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

By

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Abstract

Tap water supply is an essential resource for human societies. However with increasing water use and global warming, this resource needs to be monitored and managed sustainably. Here we use stable isotopes to identify potential issues associated with tap water resources in Canada. We analyze isotopes of 576 tap water samples collected from across Canada and classified them based on their supply sources including groundwater ($\mathrm{Tap}_{\mathrm{Groundwater}}$), river ($\mathrm{Tap}_{\mathrm{River}}$) and lake ($\mathrm{Tap}_{\mathrm{Lake}}$). We found, isotopic values in tap water correlate strongly with those predicted in local precipitation across Canada, suggesting precipitation is the parent source of tap water. However, this correlation is stronger for $\mathrm{Tap}_{\mathrm{Groundwater}}$ and $\mathrm{Tap}_{\mathrm{River}}$ than $\mathrm{Tap}_{\mathrm{Lake.}}$ To explain this difference, we constructed a series of water balance models to predict isotopic values of surface water across Canada validated against Canadian rivers isotopes data. We then compared the tap water isotopic values to those predicted in local surface water, which improved the predictability of $\mathrm{Tap}_{\mathrm{River}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ but not $\mathrm{Tap}_{\mathrm{Groundwater}}$. We suggest, $\mathrm{Tap}_{\mathrm{Groundwater}}$ usually reflects isotopic values of annually averaged precipitation whereas $\mathrm{Tap}_{\mathrm{River}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ reflect post-precipitation processes. We used the residuals between our observed and predicted isotope data to assess regional sources and processes influencing tap water isotopes across Canada. Regionally, snow/glacier melt from the Rockies contributes to groundwater recharge across Western Canada as well as to some rivers and lakes in Alberta and British Columbia. Also, tap water are highly evaporated across Western Canada irrespective of their sources. Across the Great Lakes and East Coast regions, lakes undergo high evaporative losses. Also, many localities in the East Coast pump and store groundwater in small lakes or ponds exposing them to evaporation. Our data and models provide a baseline for isotope monitoring of tap water resources and isotope forensic studies across Canada.

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List of Abbreviations

$\text{Tap}_{\text{Groundwater}}$	Tap water sourced from wells
$\text{Tap}_{\text{River}}$	Tap water sourced from streams and rivers
Tap_{Lake}	Tap water sourced from small or large lakes, ponds and artificial reservoirs

CHAPTER 1 - INTRODUCTION

Sustainable water resources management has become an increasing challenge across the globe with ongoing global climate change and population growth (Srinivasan et al., 2012). 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). The types of water resources (i.e. river, lake and groundwater), freshwater supplies demand, and water management strategies vary among different provinces in Canada. Therefore, the impact of climate change and growing population on Canadian water resources and tap water supply can vary significantly depending on the regions. With warming, some predicted changes in hydrological cycle include reduced snowpack, loss of glaciers and accelerated evapotranspiration water losses (Bush & Lemmen, 2019). Such changes are modifying the water balance of rivers and lakes across Canada (e.g., reduced summer flow) and affecting tap water supplied to Canadians. For example, across the Canadian Prairies anthropogenic impacts coupled with climate change have already resulted in summer streamflow depletion of major rivers (Schindler & Donahue, 2006) and serious water availability threats (Government of Canada, 2017b). Warming, reduced snow cover and glacier retreat from the Rockies and increased evaporation are predicted to further exacerbate water availability and quality across the Prairies (Bakker, 2009; Schindler & Donahue, 2006). In addition to these natural threats to water availability, Canadian water management practices vary among 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). Given some of these existing water security issues in Canada, there is an urgent need to better trace water from sources to tap water at the national scale. To protect climatically vulnerable water resources, regional water resource issues and potential risks must be clearly identified and monitored.

Naturally occurring stable isotopes of hydrogen and oxygen (δ^2H and δ^{18}O) are powerful tools to trace natural and anthropogenic water cycling processes from precipitation to tap water. The isotopic difference between tap water and its source (e.g., precipitation, groundwater, river and lake) can potentially reveal natural and anthropogenic controls on water availability such as evaporative losses (naturally or from surface reservoirs), recharge seasonality and mixing of isotopically-distinct water sources. In a pioneering study, Bowen et al. (2007) used tap water isotopes to trace regional hydrological processes and characterize regional water issues across the contiguous United States. Since then, tap water isotopic analyses have shown success in various water investigations across the globe, including partitioning regional and seasonal reliance on surface and groundwater for supply, identifying regions extracting fossil groundwater and estimating volumes of inter-basin water transfer (Du et al., 2019; Good et al., 2014; Wang et al., 2018; de Wet et al., 2020).

1.1. Objectives

The overall purpose of this research was to trace water cycling from precipitation to tap water supply and to assess regional tap water vulnerability to ongoing climate warming and potential water management issues. Four specific objectives were defined as:

1) Collect and analyze δ^2H and d-excess in 576 tap water samples collected from across Canada to provide a baseline for nationwide monitoring of important water resources (Bowen et al., 2007; de Wet et al., 2020; Zhao et al., 2017).
2) Compare the tap water isotopes to predicted local precipitation isotopes (Bowen, 2019).
3) Construct a series of water balance models to predict surface water isotopic values validated against Canadian rivers data (Gibson et al., 2020).
4) Interpret residual isotopic values between measured isotope data in tap water and predicted isotope data in precipitation and surface water to assess water sources and water cycle processes (natural and anthropogenic) that influence the Canadian tap water supply

1.2. Thesis Structure

This thesis has been written in an article format, where the body of the thesis consists of 1 independent paper (chapter two) with the intention that it will be submitted to a scientific journal for publication.

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CHAPTER 2 - SPATIAL VARIATIONS IN TAP WATER ISOTOPES ACROSS CANADA

Spatial Variations in Tap Water Isotopes Across Canada

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Key Points:

- Natural and anthropogenic processes cause significant evaporative losses across Canada
- Glacier and snow melt from the Rockies contribute to a large part of tap water sources across Western Canada
- We present the first national level maps of tap water isotopes across Canada with water management and human forensic implications

2.0. Abstract

With global warming and increasing water use, tap water resources need to be monitored and managed sustainably. Here we used stable isotopes to identify potential issues associated with tap water resources in Canada. We analyzed 576 tap water samples collected from across Canada and classified them based on their sources including groundwater ($\mathrm{Tap}_{\mathrm{Groundwater}}$), river ($\mathrm{Tap}_{\mathrm{River}}$) and lake ($\mathrm{Tap}_{\mathrm{Lake}}$). δ^2H in tap water correlate strongly with values predicted for local precipitation across Canada, suggesting precipitation is the parent source of tap water. However, this correlation is stronger for $\mathrm{Tap}_{\mathrm{Groundwater}}$ and $\mathrm{Tap}_{\mathrm{River}}$ than $\mathrm{Tap}_{\mathrm{Lake.}}$ To explain this difference, we constructed a series of water balance models to predict δ^2H of surface water across Canada validated against Canadian rivers isotopes data. δ^2H of local surface water improved the predictability of $\mathrm{Tap}_{\mathrm{River}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ but not $\mathrm{Tap}_{\mathrm{Groundwater}}$. $\mathrm{Tap}_{\mathrm{Groundwater}}$ usually reflects δ^2H of annually averaged precipitation whereas $\mathrm{Tap}_{\mathrm{River}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ reflect post-precipitation processes. We used the residuals between our observed and predicted isotope data to assess regional sources and processes influencing tap water isotopes across Canada. Regionally, snow/glacier melt from the Rockies contributes to groundwater recharge across Western Canada and to some rivers and lakes in Alberta and British Columbia. Tap water are highly evaporated across Western Canada irrespective of their sources. Across the Great Lakes and East Coast regions, lakes undergo high evaporative losses. Also, many localities in the East Coast pump and store groundwater in small lakes or ponds exposing them to evaporation. Our data and models provide a baseline for isotope monitoring of tap water resources and isotope forensic studies across Canada.

2.1. Plain Language Summary

We present a geo-hydrological study of stable isotopes in tap water and surface 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 with those of local precipitation. To understand post-precipitation processes, we constructed water balance models to predict surface water δ^2H across Canada. We compared tap water δ^2H with those predicted in local surface water. Tap water δ^2H exhibit strong correlations with both precipitation and surface water suggesting precipitation supplies most of Canadian tap water. By analyzing the isotopic difference between tap water and precipitation and surface water, we find snow/glacier melt from the Rockies is an important source of water for groundwater recharge across Western Canada

and some rivers and lakes in Alberta and British Columbia. Tap water are highly evaporated across Western Canada regardless of their source. In the Great lakes regions lakes show high evaporation. Many localities in East Coast regions rely on natural and human-made lakes including some storing pumped groundwater on surface reservoirs making them vulnerable to evaporation. We present the first national level maps of tap water isotopes providing a baseline for tap water resources monitoring with forensic applications.

2.2. Introduction

Long term sustainability of water resources has become a concern in Canada due to the rapid ongoing global climate change occurring across the country in combination with 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 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 among 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 management and of the impact of climate change from sources to tap.

Stable isotopes of hydrogen and oxygen (δ^2H and δ^{18}O) 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 tap water isotopes to trace regional hydrological processes and to characterize regional water issues across the contiguous United States. Since then, tap water isotopic analyses have shown success in various water investigations across the globe, including partitioning regional and seasonal reliance on surface and groundwater for supply, and 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 in the water distribution system and to quantify water losses due to evaporation in urban water systems.

Here, we present the first national level isotopic analysis of tap water in Canada. We collected and analyzed δ^2H and d-excess in 576 tap water samples from across Canada, which 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 document their main supply sources using publicly available records to explore risks at source level, based on the hypothesis that vulnerability to climatic change and water management can vary depending on the source type (Wang et al., 2018; de Wet et al., 2020). First we analyzed the tap water isotopic patterns over Canada and compared them with predicted local precipitation isotopic values (Bowen, 2019). We then constructed a series of water balance models to predict modifications expected in surface water isotopic values across Canada. We validated the water balance models by comparing them with river isotopic values collected from across Canada (Gibson et al., 2020), and then compared the tap water isotopic values with those of predicted local surface water. Finally, we analyzed tap water isotopic values, particularly 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. We also underline the value of our tap water and surface water databases for human forensic applications across Canada, as established previously for other regions (e.g., Ehleringer et al., 2008).

