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ORIGINAL ARTICLE

Geochemical and isotopic mass balances of kettle lakes in southern Quebec (Canada) as tools to document variations in groundwater quantity and quality

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Abstract Given increasing anthropogenic and climatic pressures on water resources, groundwater and surface water need to be better managed and preserved. As these two water stocks can be connected to each other, their evolutions are linked and need to be considered as such. However, interactions between lakes and groundwater are not well understood and, most of the time, are not taken into account. Therefore, establishing a comprehensive approach to quantify groundwater and lakes' hydrogeochemical interactions in various settings is of foremost importance for assessing the sensitivity of lakes to groundwater evolution. In this study, small kettle lakes set in fluvioglacial deposits and that are most likely well connected to shallow unconfined aquifers are specifically targeted. Geochemistry and isotopic results highlight that groundwater flux to the lakes is generally the dominant parameter of the lake water budget. The ^{222}Rn results in particular suggest that 38% of the studied lakes have a high proportion of groundwater in their balances. It appears that the different tracers are complementary: geochemistry is influenced by groundwater inflows, reflecting its quality and the local geology, whereas water stable isotopes correspond directly to the volumetric lake water budget, and both of these tracers are impacted by in-lake processes. Moreover, the third tracer considered, ^{222}Rn, highlights the location of groundwater inputs in space and time. Finally, the studied kettle lakes are characterized by a short to medium flushing time by groundwater. As a result, these lakes can be highly sensitive to environmental and climate changes affecting groundwater.

Keywords Lakes · Hydrological balance · Groundwater-surface water interactions · Geochemical tracers

Introduction

Groundwater plays a fundamental role in human and ecosystem health, as well as in social and economic development. However, despite their widely recognized importance, groundwater resources are increasingly put at risk. On the one hand, human activities involving groundwater withdrawal, land-use change, and the release of contaminants to the hydrosphere are known to affect groundwater quality (e.g., Arguelles et al. 2014). On the other hand, recent studies suggest that climate instabilities are likely to impart changes to groundwater geochemistry in different settings (Rivard et al. 2009). Groundwater is often an important inflow to surface water, and changes in groundwater quality or quantity will most likely impact surface water evolution. However, flow between groundwater and surface water is nearly always difficult to quantify. In some settings, groundwater contributions are small relative to other water budget terms and can justifiably be ignored. However, in other cases, exchange with groundwater can represent a significant component of lake water or nutrient budgets (Rosenberry et al. 2015), yet remain poorly documented.

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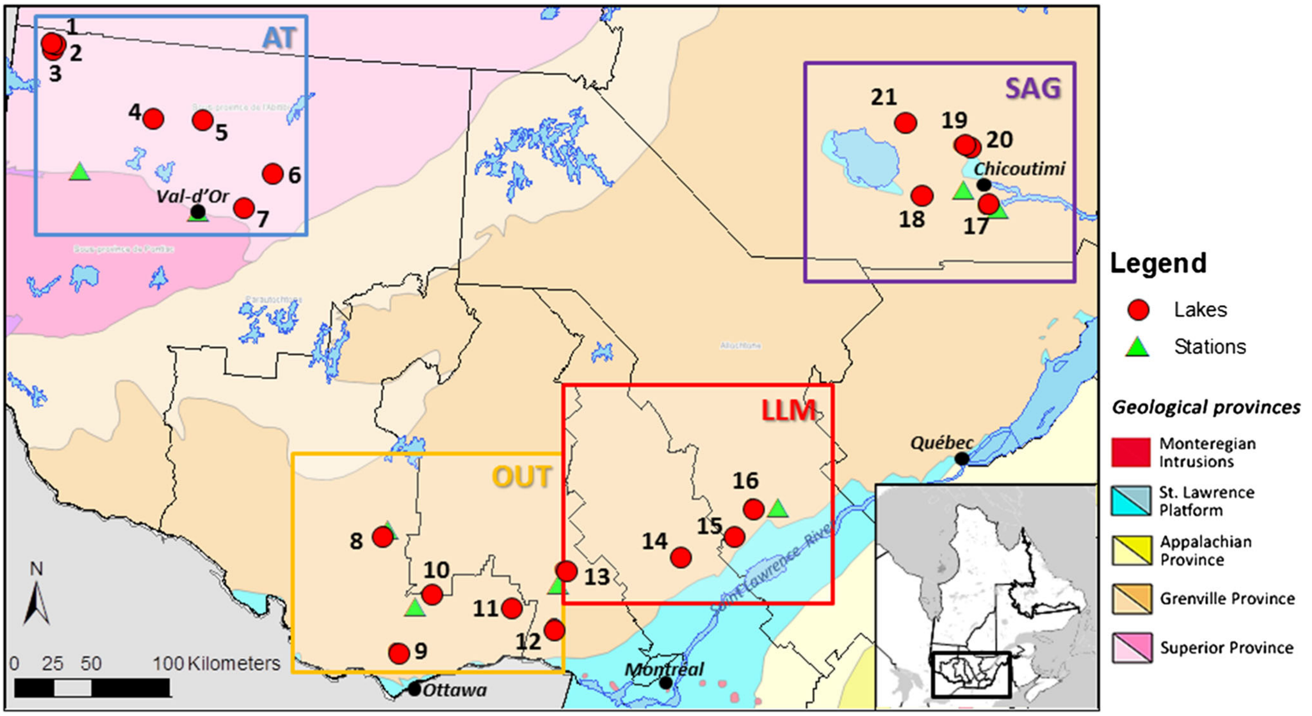


Fig. 1 Locations of the study sites within the four regions: Abitibi-Témiscamingue (AT), Outaouais (OUT), Laurentide-Lanaudière-Maurice (LLM), and Saguenay (SAG), and geological provinces (SIGEOM). Lakes designation to regions is determined on the basis of an administrative division

Furthermore, understanding the hydrogeochemical mass balance of lakes and their interactions with aquifers is critical (1) for water resource management (Showstack 2004), (2) for understanding lacustrine ecology and nutrient balances (Ala-aho et al. 2013; Kidmose et al. 2013), and (3) for quantifying the vulnerability of lakes to pollution (Kløve et al. 2011). Groundwater–lake interactions are nevertheless complex, and hydrogeochemical fluxes are highly variable, both spatially and temporally (Winter 1999), depending on shoreline and bed substrate, aquifer/aquitard characteristics, topography, and meteorological conditions (Rosenberry and LaBaugh 2008), among other factors. In addition, little is known about how climate-driven impacts on groundwater will affect lakes. In this regard, given a proper quantification of groundwater–lake water interactions, the monitoring of surface water quality in time and space could provide valuable insights into how groundwater evolution might affect lakes. The list of methods available to quantify flow between groundwater and surface water is still relatively short (Rosenberry et al. 2015), and mass balance remains one of the most commonly used methods. Mass balance can be based on (1) chemistry (LaBaugh et al. 1995, 1997; Bocanegra et al. 2012; Goldhaber et al. 2014), (2) the stable isotope signature of water (Krabbenhoft et al. 1990; Stets et al. 2010; Turner et al. 2014; Isokangas et al. 2015), and (3) ^{222}Rn activity in water (Cook et al. 2008; Kluge et al. 2007, 2012; Malgrange and Gleeson 2014). However, all of these indices of groundwater–lake interactions have their own uncertainties and, until now, they have very rarely been combined in a single study. In this paper, these three indicators will be applied and compared to determine groundwater fluxes to lakes, and the proportion of groundwater in lake water budgets. Indeed, establishing a comprehensive approach to quantify groundwater and lakes' hydrogeochemical interactions in various settings is of foremost importance for assessing the sensitivity of lakes to groundwater evolution. As such, this study aims to evaluate the potential of using lake geochemistry as a proxy for groundwater evolution in time and space. Small kettle lakes set in fluvioglacial deposits, and that are most likely well connected to shallow unconfined aquifers, are specifically targeted. The specific objectives are (1) to evaluate how these lakes and nearby groundwater can document hydrogeochemical processes at various scales, (2) to develop integrated geochemical methods to quantify groundwater fluxes to/from lakes, and (3) to determine the extent of groundwater controls on the lakes' hydrogeochemical balances.

