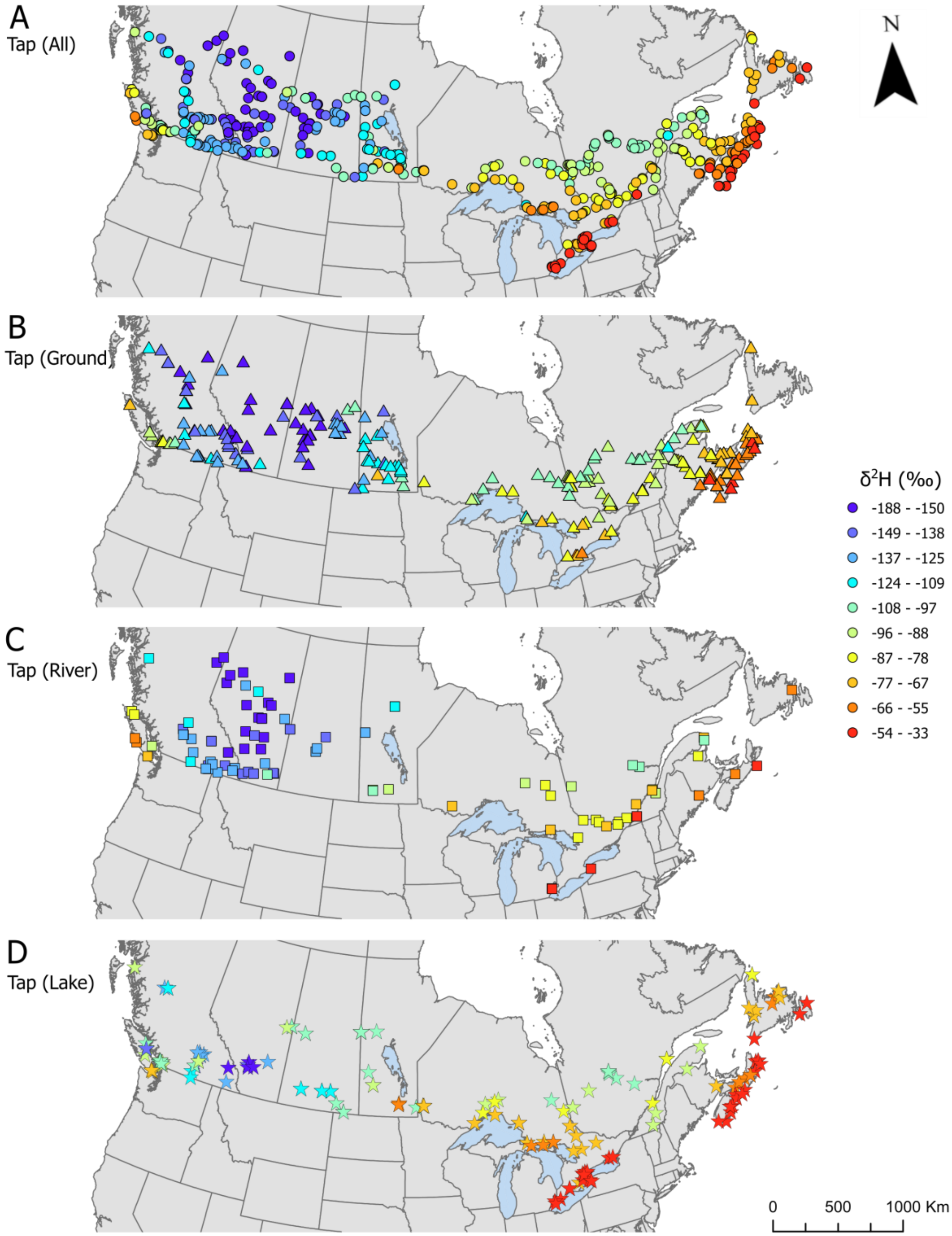

Figure 2. Spatial distribution of sample locations and δ^2H values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n=281), c: tap water sourced from rivers (n=118) and d: tap water sourced from lakes (n=177). Classification between groundwater,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