2.3. Materials and Methods

2.3.1. 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 ($\mathrm{Tap}_{\mathrm{Groundwater}}$), river ($\mathrm{Tap}_{\mathrm{River}}$) and lake ($\mathrm{Tap}_{\mathrm{Lake}}$). $\mathrm{Tap}_{\mathrm{Groundwater}}$ is defined as tap water sourced from wells. $\mathrm{Tap}_{\mathrm{River}}$ is defined as tap water sourced from streams and rivers. $\mathrm{Tap}_{\mathrm{Lake}}$ is defined as tap water sourced from small or large lakes, ponds 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.

2.3.2. 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 $\delta^{18}O$ 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 $\delta^{18}O$ using a GasBench II (Thermofisher, Bremen, Germany) with a Delta^{+}XP isotope ratio mass spectrometry (IRMS; Thermofisher, Bremen, Germany). For $\delta^{2}H$ 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 $\delta^{2}H$ using the same GasBench II with a Delta+XP IRMS as for $\delta^{18}O$. Several replicates of three internal water reference materials (RMs) were included in each analysis sequence: W-7 ($\delta^{2}H = -198.5 \pm 2.0$ ‰ and $\delta^{18}O = -24.55 \pm 0.2$ ‰), W-10 ($\delta^{2}H = -85.9 \pm 2.0$ ‰ and $\delta^{18}O = -11.84 \pm 0.2$ ‰) and W-9 ($\delta^{2}H = +11.3 \pm 2.0$ ‰ and $\delta^{18}O = -5.06 \pm 0.2$ ‰). These internal water RMs are traceable to the VSMOW scale via calibration against VSMOW ($\delta^{2}H = 0$ ‰, $\delta^{18}O = 0$ ‰ (Brand et al., 2014)), GISP ($\delta^{2}H = -189.5 \pm 1.2$ ‰, $\delta^{18}O = -24.76 \pm 0.09$ ‰ (IAEA, 2007))); and SLAP ($\delta^{2}H = -428$ ‰, $\delta^{18}O = -55.5$ ‰ (Brand et al., 2014)). Tap water $\delta^{2}H$ and $\delta^{18}O$ values were obtained using the LIMS for Light Stable Isotopes (Shrestha & Yesha, 2017). A water QC material, W-20 ($\delta^{2}H = -5.9 \pm 2.0$ ‰ and $\delta^{18}O = -7.34 \pm 0.2$ ‰), was also included in every analysis sequence. The analytical precision (2σ) of the $\delta^{2}H$ and $\delta^{18}O$ 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 $\delta^{2}H$ and $\delta^{18}O$ 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.

2.3.3. Spatial patterns of tap water isotopes and comparison with precipitation $\delta^{2}H$ values

To analyze the spatial variability of tap water isotopes across Canada, we mapped the $\delta^{2}H$ and d-excess (d) in ArcGIS Pro (with $d = \delta^{18}O - 8 * \delta^{2}H$; deviation from this equation is an indicator for post-precipitation isotopic fractionation as a result of evaporative water loss (Dansgaard, 1964)) (Figure 2 and Figure 3). 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. We also downloaded the layers predicting δ^2H in precipitation (Bowen, 2019) to analyze the correlation between the observed isotopic values of tap water (separating them as a function of their sources) and the predicted isotopic values of local precipitation (Table 2, Figure 5 and Figure S1). We extracted the predicted isotopic values in precipitation both seasonally and annually at each tap water site. As in previous studies (e.g., Bowen et al., 2007), comparing isotopic values in tap water and precipitation provide primary insights into how water cycles from its local precipitation source to the consumer faucet. 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).

2.3.4. 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 δ^2H model that only accounts for atmospheric controls of isotopic variability, water balance models incorporate isotopic variability 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 1^{-}km^2 resolution using the equations in Table 1 within the North America boundary defined by the HydroSHEDS dataset (Figure 1). We accumulated upstream Q and δQ using digital topography map with drainage direction from HydroSHEDS and the “Flow Accumulation” tool

(Spatial Analyst Toolbox; 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 isotopic values in precipitation weighed by total precipitation (P), or (2) by propagating isotopic values in precipitation weighed by effective precipitation (P-ET) (Table 1). The second method aimed to quantify whether accounting for spatial evapotranspiration (ET) variations improves the estimated predicted isotopic 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 1^2 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 or δ^{18}O)**
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	Q = Nov (P - ET) +…. Apr	δQ = (Nov (P - ET) * Nov δP) +….(Apr (P -

Weighted Winter ET Model	(P-ET)	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

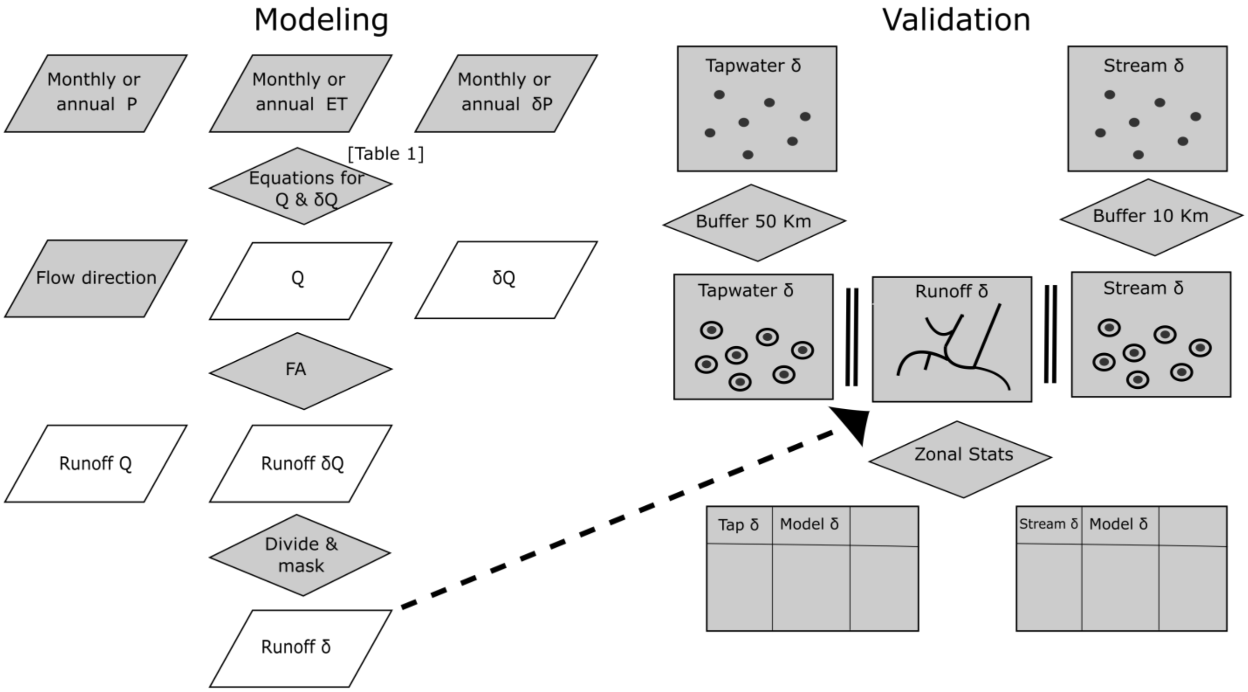

Figure 1. Workflow for GIS based water balance modeling and validation modified from (Bowen et al., 2011). Diamond = operations and rectangular (shaded) = input raster data sets. P = precipitation; ET = evapotranspiration; δP = isotopic composition of precipitation; FA = flow accumulation; Q = discharge,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

2.3.5. 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 streams isotopes dataset (Gibson et al., 2020). However, the latitude and longitude of the river water collection sites are not always lined-up with the Hydroshed. In other words, if we extracted the isotopic value of the water balance models for the pixel located at the collection site, the value extracted might not correspond to the exact river.

Most of the river samples by Gibson et al. (2020) are large streams or rivers and are distinct on the Hydroshed. In order to compare the observed isotopic values in streams with the predicted isotopic values in local surface water derived from our different models, we first masked all the pixels with total drainage areas <9 km^2 to exclude small streams (Figure 1) (Bowen et al., 2011). We then extracted the predicted isotopic 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 isotopic value within this area. We compared the annual models with observed annual average streams isotopic values at 262 sites, the summer models with observed summer average streams isotopic values at 241 sites, and the winter models with observed winter average streams isotopic values at 217 sites (Figure S4). We validated all the models based on the significantly positive linear correlation between the observed isotopic values in streams and the predicted isotopic values in local surface water from our different models (Figure S4).

2.3.6. Comparison between tap water δ^2H values and predicted local surface water δ^2H values

We then compared the observed isotopic values in tap water with the predicted isotopic values in local surface water derived from our different models (Table 3). We used a similar approach as described above but applied a larger 50 km radius circular buffer around each of the 576 tap water sampling sites (Figure 1). A larger radius is used in this case because the exact source of tap water is not always easy to locate and some large cities use more distant reservoir as main water sources (e.g., Vancouver, Calgary). We explored the correlation between the observed isotopic values in tap water and the predicted isotopic values in local surface water from our different models (Table 3, Figure 6 and Figure S2).

2.3.7 Residuals analysis

Lastly, we extracted and mapped the residuals between the observed isotopic values in tap water and predicted isotopic values in local annual precipitation (monthly weighted) (Figure S3). We also analyzed residuals between the observed isotopic values in tap water and predicted isotopic values in local annual surface water based on our Monthly Weighted Annual ET Model (Figure 7) to explore natural and anthropogenic processes imposing potential threats to tap water sources at regional level.

2.4. Results

2.4.1. Spatial patterns of δ^2H measurements in Canadian tap water

δ^2H values in tap water ranges from -188‰ to -33‰ (Figure 2). There are strong spatial patterns of decreasing δ^2H values from low latitude coastal regions towards high latitude and high-altitude inland regions (Figure 2). Generally, the lowest δ^2H values occur in Western Canada (mountainous regions) and the highest δ^2H values occur in the Eastern Canada's coastal and Great Lakes regions irrespective of tap water sources (Figure 2). The d-excess values of tap water (i.e., $d = \delta^2H – 8*\delta^{18}O$) also show large spatial variability and ranges from -35.3‰ to +19.1‰ (Figure 3). The general patterns show low d-excess dominates 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 high 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 $\text{Tap}_{\text{Groundwater}}$ and Tap_{Lake} respectively (Figure 3).