Study sites

Four study areas were identified in southern Quebec (Fig. 1); the (1) Abitibi-Témiscamingue (AT), (2) Outaouais (OUT), (3) Laurentide-Lanaudière-Mauricie (LLM), and (4) Saguenay (SAG) regions. These are characterized

Table 1 Main characteristics of the study areas; weather data for each region are obtained from mean lakes weather data provided by the *Ministry of Sustainable Development, Environment, and Action against Climate Change* (MDDELCC). Evaporation was calculated using the Thornthwaite method

Areas	Mean annual temperature (°C)	Mean annual precipitation (mm/y)	Mean annual evaporation (mm/y)	Geological province/dominant bedrock geology (SIGEOM)	Main fluvioglacial deposits associated with kettle lakes	Sampled lakes' volume (10^4 m^3) mean/standard deviation (number of lakes–lake with inlet)
AT	0.7–1.3	904–935	513–528	Superior Province Granitoids and volcanic rocks	High continuous eskers and part of the Harricana moraine (Cloutier et al. 2013)	39/52 (7–0)
OUT	3.7–4.8	974–1117	543–574	Greenville Province Granitoids and sedimentary rocks (marble, calcosilicate rocks, dolomite, and quartzite)	Located in valleys and rock depressions (Comeau et al. 2013)	48/44 (5–0)
LLM	2.6–3.5	1054–1075	543–580	Greenville Province Granitoids, mafic and ultramafic rocks, and sedimentary rocks (gneiss, parageneiss, marble, quartzite, and iron formations)	Located in valleys and rock depressions and part of the Saint-Narcisse moraine (Leblanc et al. 2013)	11/19 (4–2)
SAG	0.8–2.8	969–1113	536–538	Greenville Province Granitoids, mafic and ultramafic rocks, and sedimentary rocks (limestone, shale, dolomite, and sandstone)	Small discontinuous eskers and kame terraces (CERM-PACES 2013)	30/25 (5–1)

by contrasting climatic conditions and geological settings, as reported in Table 1. Kettle lakes, set in coarse-grained (sand/gravel) fluvioglacial deposits, from each of the four regions are specifically targeted in this study, because (1) they are small enough to be sensitive to environmental changes on a short timescale, (2) they generally present predictable and uniform geomorphological features, and (3) they are likely connected to shallow, unconfined aquifers. The kettle holes were formed during the transition period from Late Glacial to Holocene, between approximately 12,000 and 7000 years ago (Benn and Evans 2011), when ice blocks were buried in the ground and, as they melted, left depressions in the landscape. When these are filled by water, they are called kettle lakes, and are known, in many cases, to be connected with the underlying aquifer (Isokangas et al. 2015). In the AT region, fluvioglacial deposits form important eskers and moraines (Cloutier et al. 2013), in SAG, they are mainly small eskers and kame terraces (CERM-PACES 2013), and in OUT and LLM, they are mostly located in valleys and bedrock depressions (Comeau et al. 2013; Leblanc et al. 2013). As indicated in Fig. 1, a total of 21 lakes were selected for sampling in summer and autumn. These lakes have the following characteristics: (1) surface areas ranging from 0.001 to 0.3 km^2, (2) maximum depths ranging from 2 to 35 m, implying a volume ranging between approximately 1.5×10^3 and 1.4×10^6 m^3, and, to the extent possible, (3) no permanent surface inflow. The characteristics of the sampled lakes in each study region are described further in.

Materials and methods

Sampling procedures

Water samples were retrieved during two field campaigns conducted in June–July and October–November 2014. Major ions, water stable isotopes (δ^2H and $\delta^{18}O$), and ^{222}Rn activity were measured. The sampling programme included lake water, groundwater, and water from streams connected to lakes, where applicable. Lake water samples were collected from the sector corresponding to the deepest portion of the kettle. Physicochemical parameters, including pH and electrical conductivity, were acquired from water pumped from the water column through an isolated airtight cell, while temperature and dissolved oxygen were measured directly within the water column using a Hanna multiparameter probe. Electrical conductivity was measured using a WTW conductivity meter, according to the Hanna instrument standard HI 70031, and pH using a WTW pH meter, according to the Hanna instrument standards HI 70004, 70007, and 70010. When physicochemical parameters revealed a well-mixed lake, the water column was considered to be homogeneous and only one sample was collected from near the bottom of the lake. Otherwise, for stratified lakes, two samples were collected (except for ^{222}Rn): one from the top of the epilimnion, and one from near the bottom of the lake, in the hypolimnion. This was done in order to account, in as much as possible, for the vertical heterogeneity of the water column by measuring the two extremes in variations for the various parameters. Whenever possible, groundwater was sampled from private wells located in the vicinity of the studied kettle lake. At private residences, groundwater samples were collected from a tap providing untreated water, after purging the well of three times its volume. In cases where there were streams connected to the studied lake, samples were collected from the middle of the channel, and flow rate was measured using a velocimeter, once per campaign. Samples for major ions were filtered in the field using sterile syringe filters (0.45 μm), stored in 30-ml HDPE bottles, and acidified to $pH < 2$ using nitric acid for cation analyses. Unfiltered samples were stored in 35- and 250-ml bottles for stable isotope and ^{222}Rn analyses, respectively. Samples for anion, cation, and water stable isotope analyses were transported in a cooler, and subsequently stored at 5 °C until analyses were performed.

Main water chemistry

Major ion composition (Na^+, K^+, Ca^{2+}, Mg^{2+}, F^-, Cl^-, $NO_3{}^-$, $NO_2{}^-$, $SO_4{}^{2-}$), pH, electrical conductivity (EC), and alkalinity were analysed for 119 samples from the two field campaigns. Alkalinity was titrated using a Hach test kit and following the Gran method. The concentrations of major anions were analysed using a Dionex ICS-1000 ion chromatography with AS14 column. The series of samples were analysed as follows: one blank, five standards, two blanks, samples with a blank following every three samples, and, at the end of the run, one standard and two blanks to validate the calibration and the deviation of the instrument. The samples were run in order of increasing conductivity, and some were analysed twice. Major cations were measured using an AAS 240 FS atomic absorption spectrometer at the University of Paris-Sud (France). The series of samples were analysed as follows: one blank, six standards and one blank to calibrate, then two standards every six samples, and, at the end, the calibration run was analysed again to validate the calibration and the deviation of the instrument. Analysis of some samples was also duplicated.