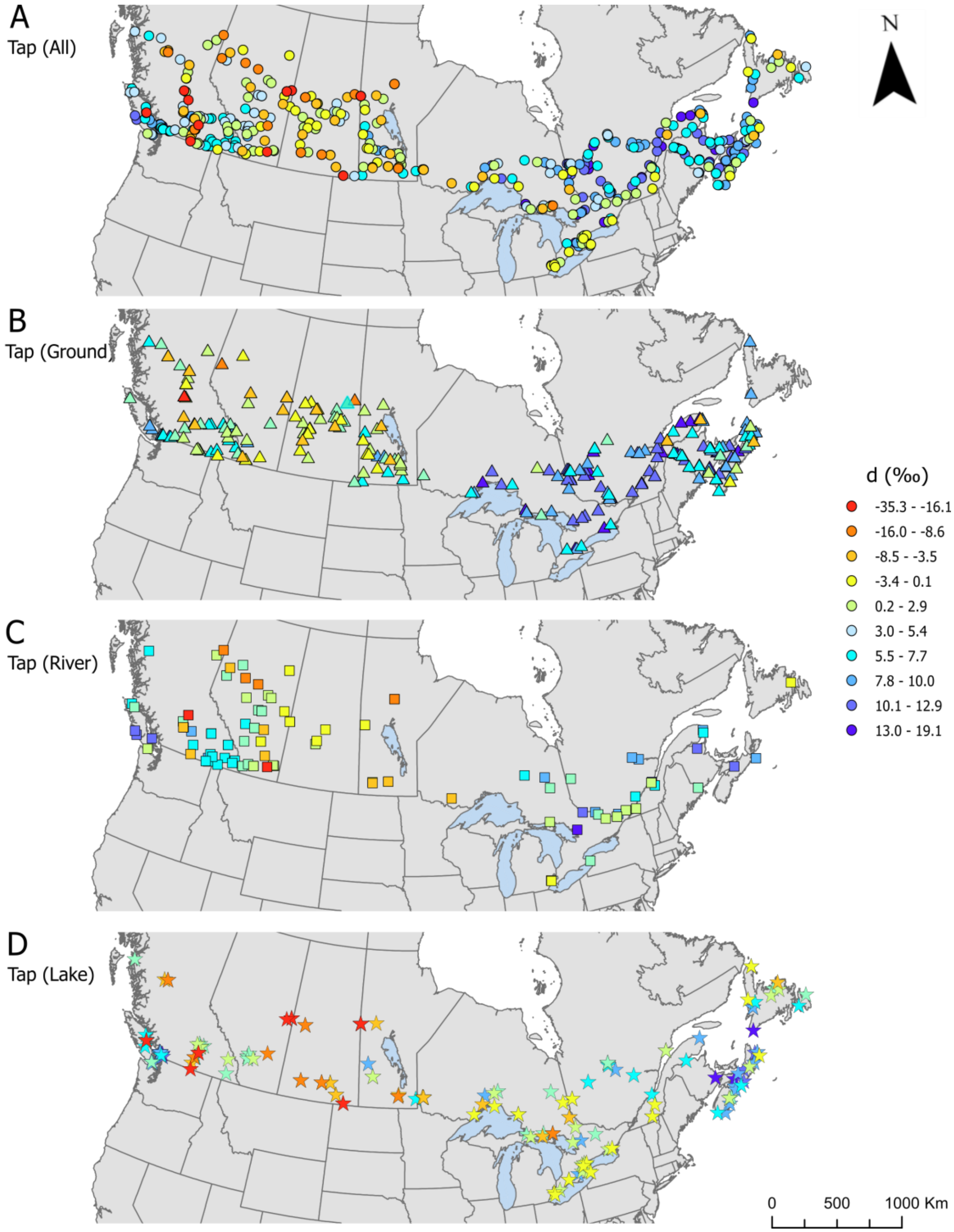

Figure 3. Spatial distribution of sample locations and *d-excess* (d) values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n=281), c: tap water sourced from rivers (n=118) and d: tap water sourced from lakes (n=177). Classification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

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

The δ^2H and δ^{18}O composition of tap water samples generally follows the Canadian Meteoric Water Line (CMWL) (Figure 4). 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, Figure 5 and Figure S1). Plotting the δ^2H in tap water grouped by their pre-classified water sources shows δ^2H values of $Tap_{Groundwater}$ and Tap_{River} have a much higher correlation with local precipitation than δ^2H values of Tap_{Lake}, both annually and seasonally (Table 2, Figure 5 and Figure S1). 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 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).