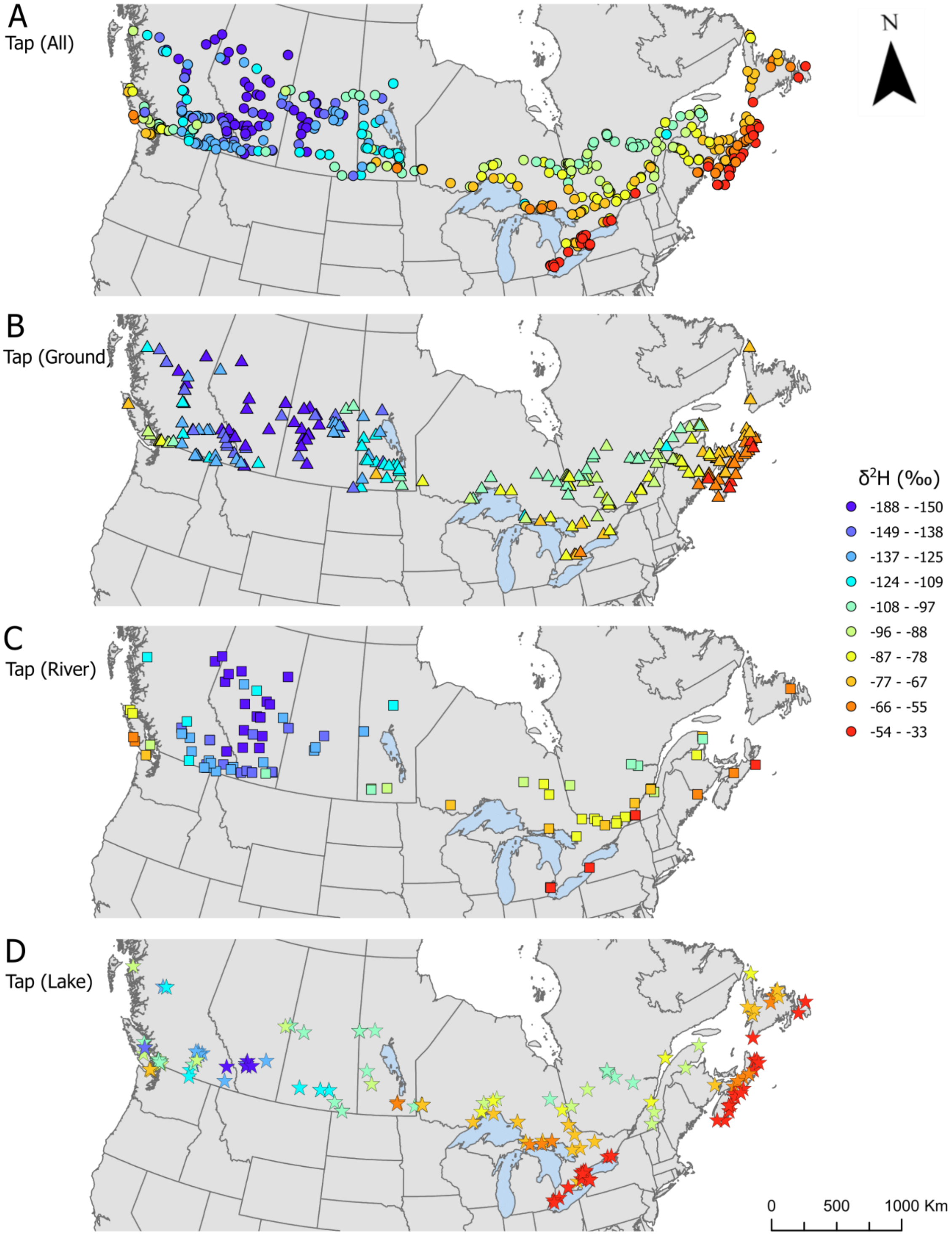

Figure 2. Spatial distribution of sample locations and δ^2H values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced primarily from groundwater (n=281), c: tap water sourced from rivers (n=118) and d: tap water sourced from lakes (n=177). Classification between groundwater, river, and lake is detailed in the Methods.

Administrative boundaries are from http://www.naturalearthdata.com/. This map was generated in ArcGIS Pro using Lambert conformal conic map projection.

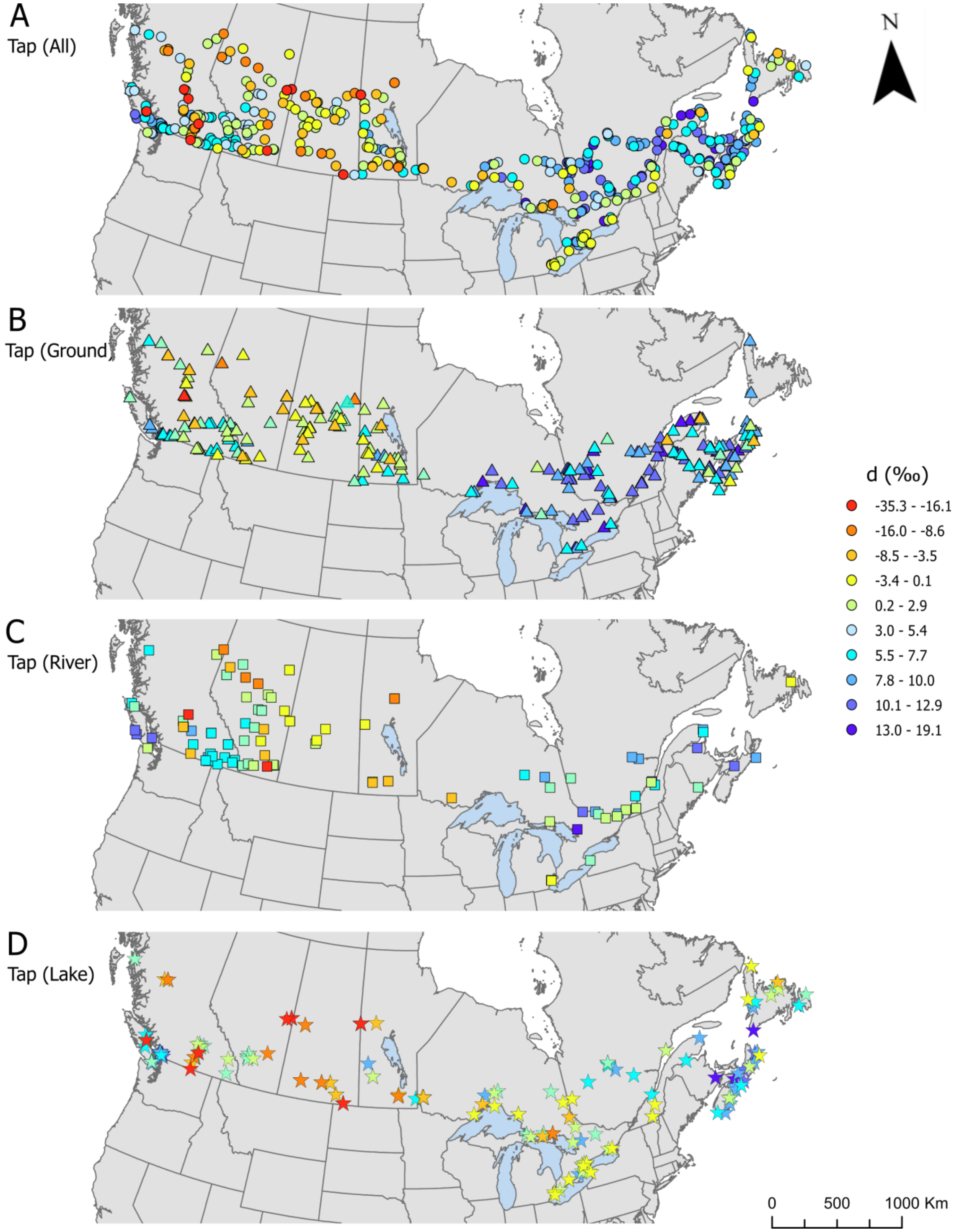

Figure 3. Spatial distribution of sample locations and d-excess (d) values in tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced primarily from

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 ArcGIS Pro using Lambert conformal conic map projection.

2.4.2. Relationship between tap water δ^2H values and precipitation δ^2H values

The isotopic 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). When plotting the δ^2H in tap water grouped by their pre-classified water sources, δ^2H values of $\text{Tap}_{\text{Groundwater}}$ and $\text{Tap}_{\text{River}}$ have a much stronger correlation with local precipitation than δ^2H values of Tap_{Lake} whether annually or seasonally (Table 2, Figure 5 and Figure S1). When accounting for precipitation seasonality, δ^2H values of Tap_{Lake} have a stronger correlation with summer precipitation, yet, they remain much less predictable relative to other sources. The correlation between δ^2H values of tap water and that predicted for winter precipitation is much weaker irrespective of the tap water source types (Table 2). Although the δ^2H values of monthly weighted annual precipitation and monthly weighted summer precipitation models have the strongest correlation with the δ^2H values of tap water, the monthly weighted annual precipitation model displays the best performance (i.e., closer to 1:1 line) (Figure 5 and Figure S1).