The ^{222}Rn mass balance

For each studied lake, ^{222}Rn was sampled from groundwater wells, inflow and outflows streams, and in the middle of the lakes near the bottom. ^{222}Rn activity was measured in the laboratory at the University of Quebec in Montreal (Canada) within three days after sampling, using liquid scintillation counting with indirect extraction, following the method described by Lefebvre et al. (2013). For the ^{222}Rn water balance, the water column was considered to be well mixed, and thus, the ^{222}Rn signal is assumed to be relatively uniformly distributed, both vertically and horizontally. However, if the lake is stratified, it has been shown that the ^{222}Rn activity in the hypolimnion is similar to or lower than the lake average (Kluge et al. 2007, 2012). Indeed, Kluge et al. (2012) showed that, in some lakes sampled during the summer, the ^{222}Rn concentration is stratified with lower concentrations near the bottom. In this study, the groundwater flow, measured in using only one sample from near the bottom of the lake, may therefore be underestimated for stratified lakes. Following the water mass budget approach, a single-box steady-state advection model of ^{222}Rn can be used to quantify groundwater discharge rates (Gleeson et al. 2009). In this application, it is assumed that groundwater discharge into the lakes is at steady-state and that this input represents the only substantial source of ^{222}Rn. In general, areas of anomalous ^{222}Rn activity are rare and are usually restricted to portions of the lake where pilings and/or artificial canals have been constructed, providing groundwater pathways to surface waters (Dimova et al. 2013). Furthermore, it is assumed that the diffusive ^{222}Rn flux from the lake sediments is negligible. However, for lakes with low groundwater discharge, the ^{222}Rn diffusive flux can be relatively important in the overall budget; in these cases, the discharge rate calculated may be overestimated. ^{222}Rn activity is negligible in the atmosphere and in precipitation. Since the radium activity in water is low, and since radium is bound to sediment particles in fresh water (Kiro et al. 2012), the ^{222}Rn production from radium decay is assumed to be negligible. The ^{222}Rn budget is then written as:

$$C_L(Q + k \times A + \lambda \times L) = I_S C_S + I_G C_G \quad \left(\mathrm{Bq \cdot l^{-1} \cdot m^3 \cdot y^{-1}}\right) \tag{1}$$

where λ is the radioactive decay constant (0.18 day^{-1}), L is the volume of the lake (m^3), C_L is the ^{222}Rn activity in the lake, C_S is the ^{222}Rn activity in the incoming surface water, C_G is the ^{222}Rn activity in the groundwater, Q is the total outflow from the lake, and k is the gas exchange velocity which is assumed to be constant as it is weakly variable for small lakes (Vachon et al. 2013). The gas exchange velocity has been estimated at 0.16 m/d by Cook et al. (2008) and used in mass balances in Quebec for small lakes by Malgrange and Gleeson (2014) and therefore is also used in this study. The total outflow from the lake is assumed to be constant and equal to the mean surface outflow measured during the two field campaigns. Lake volumes are estimated from bathymetric maps created in ArcGIS using kriging based on shorelines positions and depth profiles. Depths were measured on site with a Speedtech Depthmate portable sounder along one or two profiles per lake, with a total of only three to as many as thirty points, depending on the lake size. Only three depths points were obtained for only one lake (#16) and included the deepest point of the lake.

The stable isotopic mass balance

The stable isotope compositions were determined for a total of 119 samples from the two field campaigns. Samples were analysed with a LGR Laser Water Isotope Analyser OA ICOS DLT at the University of Paris-Sud (France). The analytical accuracy is ±1‰ for δ^2H and ±0.2‰ for δ^{18}O. The results are presented in δ values, representing deviation in per mill (‰) from the isotopic composition of Vienna Standard Mean Ocean Water (VSMOW), such that $\delta^2\mathrm{H}$ or $\delta^{18}\mathrm{O} = ((R_{\mathrm{sample}}/R_{\mathrm{standard}}) - 1) \times 1000$, where R refers to the ^{2}H/H or ^{18}O/^{16}O ratios in both the sample and standard. The annual water mass and isotope mass balance for a well-mixed lake may be written as:

$$\frac{\mathrm{d}L}{\mathrm{d}t} = I - E - Q \quad (\mathrm{m}^3 \cdot \mathrm{y}^{-1}) \tag{2}$$

and

$$L\frac{\mathrm{d}\delta_{\mathrm{L}}}{\mathrm{d}t} + \delta_{\mathrm{L}}\frac{\mathrm{d}L}{\mathrm{d}t} = I\delta_{\mathrm{I}} - E\delta_{\mathrm{E}} - Q\delta_{Q} \quad (‰ \cdot \mathrm{m}^3 \cdot \mathrm{y}^{-1}) \tag{3}$$

respectively, where L is the volume of the lake, t is time, %E is evaporation, I is instantaneous inflow, which is the sum of surface inflow from upstream, runoff from the catchment area, groundwater inflow, and precipitation on the lake surface, Q is instantaneous outflow, which is the sum of surface and groundwater outflow, δ_{L} is the isotopic composition of the lake, δ_{I} is the isotopic composition of the total inflow, and δ_{Q} is the isotopic composition of the total outflow (Gibson et al. 2015). Here, a steady state is assumed. Thus, combining Eqs. (2) and (3), groundwater inflow can be determined without information on outflow by determining the following parameters:

- *Lake (L, δ_L)*

Lake volume (L) is estimated as described in "The ^{222}Rn mass balance" section. The lakes' isotopic compositions (δ_{L}) have been measured in samples from the two field campaigns described above. However, when lakes appeared to be stratified (in summer for most of the sampled lakes), the representativeness of δ_{L} is modified due to different processes occurring in only one of the two mixed layers, such as evaporation that is restricted to the epilimnion. For the isotopic mass balance, a mean value of δ_{L} is used for the entire water column, calculated from the two values measured.

- *Evaporation (E, δ_E)*

Evaporation is calculated using the Thornthwaite method, as described in Rosenberry et al. (2007), based on monthly mean air temperatures at each lake. Monthly evaporation is calculated as time-weighted daily values summed over the month, then multiplied by the area of the lake. The isotopic composition of the evaporating moisture (δ_{E}) is not directly measured, but can be estimated using the Craig and Gordon (1965) model, as described by Gonfiantini (1986):

$$\delta_{\mathrm{E}} = \frac{(\delta_{\mathrm{L}} - \varepsilon^{+})/\alpha^{+} - h\delta_{\mathrm{A}} - \varepsilon_{\mathrm{K}}}{1 - h + 10^{-3}\varepsilon_{\mathrm{K}}} (‰) \tag{4}$$

where h is the relative humidity at the lake surface temperature (decimal fraction), δ_{A} is the isotopic composition of the local atmospheric moisture, $\varepsilon^{+} = (\alpha^{+} - 1) \times 1000$ is the equilibrium isotopic separation, α^{+} is the equilibrium isotopic fractionation (Horita et al. 2008), and $\varepsilon_{\mathrm{K}} = -C_{\mathrm{K}}(1 - h)$ is the kinetic isotopic separation, where C_{K} is the ratio of molecular diffusivities of the heavy and light molecules (see also Gibson et al. 2015). Here, C_{K} values used are representative of fully turbulent wind conditions and a rough surface, which is the case for our study lakes [i.e., 14.2‰ for oxygen and 12.5‰ for hydrogen, respectively, based on experimental data (Horita et al. 2008)]. Horita and Wesolowski (1994) give experimental values of:

$$\alpha^{+}(^{18}\mathrm{O}) = \exp(-7.685/10^3 + 6.7123/T - 1666.4/T^2 + 350410/T^3) \tag{5}$$

$$\alpha^{+}(^{2}\mathrm{H}) = \exp(1158.8 \times T^3/10^{12} - 1620.1 \times T^2/10^9 + 794.84 \times T/10^6 - 161.04/10^3 + 2999200/T^3) \tag{6}$$

for δ^{18}O and δ^2H, respectively, where T is temperature in Kelvin. The isotopic composition of atmospheric moisture (δ_{A} in ‰ vs SMOW) was calculated assuming equilibrium isotopic exchange between the liquid and vapour phases, as follows:

$$\delta_{\mathrm{A}} = \frac{\delta_{\mathrm{P}} - \varepsilon^{+}}{1 + 10^{-3}\varepsilon^{+}} (‰) \tag{7}$$

where δ_{P} (in ‰) is the isotopic composition of precipitation weighted here by the evaporation flux (Gibson et al. 2015). Mean temperature and humidity are measured at land-based meteorological stations located as near as possible to the lakes (2000–2014 period; Fig. 1).