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
Groundwater	0.86	0.87	0.79
River	0.86	0.88	0.76
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**

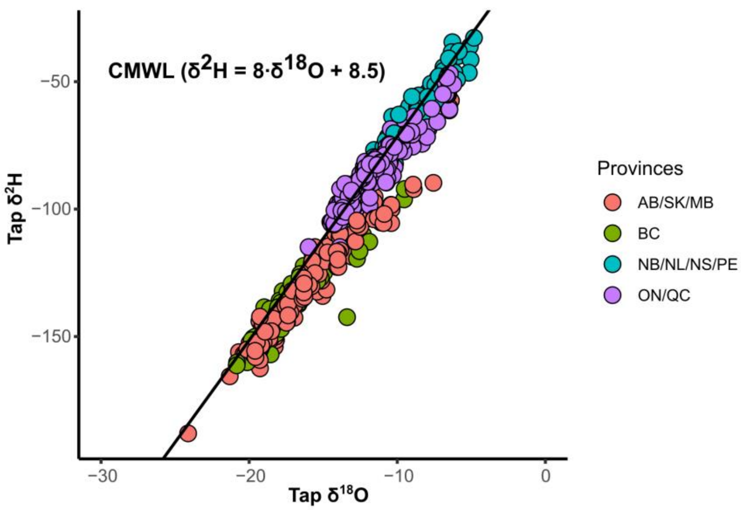

Figure 4. Covariation of tap water δ^2H and δ^{18}O values (n = 576) in relation to Canadian meteoric water line (CMWL) (Gibson et al., 2020).