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

Tap sources	δ^2H	Monthly Weighted Annual precipitation	Monthly Weighted Summer precipitation	Monthly Weighted Winter precipitation
		R^2	R^2	R^2
All	δ^2H	0.79	0.81	0.69
Groundwater	δ^2H	0.86	0.87	0.79
River	δ^2H	0.86	0.88	0.76

Lake	δ^2H	0.62	0.67	0.45

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

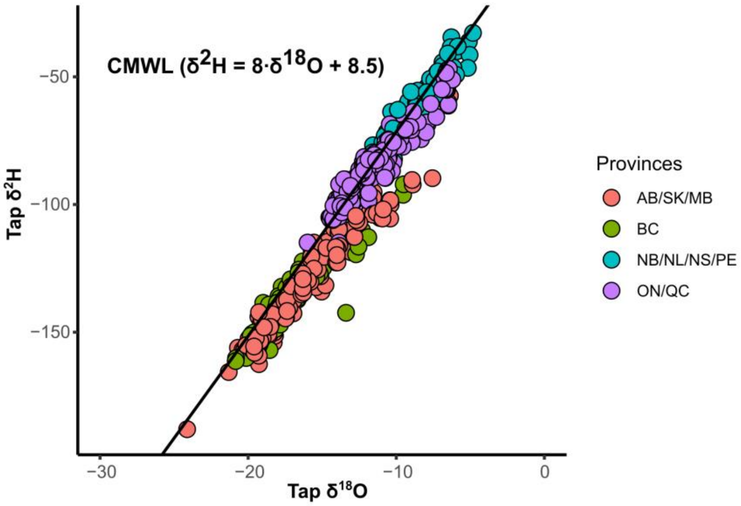

Figure 4. Covariation of tap water δ^2H and δ^{18}O values (n = 576) in relation to Canadian meteoric water line (CMWL) (Gibson et al., 2020).

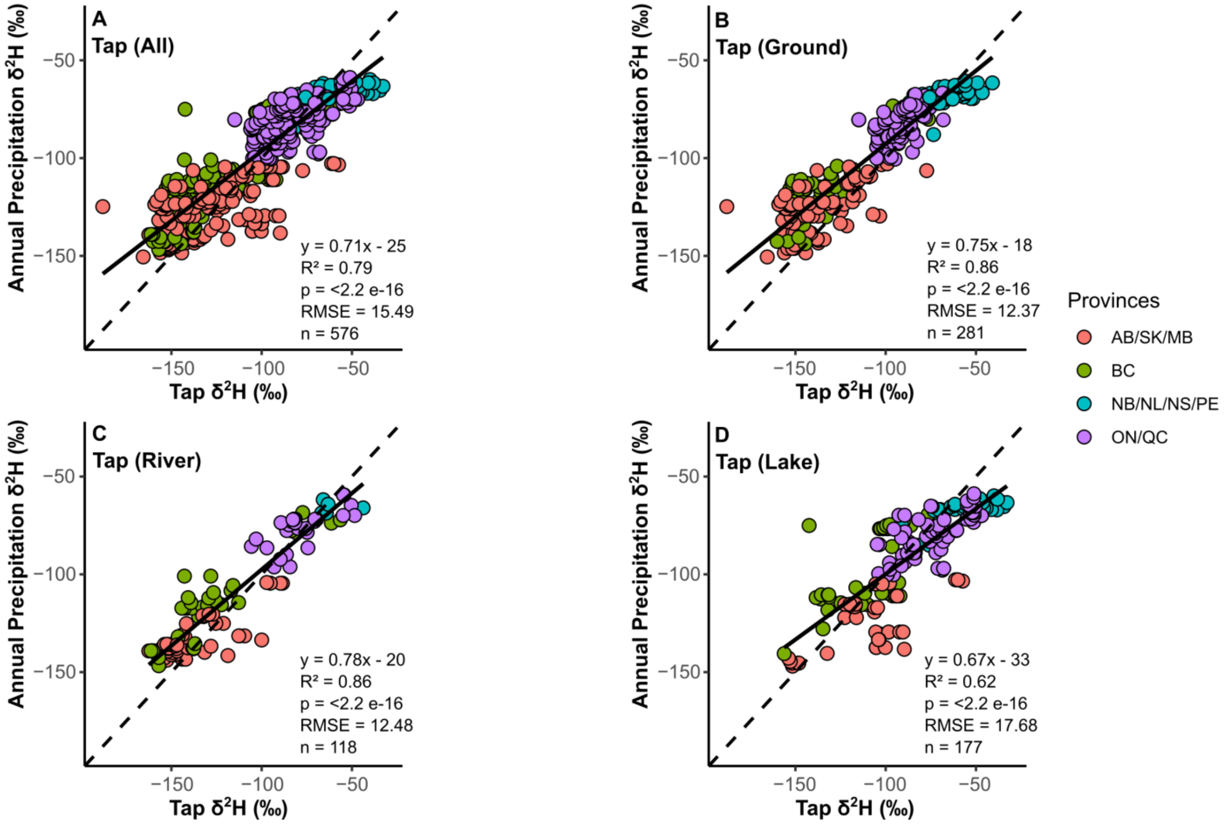

Figure 5. Correlation between tap water δ^2H values and monthly weighted local annual precipitation δ^2H values. a: all the tap water samples combined, b: tap water sourced primarily from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1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

2.4.3. 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 observed δ^2H values in streams and predicted δ^2H values in surface water whether annually or seasonally (Figure S4) validating our water balance modelling approach. When looking more closely at individual models, the monthly weighted annual models perform better (i.e., closer to 1:1 line) than annual average models and seasonal models. For these models, the relationship between predicted and observed values fall close to the 1:1 line except for the semi-arid regions of Alberta, Saskatchewan and Manitoba (the Prairies) where the observed stream values are consistently higher than the predicted values. Overall, the Monthly Weighted Annual ET model displays the best performance. Models predicting isotopic values in the winter always underperform relative to those predicting isotopic values annually or in the summer (Figure S4). To analyze why observed winter streams have a much weaker correlation with the monthly weighted winter models, we compared the observed δ^2H values in winter streams with both local winter precipitation δ^2H and local 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).

2.4.4. 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). When plotting the δ^2H values in tap water grouped by their pre-classified water sources, the water balance models do not improve $\text{Tap}_{\text{Groundwater}}$ δ^2H prediction (Table 3 and Figure 6) relative to the precipitation-only model (Table 2 and Figure 5). Conversely, the water balance models improve the prediction of $\text{Tap}_{\text{River}}$ 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. The monthly weighted summer models perform much better than the monthly weighted winter models. However, overall the Monthly Weighted Annual ET model displays the best performance.

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

Tap sources	δ^2H	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	δ^2H	0.81	0.81	0.76	0.81	0.81	0.75	0.78	0.78
Groundwater	δ^2H	0.84	0.84	0.83	0.84	0.84	0.83	0.84	0.84
Rivers	δ^2H	0.89	0.90	0.83	0.90	0.90	0.82	0.85	0.85
Lakes	δ^2H	0.73	0.74	0.60	0.73	0.73	0.60	0.65	0.65

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

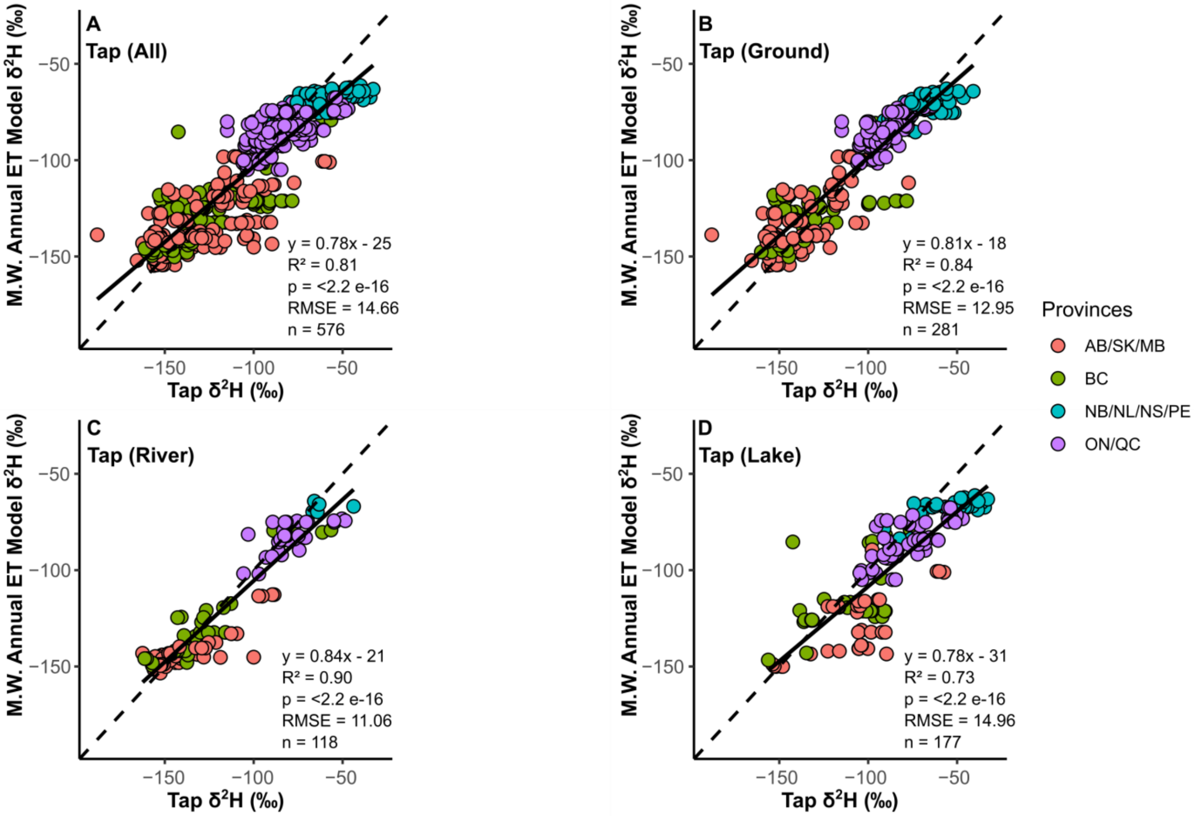

Figure 6. Correlation between tap water δ^2H values and local predicted surface water δ^2H values (based on the Monthly Weighted Annual ET Model). a: all the tap water samples combined, b: tap water sourced primarily from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes.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

2.4.5. δ^2H Residuals across Canada

We present the residual isotopic values between tap water and local annual precipitation (monthly weighted) (Figure S3) and residual isotopic values between tap water and local annual surface water based on our Monthly Weighted Annual ET Model (Figure 7). Large scale residual patterns show $\text{Tap}_{\text{Groundwater}}$ have lower isotopic values than the isotopic values predicted in local precipitation and local surface water (positive residuals, Figure S3 and Figure 7) across the Prairies and British Columbia. $\text{Tap}_{\text{River}}$ and Tap_{Lake} have higher isotopic values than the isotopic values predicted in local precipitation and local surface water (negative residuals, Figure S3 and Figure 7) across Saskatchewan and Manitoba. However, $\text{Tap}_{\text{River}}$ and Tap_{Lake} have a mix of positive and negative residuals across Alberta and British Columbia. The Great Lakes and East

Coast regions are dominated by negative residuals for Tap$_{\mathrm{River}}$ and Tap$_{\mathrm{Lake}}$, with Tap$_{\mathrm{Lake}}$ having the largest negative residuals. Conversely, Tap$_{\mathrm{Groundwater}}$ in the East Coast regions have some small positive residuals.