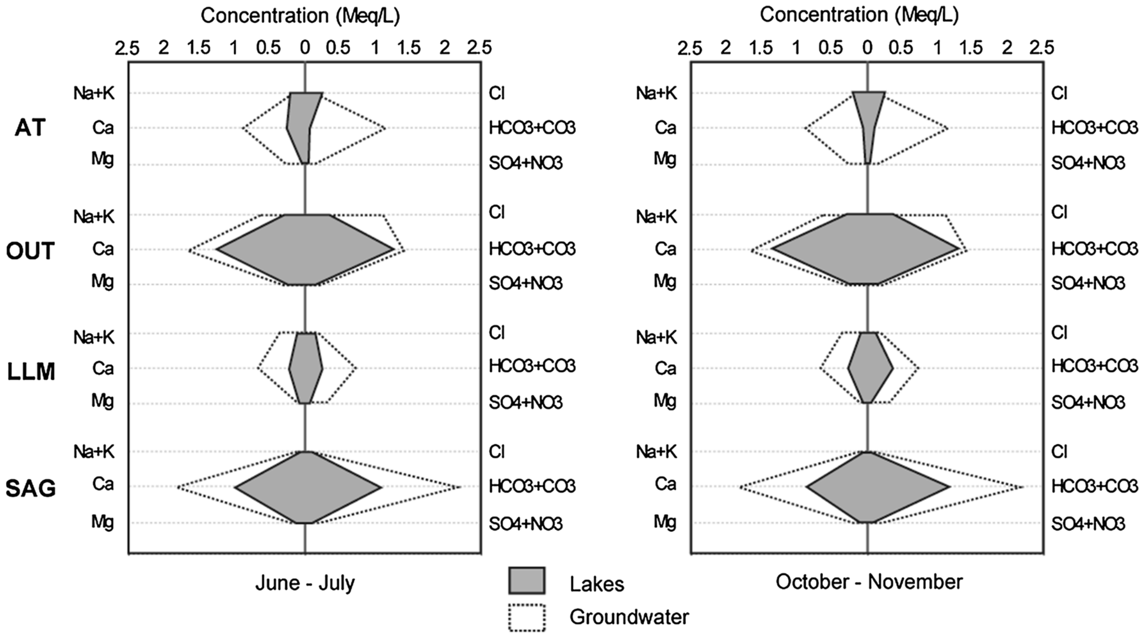


Fig. 2 Mean major ion contents for each study region in lakes (*grey*) and in groundwater (*dashed lines*) for the two sampling campaigns

- *Precipitation (P, δ_P)*

Hydrological inputs from precipitation (P) were estimated from a temporal series of land-based meteorological stations by interpolation. These data have been provided by the CEHQ *(Centre d'Expertise Hydrique du Québec)* from 1900 to 2010 for a 0.1° grid size (Poirier et al. 2014). The mean annual precipitation value is based on monthly precipitation values at each sampled lake from 1900 to 2010, as summarized in Table 1. The monthly mean isotopic composition of precipitation (δ_P) was assessed in the 4 regions from GNIP (*Global Network of Isotopes in Precipitation*) and PACES (*Programme d'Acquisition de Connaissances sur les Eaux Souterraines*) datasets. Mean annual precipitation isotopic composition used in the lake balances is flux-weighted by monthly values.

- *Groundwater input ($\boldsymbol{I_G}$, δ_{Gi}), stream input ($\boldsymbol{I_S}$, δ_s), and inflow from catchment runoff ($\boldsymbol{I_R}$, δ_R)*

The evaporation to inflow ratios (*E/I*; dimensionless) are used to discuss relative parameter dominance (Isokangas et al. 2015, Gibson et al. 2015, Turner et al. 2010):

$$E/I = \frac{\delta_I - \delta_L}{\delta_E - \delta_L} = x(\text{dimensionless}) \tag{8}$$

where $I = P + I_S + I_R + I_G$. In many studies, runoff has either been assumed to be a percentage of rainfall minus evapotranspiration, depending on soil composition (Stets et al. 2010), or it is simply not considered at all (Krabbenhoft et al. 1990; Sacks et al. 2014). Thus, for a headwater lake, if the annual runoff is assumed to be negligible, annual groundwater inflow can be defined as:

$$I_G = \frac{E}{x} - P\left(\mathrm{m}^3 \cdot \mathrm{y}^{-1}\right) \tag{9}$$

A sensitivity analysis was conducted in order to evaluate the robustness of the calculations proposed above. Model sensitivity to parameter uncertainty is first assessed by observing the variations in groundwater influence as a function of each parameter for a lake. During the sensitivity analysis, the values corresponding to each parameter are modified in a stepwise manner and the differences in the calculated groundwater discharge rates are noted. The cumulative uncertainty associated with all of the model parameters (i.e., lake area, lake depth, isotopic signatures, precipitation, and evaporation rates) is subsequently calculated.

Results and discussion

Hydrochemistry

Mean major ion compositions of water samples from the two field campaigns are presented in Fig. 2. The ionic balance errors of analyses are less than 10% in 57.5% of the 119 samples. The standard deviation of the ionic balance is 12%, the mean is 10%, and the median is 7%. The very low ion content of the sampled water is presumed to be responsible for the relatively high ionic balance errors. In Fig. 2, three groups of data can be observed when comparing lake water with groundwater compositions, depending on the study region. The AT kettle lakes tend to present a completely different pattern

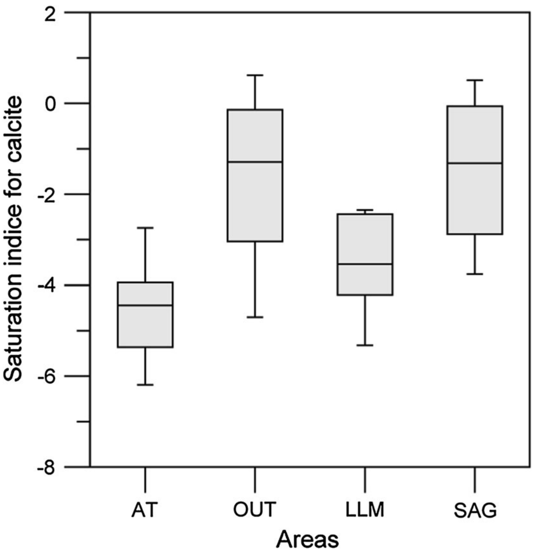


Fig. 3 Median, first and third quartiles, and maximum and minimum values of the mean calcite saturation index from lake samples from the *four* study areas