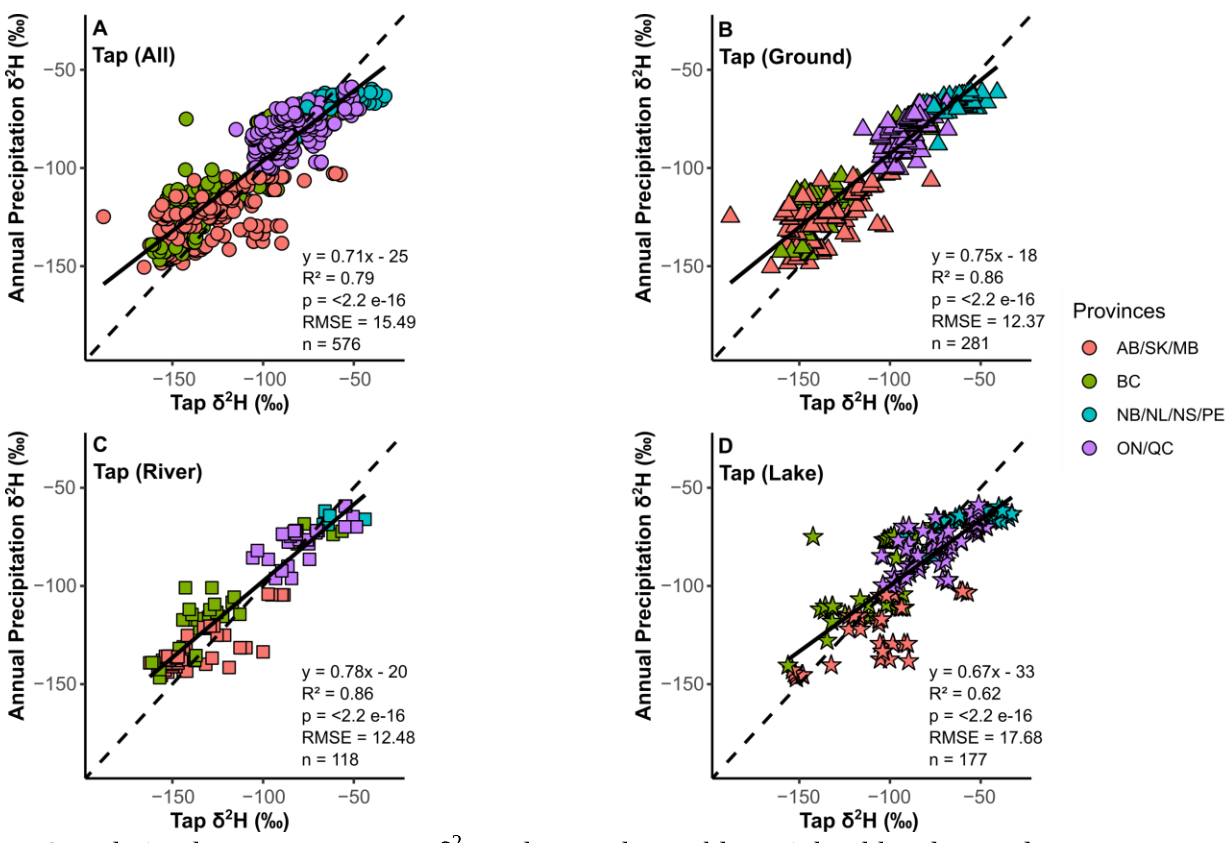

Figure 5. Correlation between tap water δ^2H values and monthly weighted local annual precipitation δ^2H values. a: all the tap water samples combined, b: tap water sourced from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1 line. The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

3.3 Validation of the eight water balance models: relationship between observed stream water δ^2H values and predicted surface water δ^2H values

There is a strong positive correlation between the measured δ^2H values in stream water and predicted δ^2H values in surface water, both annually and seasonally (Figure S4) validating our water balance modelling approach. With the exception of the semi-arid regions of Alberta, Saskatchewan and Manitoba (the Prairies), where the measured stream water δ^2H values are consistently more positive than the predicted values (Figure S4), the monthly weighted annual models perform better (i.e., closer to 1:1 line) than annual average models and seasonal models (Figure S4). Models predicting surface water δ^2H values in the winter always underperform relative to those predicting surface water δ^2H values annually or in the summer (Figure S4). To analyze why measured winter stream water have a much weaker correlation with the monthly weighted winter models, we compared the measured δ^2H values in winter stream water with both local winter and summer precipitation δ^2H values. This analysis shows that observed δ^2H in winter streams have a weaker correlation with local winter precipitation δ^2H and rather a stronger correlation with local summer precipitation δ^2H (Figure S5 and Table S1).

3.4 Relationship between 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, Figure 6 and Figure S2). Plotting the δ^2H values in tap water grouped by their pre-classified water sources shows the water balance models do not improve $Tap_{Groundwater}$ δ^2H prediction (Table 3 and Figure 6) relative to the precipitation-only model (Table 2 and Figure 5). Conversely, the water balance models do improve the prediction of Tap_{River} δ^2H and Tap_{Lake} δ^2H relative to the precipitation-only model. The monthly weighted annual models predict δ^2H values in tap water better than the annual average models, and the monthly weighted summer models perform much better than the monthly weighted winter models.

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 Winter Model	Monthly Weighted Annual ET Model	Monthly Weighted Summer ET Model	Monthly Weighted Winter ET Model	Annual Average Model	Annual Average ET Model
	R^2	R^2	R^2	R^2	R^2	R^2	R^2	R^2
All	0.81	0.81	0.76	0.81	0.81	0.75	0.78	0.78
Groundwater	0.84	0.84	0.83	0.84	0.84	0.83	0.84	0.84
Rivers	0.89	0.90	0.83	0.90	0.90	0.82	0.85	0.85
Lakes	0.73	0.74	0.60	0.73	0.73	0.60	0.65	0.65