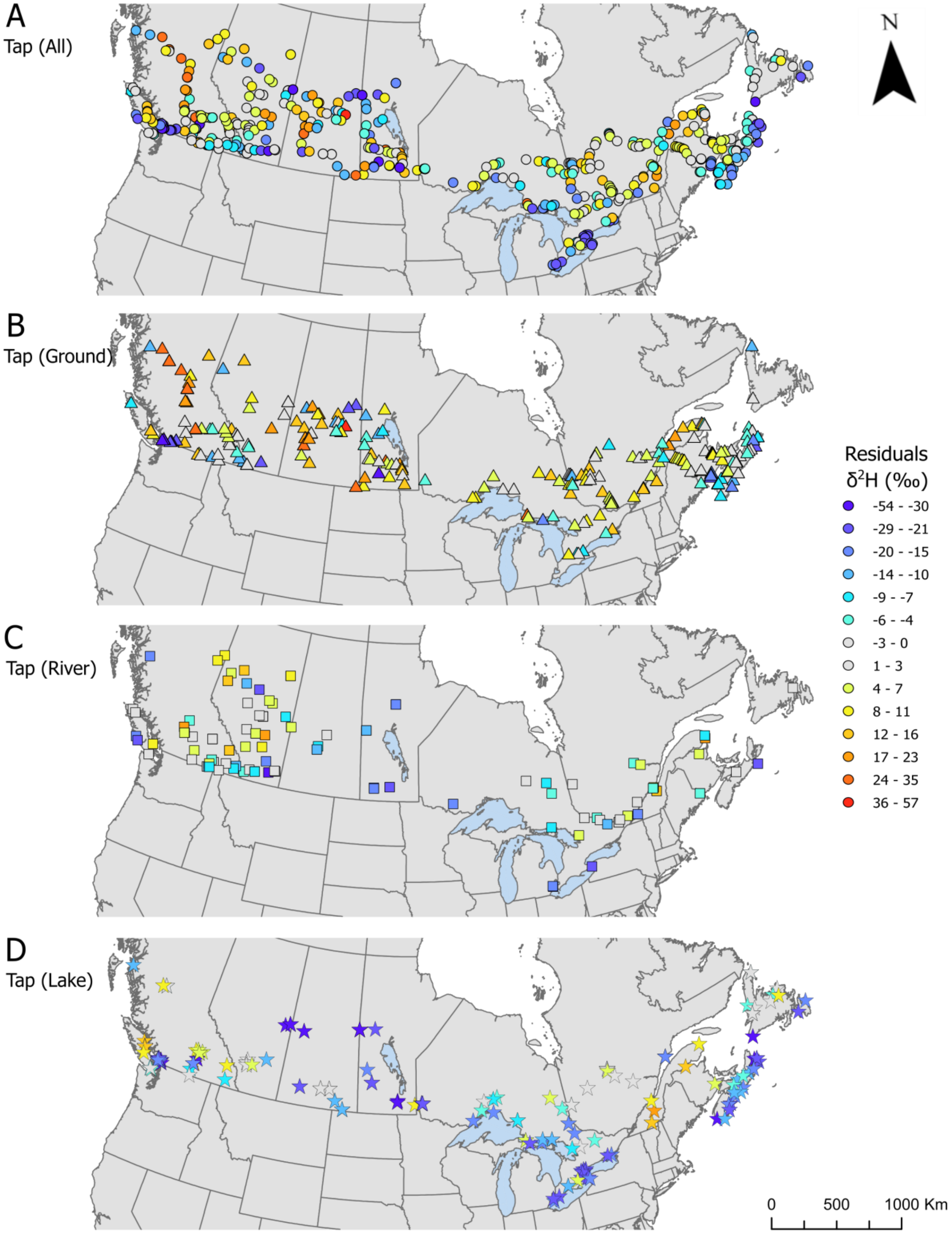

Figure 7. Residuals of δ^2H values between predicted local surface water (based on the Monthly

Weighted Annual ET Model) and tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced primarily 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 ArcGIS Pro using Lambert conformal conic map projection.

2.5. Discussion

2.5.1. General patterns of tap water and its relationship to local precipitation and local surface water

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 isotopic composition in Canada as seen in other studies (Bowen et al., 2007, 2011; Stahl et al., 2020; Wang et al., 2018). This also suggests use of non-local water (e.g., water diversion) is perhaps not an issue for Canadian regions. Water balance model (annual/summer) improves the predictability of δ^2H values in tap water sourced from rivers and lakes ($\text{Tap}_{\text{River}}$ and Tap_{Lake}) but not those sourced from groundwater ($\text{Tap}_{\text{Groundwater}}$) (Figure 6, Figure S2 and Table 3). It suggests that a comparison between the δ^2H values in tap water sourced from surface water (i.e. $\text{Tap}_{\text{River}}$ and Tap_{Lake}) with the δ^2H values predicted in local surface water provides insights into post precipitation processes. The water balance modeling approach described above does not account for isotopic fractionation due to evaporation. It also does not account for infiltration. As infiltration rates can vary seasonally, this might also influence the predicted isotopic values. In this study, we quantified the residual isotopic values between our predicted local surface water and observed tap water (Figure 7). We interpreted the trends and patterns in these residuals as reflecting either evaporative losses or other processes not accounted for in the water balance modeling (Bowen et al., 2011).

2.5.2 Regional patterns

East Coast regions (New Brunswick, Nova Scotia, Prince Edward Island and Newfoundland)

In the East Coast regions, high δ^2H values and moderate to high 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. We found some small positive residuals between isotopic values in tap water and predicted isotopic values in local precipitation and surface water (in red, Figure S3 and Figure 7). These positive residuals are mainly for $\mathrm{Tap}_{\mathrm{Groundwater}}$ (~38% or 26 out of 69 $\mathrm{Tap}_{\mathrm{Groundwater}}$ samples) and some $\mathrm{Tap}_{\mathrm{River}}$ (~33% or 2 out of 6 $\mathrm{Tap}_{\mathrm{River}}$ samples) and $\mathrm{Tap}_{\mathrm{Lake}}$ (~17% or 6 out of 36 $\mathrm{Tap}_{\mathrm{Lake}}$ samples). Similarly, Gibson et al. (2020) also observed such positive residuals when measuring isotopes in the eastern rivers and suggested that evaporation into humid oceanic air masses can lead to isotopic enrichment of surface waters along high slope evaporation lines.

However, ~36% or 25 out of 69 $\mathrm{Tap}_{\mathrm{Groundwater}}$ samples in the East Coast regions have low d-excess values (< 8.5) (Figure 3) and ~62% or 43 out of 69 $\mathrm{Tap}_{\mathrm{Groundwater}}$ samples have higher isotopic values than the isotopic values predicted in local precipitation and surface water (negative residuals in blue, Figure S3 and Figure 7) indicating significant evaporative losses. By looking individually at each of these anomalous samples, we found that some of the tap water sources were probably misclassified in these municipalities and that additional human management practices contributed to evaporative losses. For example, we found that many localities in these regions pump and store groundwater in open surface reservoirs and small lakes or ponds. With climate warming, such water management strategy could lead to high evaporative losses of exploited groundwater.

Approximately 58% or 21 out of 36 $\mathrm{Tap}_{\mathrm{Lake}}$ samples also display low d-excess (<8.5) and ~83% or 30 out of 36 $\mathrm{Tap}_{\mathrm{Lake}}$ samples have higher isotopic values than the isotopic values predicted in local precipitation and surface water (negative residuals, mainly in Newfoundland and Nova Scotia) suggesting these coastal lakes are undergoing significant evaporative losses. Most of these samples originate from small lakes or artificial ponds such as Lake George, Little Lake, Sand Lake, Landrie Lake, Lake Major and Rodney Lake for which higher evaporative losses is expected. A lot of these lakes are used to supply water to small towns or communities. These water management practices in small communities are not optimal for water security in a warming climate with increasing evaporative losses.

The Great Lakes regions (Ontario and Quebec)

In the Great Lakes regions, high isotopic values dominate for tap water similar to what is seen in precipitation (Brown, 1971). However, these tap water samples show an interesting combination of high and low d-excess values for $\mathrm{Tap}_{\mathrm{Groundwater}}$ and $\mathrm{Tap}_{\mathrm{Lake}}$ respectively. $\mathrm{Tap}_{\mathrm{Groundwater}}$ have d-excess similar to those found in precipitation in these regions, suggesting limited evaporative losses (Gibson et al., 2020). The high d-excess of the $\mathrm{Tap}_{\mathrm{Groundwater}}$ reflects the amount of recycled water fluxes ('lake-effect' precipitation events) in the Great Lakes regions suggested by earlier studies (Gat et al., 1994; Machavaram & Krishnamurthy, 1995). Aquifers that recharge near the lakes have higher d-excess values than areas that are further away from these lakes (Bowen et al., 2012).

Conversely, $\mathrm{Tap}_{\mathrm{Lake}}$ have lower d-excess as well as higher isotopic values than the isotopic values predicted in local precipitation and surface water (negative residuals) because they have undergone more evaporative losses associated with kinetic 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. Our study reinforces the idea that at source level lakes are undergoing significant evaporation in these regions (Jasechko et al., 2014). Except 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. The risks and issues associated with these water resources with climate change plays over longer timescale and require a good understanding of the long-term water balance of the Great Lakes (Jasechko et al., 2014; Jones et al., 2016; Steinman & Abbott, 2013). We found little information about regional water management practices around the Great Lakes regions. For example, whether it is a common practice to store water in small open reservoirs contributing to additional evaporation and how current water management practices impact the overall water budget of the Great Lakes. However, isotopic monitoring appears promising in identifying significant changes to these water resources with changes in climate or water management practices as well as in distinguishing water supply specific threats at source level (e.g., groundwater vs lake water).