from groundwater, with a composition closer to that of rain water, with a very low ionic content, suggesting very limited connection with groundwater. For the other three areas, the patterns are similar between lakes and groundwater. LLM and SAG lakes' water are close to groundwater composition, and most likely reflect the groundwater influence on lakes in this area, albeit with higher dilution from rain water than is observed for the OUT region. The OUT lakes' waters are indeed very close to the groundwater composition, suggesting an important interconnection between the sampled kettle lakes and groundwater in this region. The saturation indices with respect to calcite ($SI_{calcite}$), calculated from the ion contents of lake water samples, are significantly different between AT–LLM and OUT–SAG, as illustrated in Fig. 3. In the OUT and SAG regions, this index is closer to equilibrium (with mean values of $SI_{calcite}$ equal to −1.6 and −1.5, respectively) than for the AT and LLM regions, in which $SI_{calcite}$ shows unsaturated lakes with respect to calcite (with mean SI_{cacite} values equal to −4.6 and −3.5, respectively). $SI_{calcite}$ is supposed to be close to or higher than equilibrium for groundwater in areas where carbonates are abundant. In the AT and LLM regions; $SI_{calcite}$ for groundwater is undersaturated (with mean values equal to −1.8 and −2.8, respectively). However, in OUT and SAG regions, where calcite deposits can be found, the $SI_{calcite}$ for groundwater remains undersaturated (with mean values equal to −1.1 and −1.7, respectively). The water from the sampled wells taken to represent groundwater are therefore probably a mix of lake water and groundwater instead, and groundwater $SI_{calcite}$ is therefore not considered further in this analysis. The lake water $SI_{calcite}$ shows that lakes are representative of the carbonate bearing geology in OUT and SAG. It suggests that lakes can be connected to groundwater in these two areas. For the other two areas, AT and LLM, low $SI_{calcite}$ may either indicate the absence of carbonate deposits or the disconnection of lakes from groundwater. Therefore, there are two components underlying the geochemistry of these kettle lakes: their geological setting followed by their connection to groundwater.

^{222}Rn mass balance

Analyses of groundwater collected from wells near all studied lakes show that the ^{222}Rn activity in groundwater is 2–3 orders of magnitude higher than in the lake waters. ^{222}Rn activity is therefore well differentiated between lakes and groundwater (Fig. 4b). The results reveal that ^{222}Rn activity shows significant spatial variability in groundwater, even within a single region (Fig. 4a). Groundwater ^{222}Rn activity used in the mass balance model for each region was the mean activity measured in the wells (8539, 5024, 7585, and 24,033 Bq/m^3 for LLM, SAG, AT, and OUT, respectively). ^{222}Rn activity is much higher in OUT than in the other regions. This likely reflects the geology of the region, which is known to be rich in granitic pegmatites and skarn (SIGEOM). A well-mixed single-box model is used to calculate groundwater flows to each lake (Malgrange and Gleeson 2014). A more precise multi-box model has already been developed for a lake (Kluge et al. 2012). However, this same study shows that the results of single-box models are comparable to the results of the multi-box model calculations and supports the use of one well-mixed single-box model with a mean ^{222}Rn concentration for inflow calculations (Kluge et al. 2012). In many of the sampled lakes, ^{222}Rn activity was below the detection limit. However, the absence of the tracer at detectable levels is not a proof of no groundwater inflow to the lake, since it can also be due to internal mixing in the lake or a long water residence time compared to the ^{222}Rn half-life. It is important to keep in mind that ^{222}Rn, because of its very short half-life, is an indicator of groundwater inflows to surface water but only at the specific time of the sampling. Moreover, considering that the measured value represents the mean value of the lake and that groundwater inflows do not change throughout the year, a mean groundwater flux towards the lakes can be calculated (Fig. 5). Although the value obtained is only an approximation of groundwater fluxes, because ^{222}Rn activity can follow stratification in the lake, it provides a first estimate to compare behaviour between lakes. Computed mean fluxes normalized to the lake surface area range between 0.12 and 141 m/y, as shown in Fig. 5, with most of the

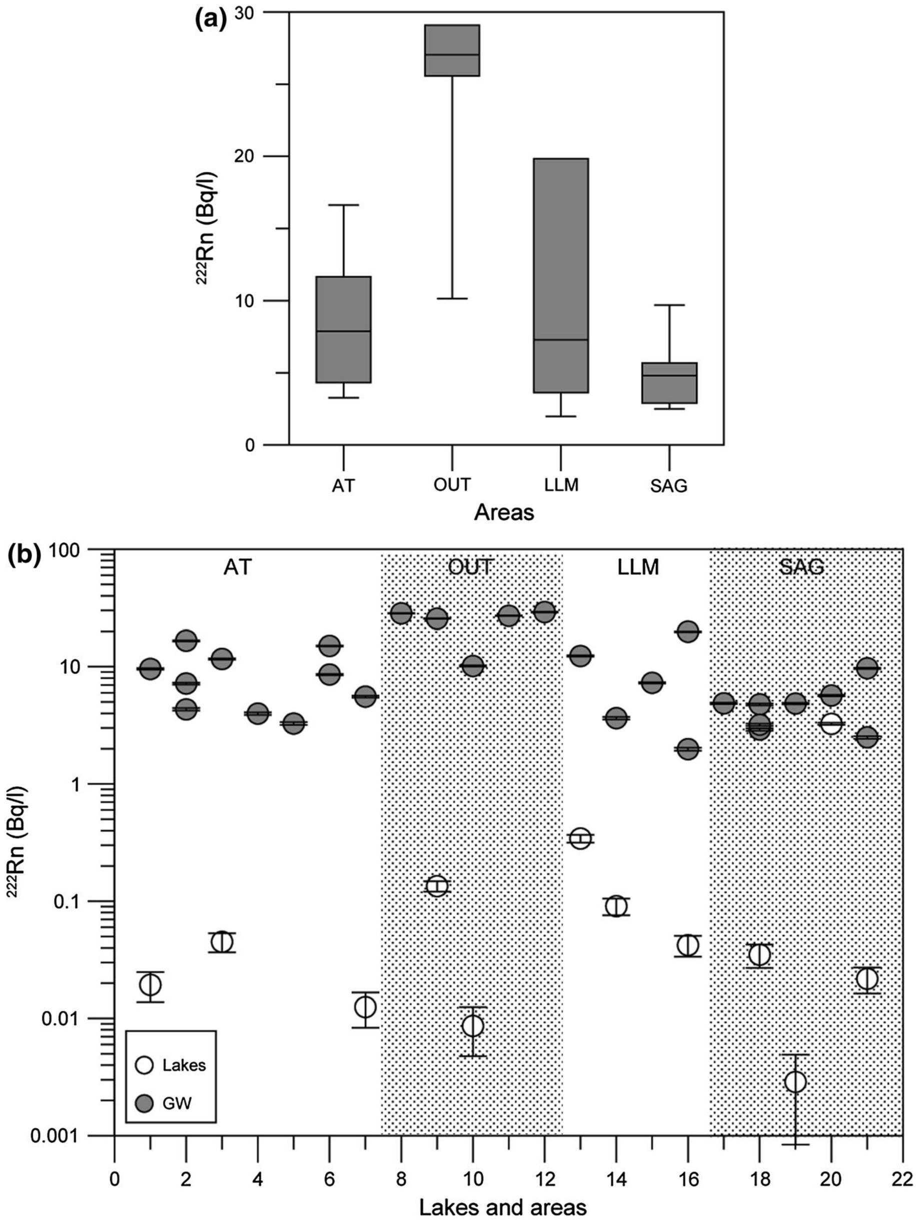


Fig. 4 **a** Median, first and third quartiles, and maximum and minimum of groundwater ^{222}Rn activity for each area and **b** ^{222}Rn activity measured in the deep part of the lakes (*white circles*) and associated groundwater measured in nearby wells (*black circles*)