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

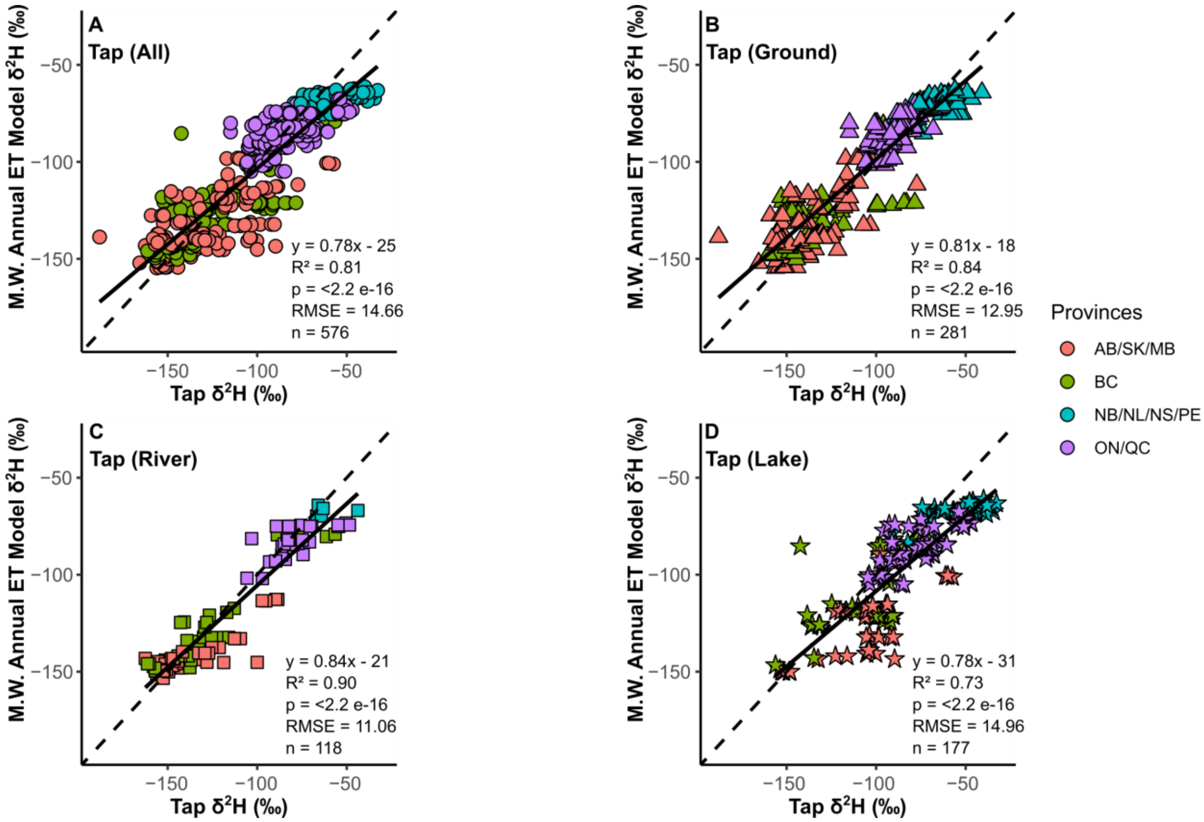

Figure 6. Correlation between tap water δ^2H values and local predicted surface water δ^2H values (based on the Monthly Weighted Annual ET Model). a: all the tap water samples combined, b: tap water sourced from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash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

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) (Figure S3), and the residual δ^2H values between tap water and local annual surface water based on our Monthly Weighted Annual ET Model (Figure 7). We defined the δ^2H residual value as: predicted δ^2H value (either from precipitation or surface water) – measured tap water δ^2H value. In cases where the predicted δ^2H value is more positive than the measured tap water δ^2H value, the residual will be positive; conversely when the measured 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 residual patterns show $\text{Tap}_{\text{Groundwater}}$ sources have more negative δ^2H values than that predicted in local precipitation or in local surface water (positive residuals, Figures S3 and 7). $\text{Tap}_{\text{River}}$ 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 δ^2H residuals, Figure S3 and Figure 7) across Saskatchewan and Manitoba. However, $\text{Tap}_{\text{River}}$ 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 δ^2H residuals with local precipitation and local surface water for $\text{Tap}_{\text{River}}$ 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 δ^2H residuals.