The Prairies (Alberta, Saskatchewan and Manitoba) and British Columbia regions

As we move toward the Prairies, tap water isotopic values decrease and are generally associated with low 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 lowest isotopic values in our dataset. These mountainous regions also display low d-excess values, suggesting substantial evaporative losses as expected with continental and seasonal climate patterns (Brooks et al., 2014; Gibson & Edwards, 2002). These isotopic patterns are consistent with earlier findings in precipitation and surface waters in these regions (Brown, 1971; Gibson et al., 2020).

$Tap_{Groundwater}$ across the Prairies and British Columbia

Although we generally presume groundwater sources to be more sheltered from evaporation, across the Prairies ~96% or 81 out of 84 $Tap_{Groundwater}$ samples and across British Columbia 92% or 46 out of 50 $Tap_{Groundwater}$ samples have low d-excess (Figure 3). Also, in the Prairies 62% or 50 out of 81 $Tap_{Groundwater}$ samples and in British Columbia 62% or 31 out of 50 $Tap_{Groundwater}$ samples have lower isotopic values than the isotopic values predicted in local precipitation and surface water (positive residuals in red, Figure S3 and Figure 7). The lower δ^2H values in $Tap_{Groundwater}$ suggest two possibilities, recharge contribution from winter precipitation and mountain water (snow/glacier melt runoff). Strong water contribution from mountains is well-established across the semi-arid regions of North America (Bowen et al., 2007; Castellazzi et al., 2019). Also, in Canada, winter precipitation has been reported as major contributor in recharging Prairies aquifers (Jasechko et al., 2017). Our study suggests that groundwater aquifers in British Columbia are also dependent on mountainous runoff (snow/glacier melt). However, the low 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 rate in arid regions (Gibson et al., 2020; St Amour et al., 2005).

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 and negative residuals with the isotopic values predicted in local precipitation and surface water (Figure S3 and Figure 7). ~53% or 36 out of 68 Tap_{River} samples have positive residuals and ~47% or 32 out of 68 Tap_{River} samples have negative residuals across Alberta and British Columbia. Similarly ~37% or 19 out of 52 Tap_{Lake} samples have positive residuals and ~63% or 33 out of 52 Tap_{Lake} samples have negative residuals across Alberta and British Columbia. ~83% of the Tap_{River} and Tap_{Lake} samples in these regions also have very low d-excess. An exception is some of the samples in British Colombia, which have higher d-excess (20 samples and mainly river and reservoirs) reflecting the higher relative humidity in coastal setting.

The positive residuals combined with low d-excess in Alberta and British Columbia is similar to what was seen for the $Tap_{Groundwater}$ across the 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). In addition, the negative residuals in these regions also suggest further evaporative losses from rivers and lakes. From 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 -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 climate change. The impact of climate change to these surface reservoirs and small lakes need to be further investigated to support water security in British Colombia.

Tap_{River} and Tap_{Lake} in Manitoba and Saskatchewan

As we go further to the Rockies, Tap_{River} and Tap_{Lake} of Manitoba and Saskatchewan show only negative residuals with the isotopic values predicted 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 and often exceed local precipitation (Government of Canada, 2017a; Liu et al., 2014) making these regions highly dependent of snowmelt and winter recharge. High evaporative losses occur along the path of large rivers throughout the Prairies (e.g., Athabasca River) from the slow circulation of waters from open surface reservoirs such as lakes or peatlands (Gibson et al., 2016). These evaporation mechanisms in the uplands or valleys lead to highly evaporated isotope signature and low d-excess in these regions (Gibson et al., 2020). Small changes in winter precipitation in these regions will have a significant impact on availability of the water resources (Jasechko et al., 2014). From the Prairies, out of 37 Tap_{Lake} samples at least 12 are human made reservoirs. Also some of the $\text{Tap}_{\text{Lakes}}$ samples are sourced from small lakes such as Cold Lake, Douglas lake, Meadow Lake, Nickel Lake and Shoal Lake.Water management practices need to take into account these regional particularities avoiding storage in open reservoirs to limit evaporative losses (Jameel et al., 2016).

2.5.3. 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 reduction in snow and ice cover will alter the water balance of many watersheds (Medeiros et al., 2017). The earlier and reduced runoff volume seen 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 glacier 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 depending on these water sources (Bakker, 2009). Evapotranspiration related water losses will also accelerate in the next decades with increasing warming (Bush & Lemmen, 2019) further modifying the water balance of rivers and lakes that are often critical for human water supply throughout Canada.

Warming and population growth will also continue to weigh on tap water demand across Canada. As seen in other countries, poor water management practices might exacerbate water losses in semi-arid regions (e.g., the Prairies) (Jasechko & Perrone, 2020). Some of these regions are critical for Canadian food security (e.g., the Prairies). It is therefore critical to take into account the regionally specific and long-term impacts of water management practices on Canadian water resources (Gleeson et al., 2012; Jasechko & Perrone, 2020). Where necessary, water management plan should integrate these regional water balance considerations in their water management plan. However, such regional considerations are often limited by the fragmented and localized water governance (Bakker & Cook, 2011). Isotopic monitoring is an easy and cost-effective approach to identify early climatic and hydrologic changes to the water resources at the regional scale from water sources to the tap. Our models and databases will contribute to improving water resources management effort across Canada (CWA, 2017) by providing an isotopic baseline tool for ongoing monitoring. However, a limitation of our tap water isotopes dataset is sampling bias in summer season which provides a baseline for only summer season. Therefore, we recommend long-term multi-year collection of isotope data to conduct more specific assessments of climatic and anthropogenic threats to the Canadian tap water resources in different regions. We also recommend future work to understand how glacier retreat might impact water availability in the western Canadian regions (e.g., partitioning of how much water is derived from annual snowmelt and annual glacier ice melt).

2.5.4. Forensic application

In addition to its potential use in water resource monitoring, our database is also a valuable tool in forensic studies. Tap water is the source of many manufactured products from food to explosive precursors or drugs. The isotopic values of tap water are usually transmitted to these organic or inorganic materials providing an "isotope fingerprint" to trace their origin. For example, a strong relationship exists between the isotopic 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). Hydrogen and oxygen isotopes in tap water has been critical to trace the mobility or origin of individuals in cold cases, in certifying food provenance, or in authentifying 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 organic and inorganic material in forensic studies 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).

2.6. Conclusions

Our study suggests that precipitation is the primary source of tap water across Canada unlike some western US regions where they largely rely on non-local water (e.g., importing water through inter-basin transfer). 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 demonstrate 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.

2.7. Credit authorship contribution statement

CPB designed the project and supervised SAB. MMGC and GSJ conducted laboratory analysis of the samples. SAB conducted all the data analysis. 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 approved the submitted version.

2.8. Acknowledgments

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Abdi and Paul Middlestead) at University of Ottawa for assisting with laboratory analysis. All data to verify the conclusions of this work have been made available. The tap water isotope dataset is available in the supporting information. 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 rivers isotopes data that were used for models validations can be requested from Dr. John Gibson (jjgibson@uvic.ca) at University of Victoria.

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CHAPTER 3 – CONCLUSION

Tap water isotopes exhibit strong correlation with both local precipitation and local surface water suggesting that precipitation is the primary source of water for tap water supply. However, across Western Canada the consistent positive isotopic residuals between tap water and local environmental water (precipitation and local surface water) suggests that snow and glacier melt from the Rockies is an important source for groundwater recharge across the Prairies and British Columbia as well as for some rivers and lakes in Alberta and British Columbia. Notably, low d-excess in tap water across these regions of Western Canada irrespective of their sources (groundwater, river and lake) is alarming for evaporative losses. Canada has already experienced significant glacier loss and as warming progresses further reduction in glacier from the Rockies and changes in rainfall patterns are expected. Particularly, the Prairies are already very dry and faced with water availability challenges. With warming, an increase in temperature will lead to further evaporative water losses from across Western Canada. Also further reduction in snow and glacier from the Rockies will affect water resources that are dependent on the runoff contribution from the Rockies.

Similarly, low d-excess and negative residuals across the Great Lakes and East Coast regions suggest substantial evaporative losses from surface water sources. However, in the East Coast regions many groundwater sources also show low d-excess and negative residuals suggesting even these groundwater sources are undergoing significant evaporation. This is largely due to pumping and storing groundwater in small lakes or ponds. In addition, many small towns and communities in the East Coast regions are dependent on small lakes (natural) for tap water supply. With ongoing warming and accelerated evaporative losses these communities can experience significant water availability threats. As warming progresses across Canada, effective regional water supply management strategies need to be implemented to limit negative impacts on the water resources. Our study provides the first national level maps of isotopic values in tap water providing an isotopic baseline to monitor tap water resources across Canada. Also, our databases have implications for forensic studies across Canada.