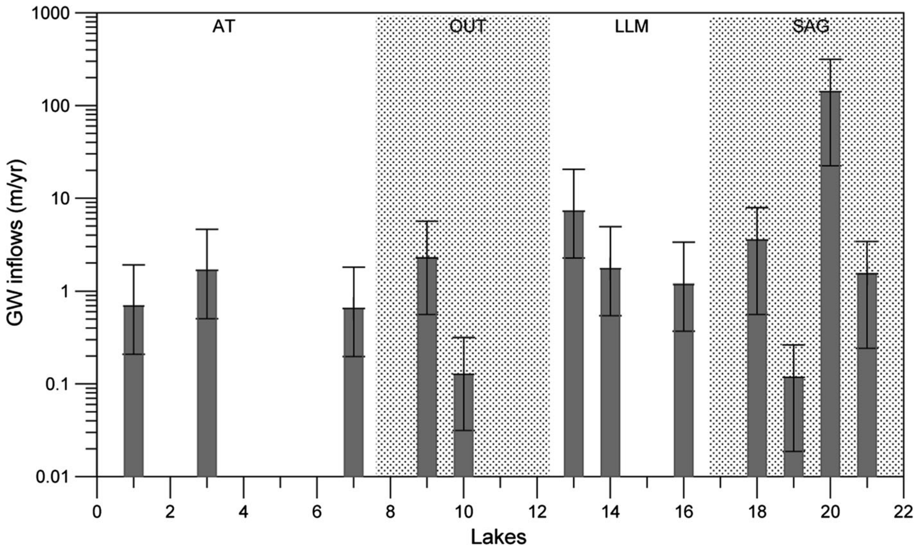
lakes having a groundwater input of close to 1 m/y. The groundwater fluxes calculated from ^{222}Rn do not show any trends between regions.

Stable isotopic mass balance

Stable isotopic composition of water

The isotopic composition of lakes depends on many factors, including the dynamics of inflows (their quantities and their signatures), stratification, evaporation, sampling time, and geomorphology (depth, surface, and volume). The isotopic data presented in Fig. 6 lie on two different local evaporation lines (LEL) depending on their location ($\mathrm{LEL_{North}}$/$\mathrm{LEL_{South}}$). It has previously been shown that the intersection of the LEL with the local meteoritic water line (LMWL) often provides an valid approximation of the weighted-mean isotopic composition of input waters to a catchment (δ_I), while the displacement of a given lake water (δ_L) along the LEL provides an indication of water balance (Edwards et al. 2004). Thus, the current results confirm that total inflows to the studied lakes have an isotopic composition close to the mean isotopic composition of groundwater, which is similar to the amount-weighted mean annual isotopic value for precipitation (δ_{PFW}) of each study area. Generally, the amount-weighted mean annual isotopic composition of precipitation is controlled by altitude and latitude (Delavau et al. 2011). Here, as the studied regions are relatively flat, this is controlled mainly by variations in latitude. It can be observed that a

Fig. 5 Groundwater inflow to lakes obtained through ^{222}Rn mass balance, the fluxes normalized to the lakes' surface areas. The *bars* represents groundwater inflows calculated for each lake (numbered as per Fig. 1) from the mean ^{222}Rn activity in groundwater per region, ±the standard deviation of ^{222}Rn activity in groundwater per region



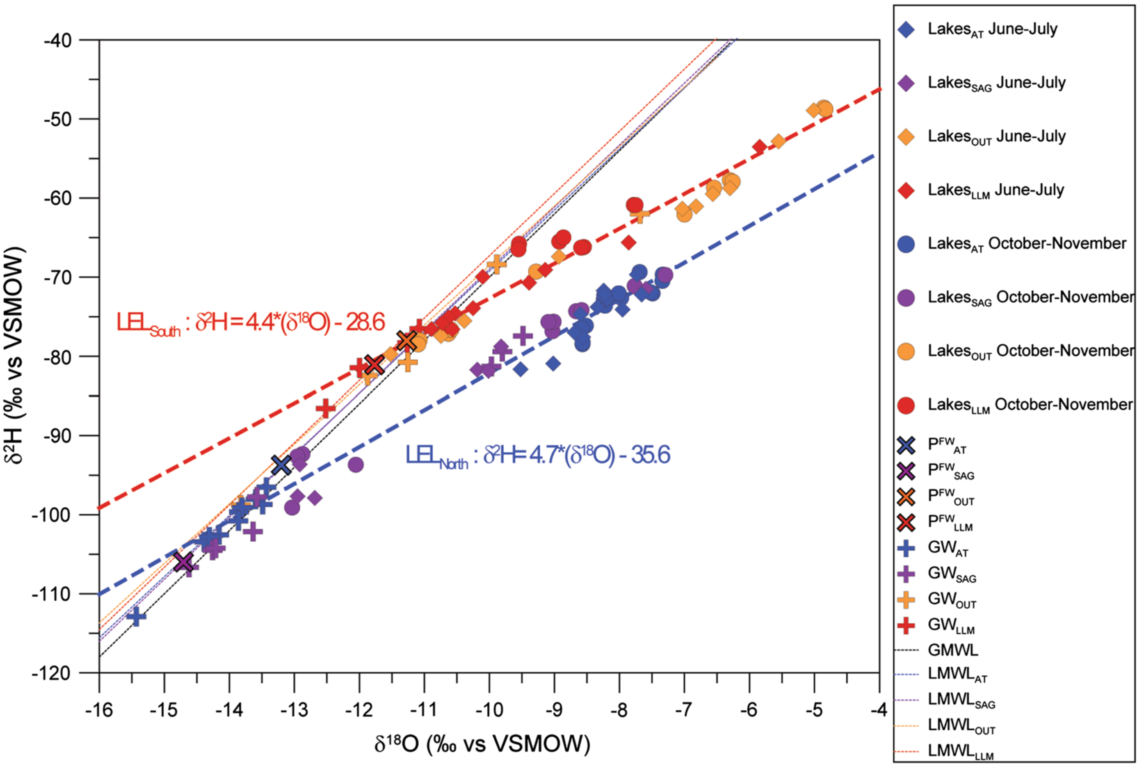


Fig. 6 Isotopic signatures in precipitation, lake water, and groundwater in the *four* areas

few groundwater values do not follow this trend, and are instead close to lakes values, suggesting that some sampled wells might represent a mixture between lake water and groundwater.