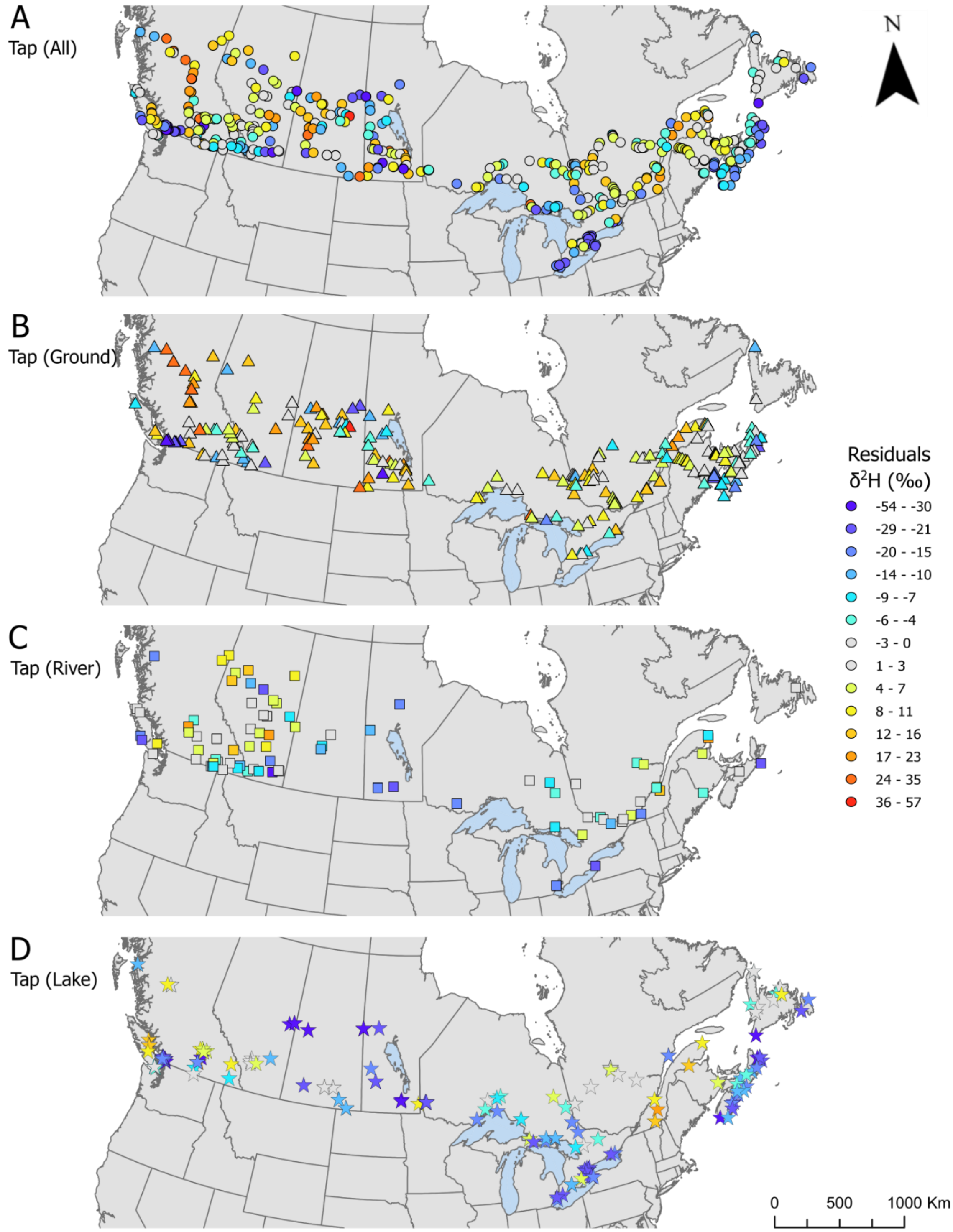

Figure 7. Residuals of δ^2H values between predicted local surface water (based on the Monthly Weighted Annual ET Model) and tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced from groundwater (n=281), c: tap water sourced from rivers

(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/.

4 Discussion

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

As demonstrated in other studies (Bowen et al., 2007, 2011; Stahl et al., 2020; Wang et al., 2018), the spatially coherent regional patterns of tap water δ^2H (Figure 2) and their strong correlation with local precipitation (annual/summer) (Figure 5, Figure S1 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 Tap_{River} and Tap_{Lake}, but not $Tap_{Groundwater}$ (Figure 6, Figure S2 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, nor 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 measured tap water (Figure 7) as reflecting either evaporative losses (for negative residuals) or other processes not accounted for in the water balance modeling (Bowen et al., 2011).

4.2 Regional patterns in measured 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 (Figure 2 and Figure 3) coincide with warm and humid summers and a year round rainy climate (Geographic, 2020; Hall et al., 2020). 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 δ^2H residuals with respect to both predicted local precipitation and predicted surface water mainly for $Tap_{Groundwater}$ (~38%) and Tap_{River} (~33%) compared to Tap_{Lake} (~17%) (in red, Figure S3 and Figure 7). Similarly, Gibson et al. (2020) observed positive δ^2H residuals when measuring δ^2H in eastern Canadian river water, suggesting evaporation into humid oceanic air masses can lead to isotopic enrichment of surface waters along high slope evaporation lines.