CHAPTER 4 – SUPPLEMENT INFORMATION

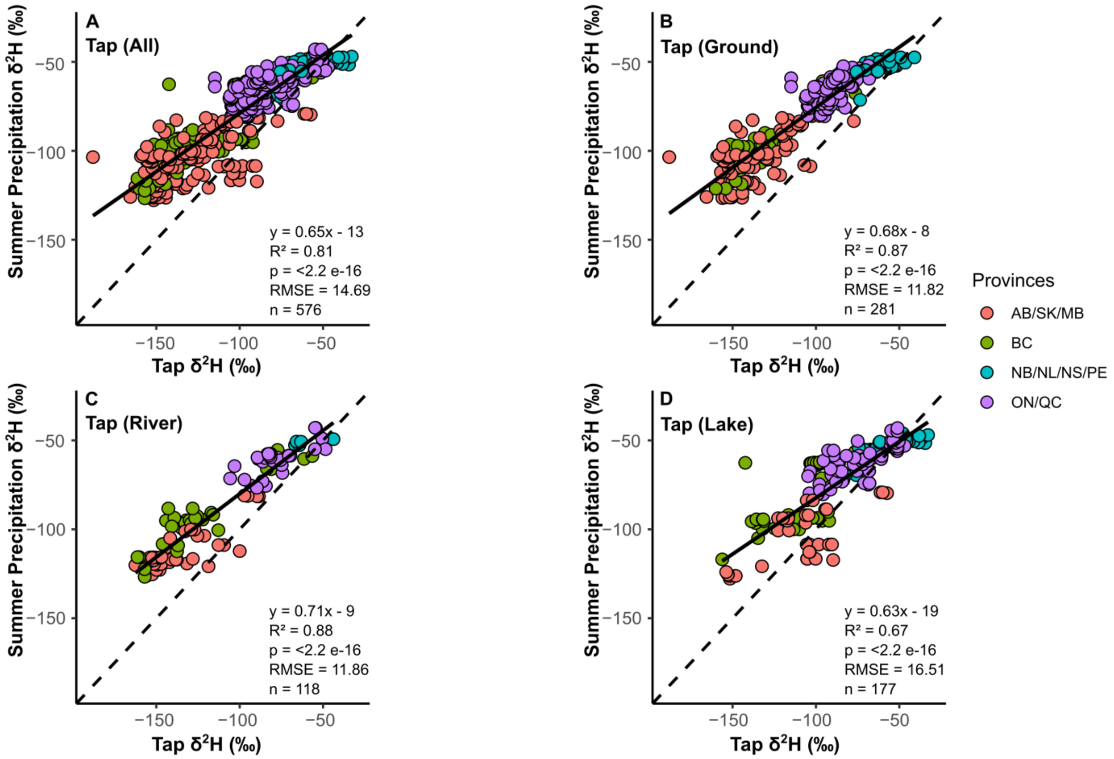

Figure S1. Correlation between tap water δ^2H values and monthly weighted local summer precipitation δ^2H values. a: all the tap water samples combined, b: tap water sourced primarily from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1 line. The black line represents the best fit linear model.

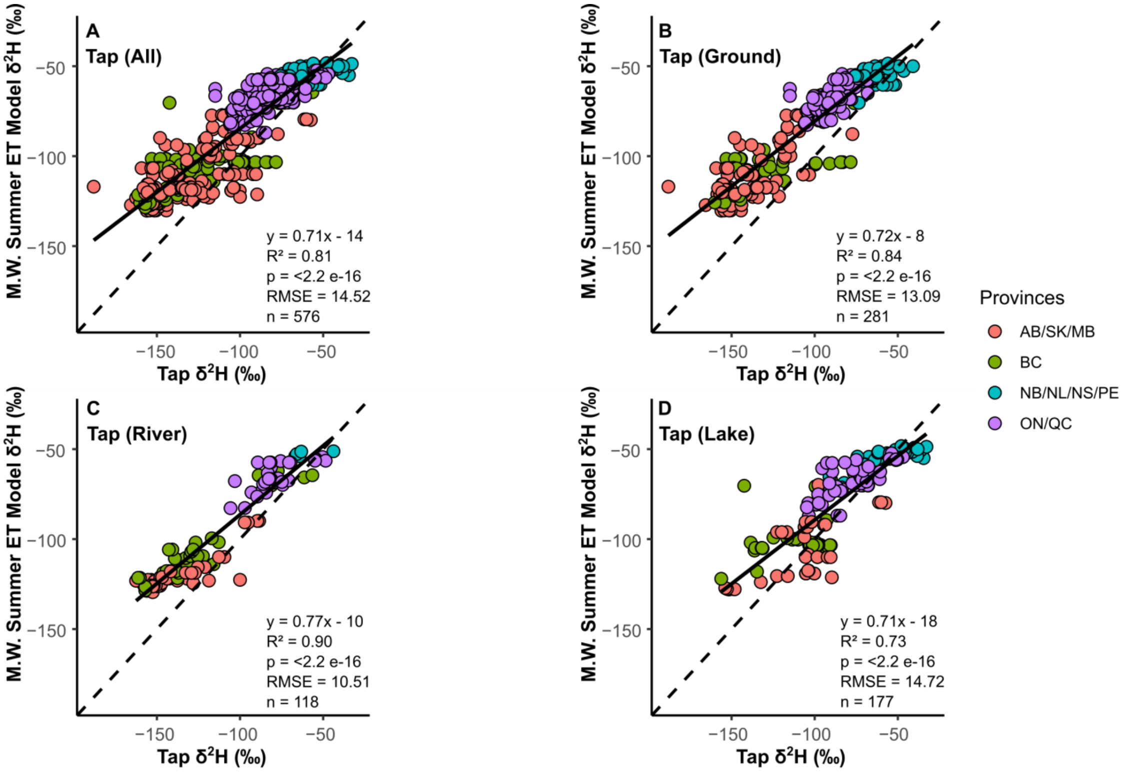

Figure S2. Correlation between tap water δ^2H values and local modelled surface water δ^2H values (based on the Monthly Weighted Summer ET Model). a: all the tap water samples combined, b: tap water sourced primarily from groundwater, c: tap water sourced from rivers and d: tap water sourced from lakes. The dash line represents the 1:1 line. The black line represents the best fit linear model.

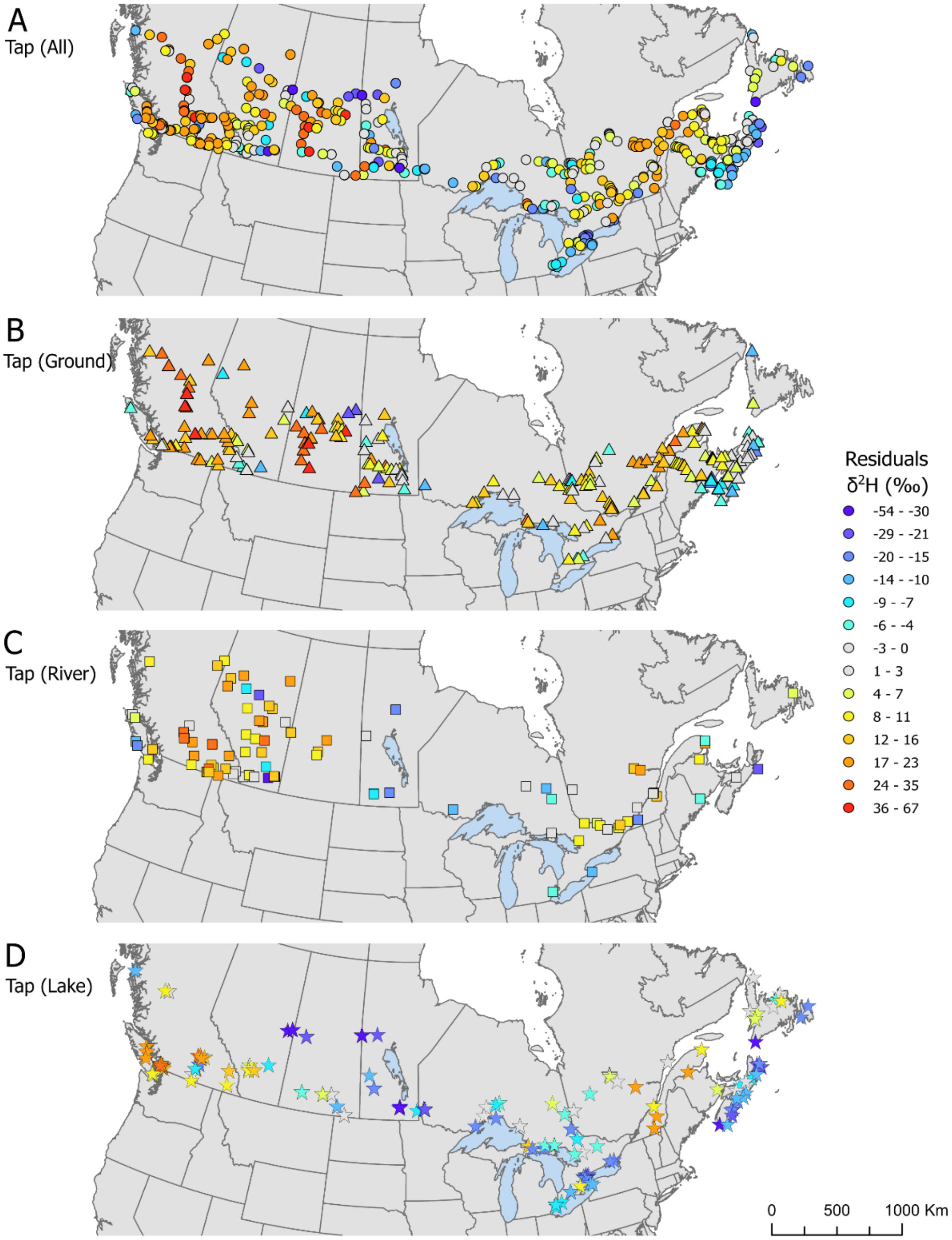

Figure S3. Residuals of δ^2H values between modelled local annual precipitation (based on the Monthly Weighted Annual Precipitation) and tap water (n = 576) across Canada. a: all the tap water samples combined, b: tap water sourced primarily from groundwater (n=281), c: tap water sourced from rivers (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 ArcGIS Pro.