The calculated E/I of the studied lakes, based on δ^{18}O isotope mass balance, are shown in Fig. 7, and suggest that kettle lakes are mainly dominated by groundwater inflow and not by evaporation, even after summer. Three groups of data are identified in Fig. 7: (1) *E/I* values of between 0 and 0.2, where lakes are typically through-flow lakes dominated by groundwater inflow, (2) *E/I* values varying from 0.2 to 0.4, which are still dominated by groundwater inflows, but with a higher influence of evaporation (this is the case for all lakes located in AT region), and (3) two lakes (#10, 18) with an *E/I* value of close to 0.5, the value above which the lake becomes evaporation-dominated (Turner et al. 2010). It is worth noting that lake #18 (SAG) is likely affected by anthropogenic impacts, because it is

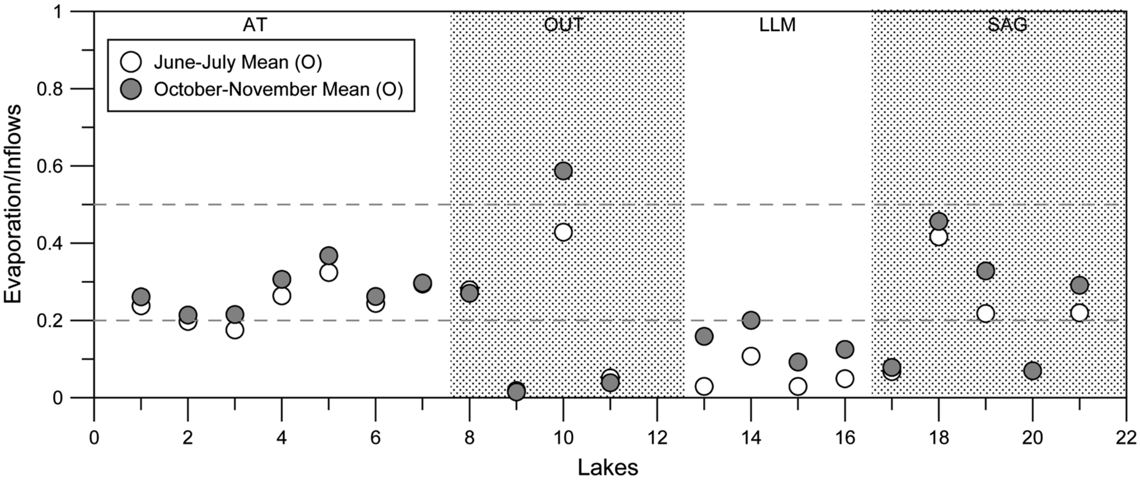


Fig. 7 Evaporation to inflow ratios (*E/I*) calculated from the isotopic water balance model for the two sampling periods

located in the vicinity of a municipal well. These anthropogenic impacts could, at least in part, explain the decrease in groundwater level and therefore in groundwater inflows to this lake. Another reason for their high E/I values could be the proximity of these two lakes to a larger lake, which can lead to groundwater inflow already enriched by evaporation.

Sensitivity analysis

Uncertainties are associated with the proposed methodology for quantifying groundwater fluxes to lakes. These are based on both the hypotheses and on the uncertainties of each parameter included in the balance calculation. Sensitivity tests were performed to derive the range of uncertainties associated with the quantities being evaluated, such as total *E/I*. The sensitivity analysis focused on the main parameters used in the isotopic model described above. All variables present in the model were considered in this process. As illustrated in Fig. 8, each parameter (lake isotope signature, mean isotope signature of precipitation, flux-weighted by evaporation, mean isotope signature of precipitation, flux-weighted by precipitation isotope signature, lakes surface area, precipitation, evaporation, temperature, and relative humidity) was varied by ±10% of the obtained value. It can be observed that (1) the model is more sensitive to evaporation than to any of the other parameters, and (2) the model diverges if the isotopic value of the lake water is too high or too close to the maximum enrichment calculated for the lake (which is the case for lake #12).

Comparison of the three approaches

The dependence of the studied lakes on groundwater can be quantified through the G-index, defined as the contribution of groundwater inflow to the total inflow of water to a given lake (Isokangas et al. 2015). The groundwater inflow obtained from the isotopic mass balance is derived by subtracting precipitation from the total inflow. The values of the G-index obtained for the studied lakes are presented in Fig. 9. G-indices were found to vary from 0 to more than 95% with the isotopic mass balance and from 0 to 90% with the ^{222}Rn mass balance. A chemical index (C-index) has been determined from the stiff diagrams, based on the ratio between stiff areas formed by lake water compositions and mean groundwater composition of the four areas, shown in Fig. 2. Thus, if the C-index is equal to 1, the chemical composition of the lake is the same as the mean groundwater composition. The lake is then less (or more)

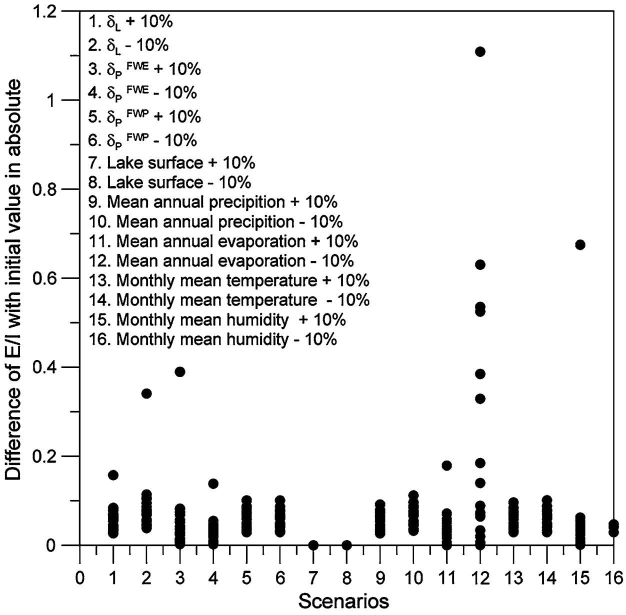


Fig. 8 Sensitivity analyses of the isotopic mass balance for all the lakes, following the different scenarios

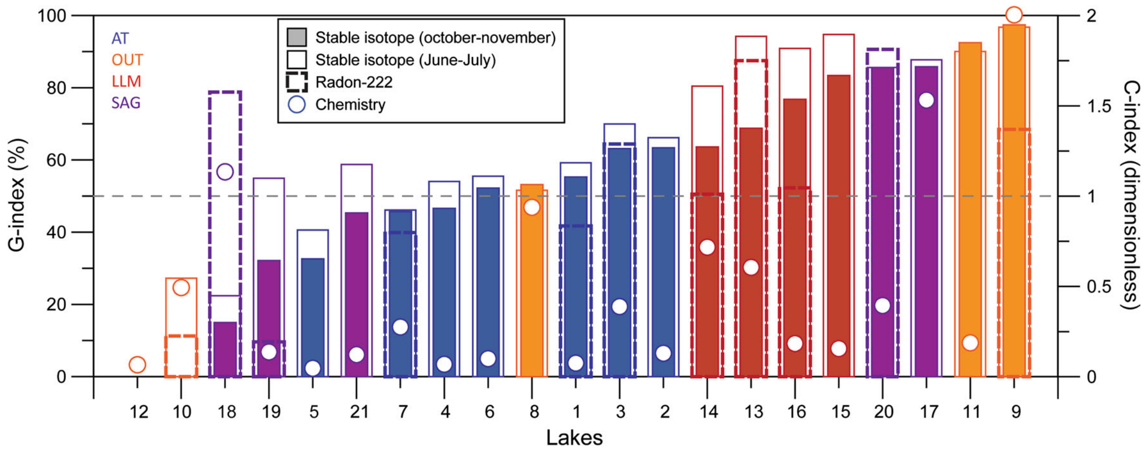


Fig. 9 G-index and the C-index values of each of the lakes, quantifying their groundwater dependency. Results are presented in order of increasing G-indices obtained from autumn stable isotope signatures in lakes

mineralized than the mean groundwater if this C-index is below (or above) 1. In the AT region, the C-index is close to zero, suggesting that the lakes' ionic compositions are much closer to rain than to groundwater. In the three other regions (representing 13 lakes), the C-index is between 0 and 2, with 4 lakes between 0.5 and 1, and 3 lakes between 1 and 2. With the two indexes (G-index and C-index), based on the three approaches, two families can be highlighted: lakes that are not well connected to groundwater—the AT region—and lakes showing stronger connections—the other three regions—(Fig. 9).