Many of the $Tap_{Groundwater}$ samples in the East Coast regions (~36%) have low (more negative) *d-excess* values (< 8.5 ‰) (Figure 3) indicating significant evaporative losses. Also ~62% of the total $Tap_{Groundwater}$ samples showed negative δ^2H residuals with respect to local precipitation and local surface water (in blue, Figure S3 and Figure 7), which also supports evaporative losses in these waters. Such evaporative losses are alarming and indicative of anthropogenic processes. Comparatively, a recent study suggests the Maritime regions exhibit some of the lowest evaporation related losses in Canada (Gibson et al., 2021). Some of these anomalous δ^2H values likely reflect misclassification of municipal water sources, or additional human management practices that contributed to evaporative losses. For example, many localities in these regions pump and store groundwater in open surface reservoirs and small lakes or pounds imprinting evaporation signal in $Tap_{Groundwater}$. 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 (Jameel et al., 2016; Tipple et al., 2017).

Approximately 58% and 83% of the total Tap_{Lake} samples display low (more negative) *d-excess* (<8.5 ‰) and negative δ^2H residuals (with respect to local annual precipitation and local surface water) respectively (mainly in Newfoundland and Nova Scotia), both independently 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. Many of these lakes are used to supply water to small towns or communities. Here again, isotopic data would be practical to assess local water management strategies and quantify regional water losses to water sustainability targets.

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 (Brown, 1971). However, these tap waters show an interesting combination of positive and negative *d-excess* values for $Tap_{Groundwater}$ and Tap_{Lake} respectively. $Tap_{Groundwater}$ samples have *d-excess* similar to those found in precipitation in these regions, suggesting limited evaporative losses (Gibson et al., 2020). The more positive *d-excess* of the $Tap_{Groundwater}$ reflects the amount of recycled water fluxes ('lake-effect' precipitation events) in the Great Lakes regions, as suggested by earlier studies (Gat et al., 1994; Machavaram & Krishnamurthy, 1995). Aquifers that recharge near the lakes have more positive *d-excess* values than areas that are further away from these lakes (Bowen et al., 2012). Conversely, Tap_{Lake} have more negative *d-excess* values, and negative δ^2H residuals with respect to both predicted local precipitation and predicted local surface water, suggesting they have undergone more evaporative losses with its associated fractionation (Gat & Gonfiantini, 1981). Bowen et al. (2007) showed similar patterns of "low *d-excess* regions" around the Great Lakes in the United States, however the sources for those tap water samples were not known. Tap water from lakes can undergo significant evaporation in these regions (Jasechko et al., 2014). Such high evaporative losses can be partially due to tap water management related issues as recent study suggests this region to have very limited evaporative losses (Gibson et al., 2021). Except a few small lakes such as Aspey Lake, Lauzon Lake, Lake Sassagianga and Lake Wawa, most of the Tap_{Lake} samples in these regions are sourced from the Great Lakes. 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 (Jasechko et al., 2014; Jones et al., 2016; Steinman & Abbott, 2013). Long-term seasonal and multi-annual isotopic monitoring of tap waters appears promising in identifying the effect of climate or water management practices on tap water supplied by different sources (e.g., groundwater vs lake water).

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 (Figure 2 and Figure 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 (Geographic, 2020; Zhao et al., 2017). The glacier and snow covered Rockies receive substantial orographic rainfall (mountain effects) (Dansgaard, 1964; Gat, 1996; Hall et al., 2020), 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 (Brooks et al., 2014; Gibson & Edwards, 2002). These δ^2H patterns are consistent with earlier findings in precipitation and surface waters in these regions (Brown, 1971; Gibson et al., 2020). The overall evaporative losses patterns follow the natural evaporative losses found in these regions (Gibson et al., 2021).

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 (Figure 3). Also, 62% of $Tap_{Groundwater}$ samples in both the Prairies and British Columbia have positive δ^2H residuals with respect to both predicted local precipitation and predicted surface water (in red, Figure S3 and Figure 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 (Bowen et al., 2007; Castellazzi et al., 2019). 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 (Jasechko et al., 2014; 2017). 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 (Gibson et al., 2020; St Amour et al., 2005).