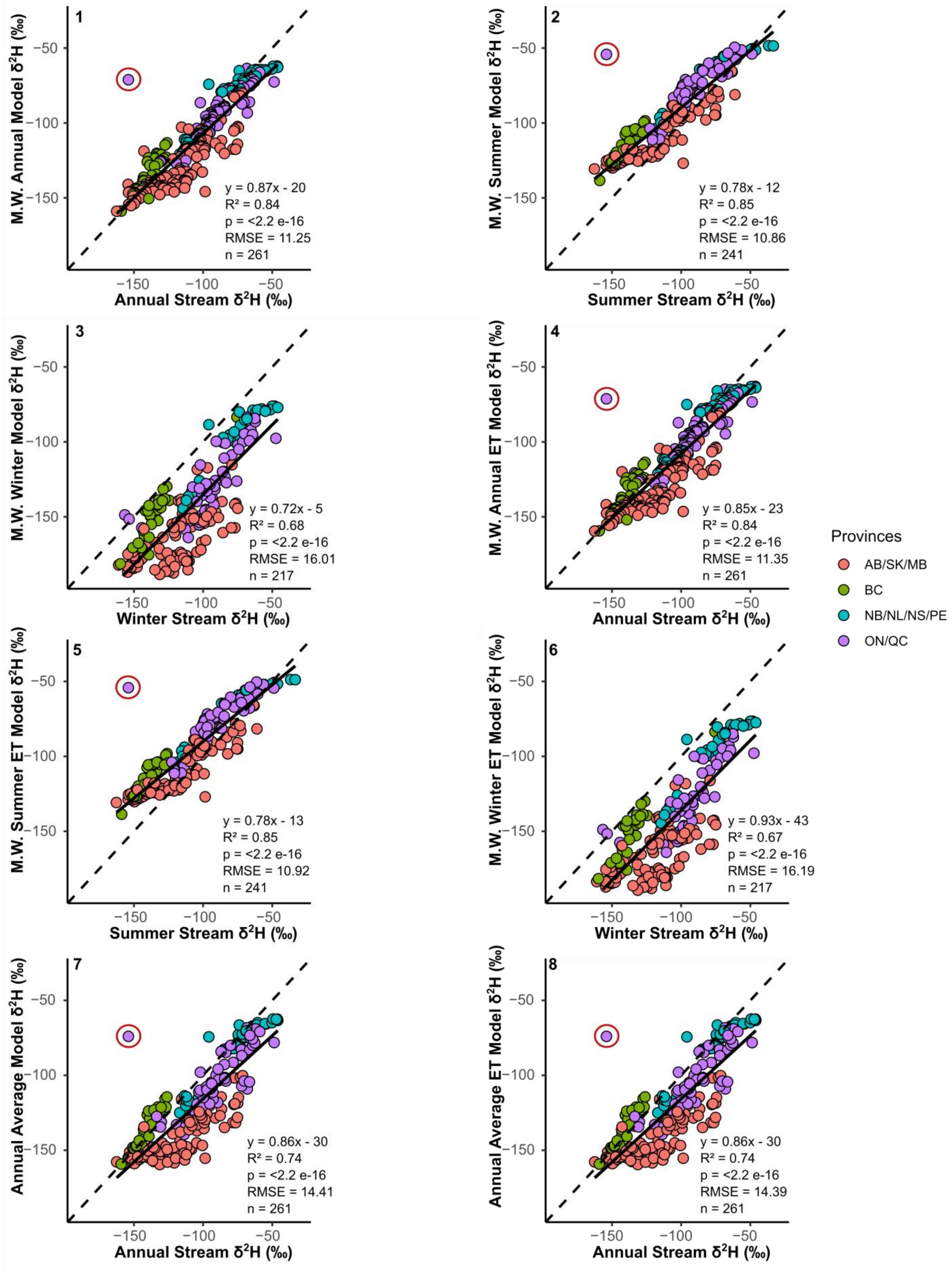

Figure S4. Correlation between δ^2H values derived from the eight water balance models and observed streams δ^2H values annually and seasonally .The dash line represents the 1:1 line. The black line represents the best fit linear model. The outlier data point (red circle) is not reflected in the regression line equation**.**

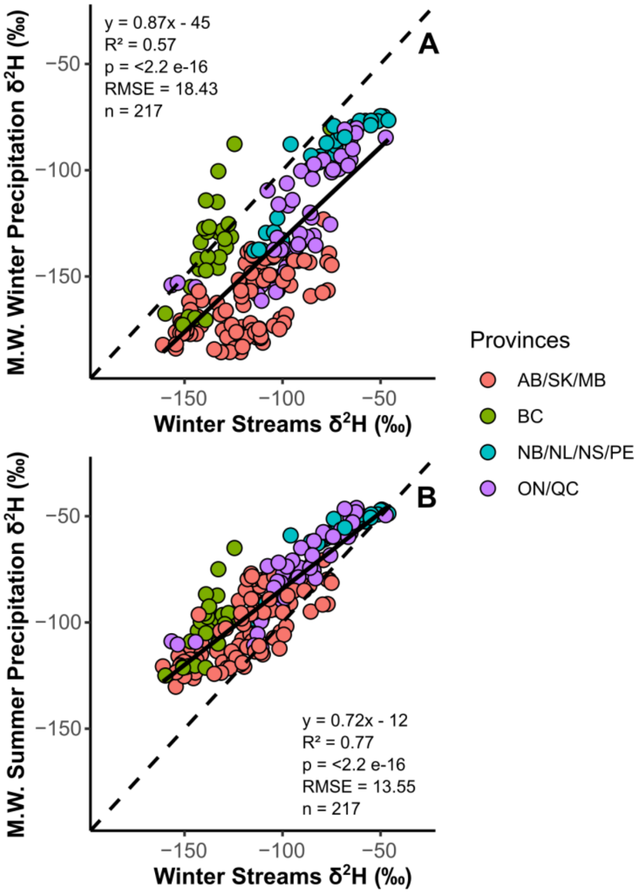

Figure S5. a: Correlation between winter streams δ^2H values and monthly weighted local winter precipitation δ^2H values, b: Correlation between winter streams δ^2H values and monthly weighted local summer precipitation δ^2H values.

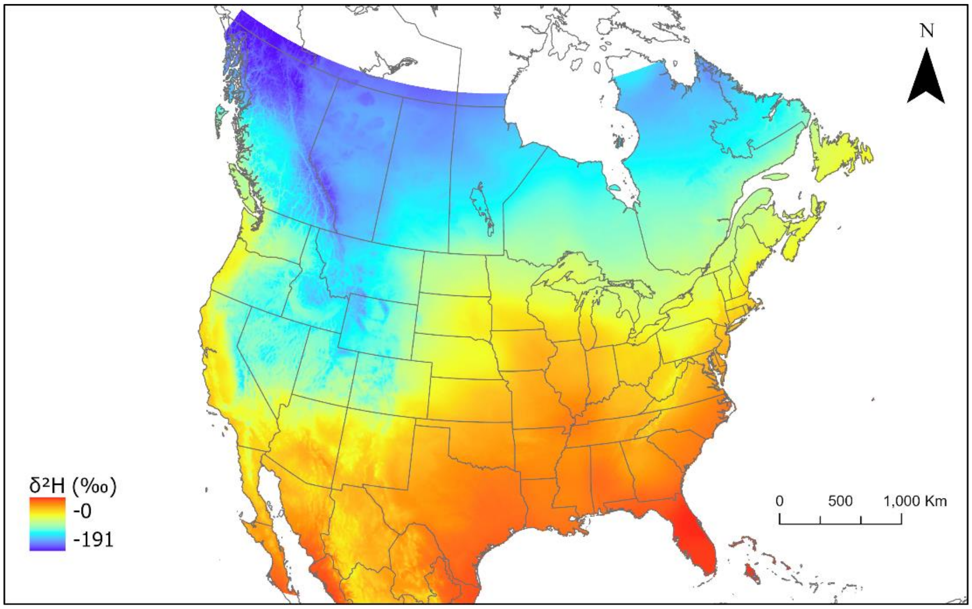

Figure S6. Modelled δ^2H values in precipitation across North America based on the long-term monthly mean isotopic values for global precipitation derived from Waterisotopes.org (Bowen, 2019).

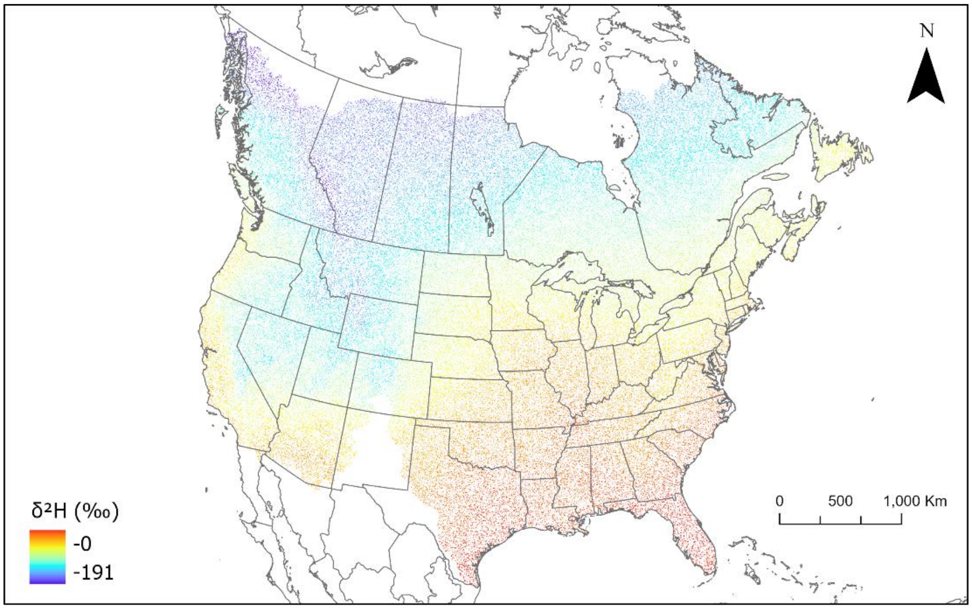

Figure S7. Modelled δ^2H values in surface water (major streams) across North America based on the Monthly Weighted Annual ET Model.

Table S1. Results of linear correlation model between observed streams isotopes and local precipitation isotopes

Observed Streams	δ^2**H**	**Annual precipitation (monthly weighted)**	**Summer precipitation (monthly weighted)**	**Winter precipitation (monthly weighted)**
		$\mathbf{R^2}$	$\mathbf{R^2}$	$\mathbf{R^2}$
Annual streams (262 sites)	δ^2**H**	0.75	0.79	0.56
Summer streams (241 sites)	δ^2**H**	0.72	0.77	0.52
Winter streams (217 sites)	δ^2**H**	0.73	0.77	0.57

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

Data Set S1. Dataset of hydrogen and oxygen isotopes in tap water across Canada collected during the summer at 576 locations. The dataset and readme file is available at https://doi.org/10.6084/m9.figshare.19243518.