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 δ^2H residuals (47% and 63%, respectively) with respect to predicted local precipitation and predicted local surface water (Figure S3 and Figure 7). The majority (~83%) of the total Tap_{River} and Tap_{Lake} samples in these regions also have very negative *d-excess*. The positive δ^2H residuals 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 (Bowen et al., 2007; Gibson et al., 2020; Kendall & Coplen, 2001). The negative δ^2H residuals 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. 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. (2018) 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 δ^2H residuals, with respect to predicted δ^2H values in local precipitation and surface water suggesting significant evaporative losses (Gibson et al. 2020). Such high evaporative losses from rivers and lakes are common in the eastern Prairies (Government of Canada, 2017a; Liu et al., 2014) 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 (Gibson et al., 2016). 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 (Gibson et al., 2020). Small changes in winter precipitation in these regions can have a significant impact on availability of the water resources (Jasechko et al., 2014). 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 (Jameel et al., 2016).

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 (Figure S8). 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 maintain a relatively constant isotopic signature across multiple years and only show small seasonal fluctuations with more negative δ^2H values during snowmelt and more positive δ^2H values during the summer (Rosa et al., 2016; Telmer & Veizer, 2000). 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 much larger range and more abrupt δ^2H variations (Figure S8). These variations likely reflect a switch in water sources by water management companies from surface water to groundwater (Figure S8). Isotopic measurements of tap water are not only useful to quantify the impact of climate and evaporation on the water resources (Du et al., 2019; Wang et al., 2018; Zhao et al., 2017) but also provide a tool to track urban water supply system dynamics (Jameel et al., 2018). 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 resource 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 (Medeiros et al., 2017). The earlier and reduced runoff volume observed in many rivers across Canada can affect adequate water storage and threaten late-summer water availability (Bardsley et al., 2013), particularly in semi-arid regions. Winter streamflow 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 (Bush & Lemmen, 2019). 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 (Bakker, 2009). Evapotranspiration related water losses will also accelerate in the upcoming decades with increased warming (Bush & Lemmen, 2019) 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 (Bakker & Cook, 2011). As seen in other countries, poor water management practices might exacerbate water losses in semi-arid regions (e.g., the Prairies) (Jasechko & Perrone, 2020). It is therefore critical to take into account the specific regional and long-term impacts of water management practices on Canadian water resources (Gleeson et al., 2012; Jasechko & Perrone, 2020). 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 a baseline of isotope values in Canadian tap water for long-term monitoring of climatic and anthropogenic threats to the Canadian tap water resources.

4.5 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 "isotope 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 (Bartelink & Chesson, 2019), tracing the mobility or origin of individuals in cold cases, in certifying food provenance, or in authenticating illegal products origin (Bartelink & Chesson, 2019; Chesson et al., 2020; Fraser et al., 2006). 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 solve cold cases (Fauberteau et al., 2021) and reconstruct individual travel history (Hu et al., 2020).

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 Prairies aquifers. Those resources are vulnerable to the on-going climate change often augmented by poor human management practises. δ^2H values of tap water in those regions demonstrates strong signs of evaporative losses, caused either by natural processes (e.g., mountainous lakes) or by human water management practises (e.g., open water reservoirs). Long-term isotopic monitoring of tap water would be an effective tool to quantify evaporative losses and assess the vulnerability of different water sources to climate and anthropogenic threats. In the Eastern regions of Canada, large rivers and lakes are often the dominant source of tap water resources, and are also vulnerable to rapid climate changes that affect water balance, particularly across the Great Lakes region. Due to the abundance of the water resources in those regions, many municipalities do not consider water loss as a threat. As warming progresses across Canada, effective regional water supply management strategies need to be implemented to limit negative impacts on the water resources. Our isotopic measurements of tap water from across Canada provide a baseline, and established a foundation to develop long term isotope monitoring as a tool to better manage the water resources from source to tap by accounting for vulnerabilities specific to a region or a water source.

Credit authorship contribution statement

CPB designed the project and supervised SAB. MMGC collected all samples, and MMGC and GSJ conducted laboratory analysis of the samples. SAB conducted all the data analysis. CPB and SAB conceptualized model development steps and performed the interpretation. SAB led the writing of the manuscript (original draft) and CPB reviewed and edited the manuscript. YJ contributed to conceptualization and reviewed the manuscript. JG provided the Canadian river isotopes dataset and reviewed the manuscript. All the authors contributed to the article and approved the submitted version.

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