It can be observed that the groundwater dependence of lakes calculated using the three different methods are not consistent. This is due to the fact that each tracer represents different processes of mixing between lake water, groundwater, and other inflows in space and time. The isotopic-based mass balance does not allow for a direct quantification of groundwater inflows to lakes, because the isotopic contents of the different inflows and the lake waters are not well differentiated. However, this method allows the determination of the dominant parameters (i.e., groundwater inflows, precipitation, or evaporation) of the lake water budgets, which is essential for the determination of lake evolution and vulnerability. This approach is valuable to determine the E/I and the percentage of groundwater in the lake budget. It is important to keep in mind that the groundwater influence can be calculated adequately only if the surface inflows to the lakes are negligible; otherwise, total inflows represent the sum of groundwater and surface inflows (inlets and runoff), and the latter should be considered explicitly. The isotopic composition of a lake is a picture of its yearly hydrological budget.

The ^{222}Rn-based method provides information limited in space, because it depends on the sampling point in the lake, as well as in time, because the ^{222}Rn half-life is only 3.8 days. This tracer provides valuable information by allowing the groundwater flux to be quantified directly. This method would be more precise for a yearly budget if field measurements could be performed continuously—as achieved by Gilfedder et al. (2015) in a swamp—to precisely calculate the flux means with respect to temporal variations. The other difficult aspect of this approach is that understanding of ^{222}Rn activity in groundwater needs to be refined, because, as already established, it can be highly variable and has an important influence on the estimated fluxes. The use of a well-mixed single-box model is an assumption which may either maximize or minimize the calculated mean annual groundwater flux to lakes, depending on the horizontal and vertical variability of the ^{222}Rn signal.

The chemistry shows similar variations in lakes of the different regions between summer and autumn, as highlighted in Fig. 2, and therefore is an index adapted for yearly observation of groundwater inflows. However, the C-index remains a qualitative index, because chemical elements are not always conservative. Indeed, they are often subject to modification induced by in-lake biological processes and anthropic impacts. In this study, results suggest that each tracer has its own importance and provides complementary information to the estimation of groundwater influence on lake waters. The temporal information provided by the tracers also differs (i.e., from a few days' timescale with ^{222}Rn activity to years with stable isotopes of water and chemistry).

As highlighted by the previous indicators, the establishment of yearly budgets seems to be the most appropriate method to observe the sensitivity of lakes to groundwater changes, both in quantity and in quality. The flushing time is calculated as the volume of water in a defined system divided by the volumetric groundwater flow rate through the system (Monsen et al. 2002). Therefore, in

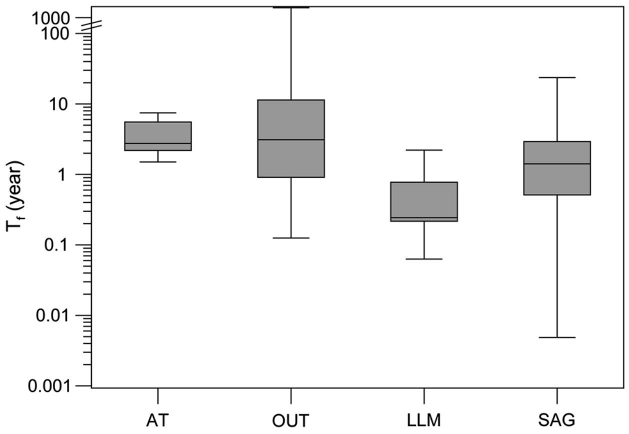


Fig. 10 Flushing time obtained with both ^{222}Rn- and δ^{18}O-based methods for the different regions

assuming that groundwater inflow does not change much during the year, the flushing time by groundwater (T_f) is expressed as:

$$T_f = {}^{L}\!/_{I_G}\,(\text{year}) \tag{10}$$

where L is the lake's volume and I_G is the groundwater inflow to the lake. Figure 10 shows the variations of lake flushing time by groundwater in the four areas obtained from the stable isotope and ^{222}Rn mass balances. It can be observed that most of the studied lakes present an average groundwater flushing time ranging between 0.1 and 10 years. 25% of the lakes have a $T_f < 1$ year, 20% have a $T_f < 2$ years, and one lake has a surprisingly long T_f (>1000 years), because of its depleted isotope signature in November, too close to the estimated maximum enrichment. Therefore, this last result is not considered to be reliable. The two indices impacting the response of the lake to a perturbation (G-index and T_f) can be combined in one graph (Fig. 11), which illustrates the resilience of the lakes if they are subjected to surface pollution. The higher the G-index of a lake, and the higher the $1/T_f$ value, the more it can be expected to be resilient to surface pollution. However, if the perturbation comes via groundwater, the lakes with a high G-index and a high $1/T_f$ will be the most sensitive. If only lakes with a G-index >50% (i.e., during one year, 50% of the total inflow comes from groundwater) and a $T_f < 5$ years (i.e., the total volume of the lake will be renewed by groundwater in less than 5 years) are considered susceptible to groundwater pollution, 75% of the studied lakes will be impacted by changes in groundwater. Lakes with a G-index >50% and a $T_f < 1$ year represent 25% of the studied lakes and are considered highly sensitive to groundwater changes (Fig. 11).

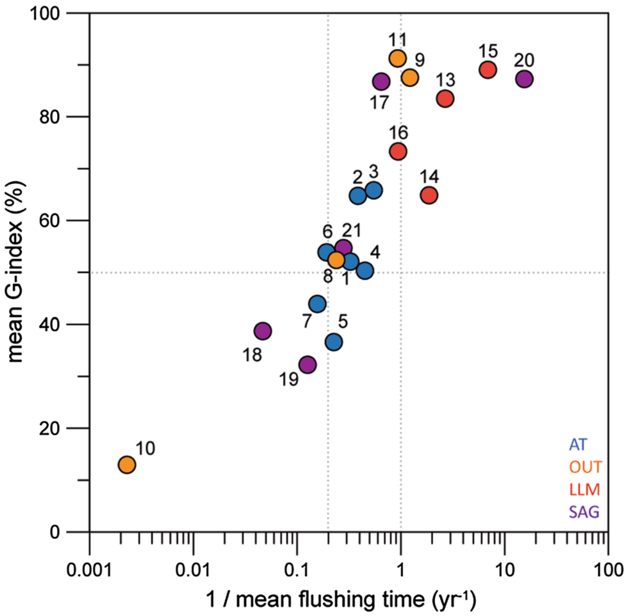


Fig. 11 Resilience of the studied lakes to surface pollution, and their sensitivity to groundwater quality or quantity changes

Conclusion

The general objective of this study was to evaluate the potential of using kettle lake geochemistry as a proxy for groundwater evolution in time and space. The geochemical and isotopic data obtained suggest that groundwater fluxes to the studied kettle lakes generally represent a major component of their water balance. The results showed that major ion concentrations remain relatively constant throughout the year, whereas the isotopic signature of the lake follows a yearly cycle. These two tracers therefore appear to be complementary methods for performing yearly hydrogeochemical budgets. Nevertheless, these two indicators are also influenced by the location of the study areas; geochemistry is influenced by the local geology, and the isotopic signatures of lake water and groundwater are influenced by the isotopic composition of precipitation, which is a function of latitude within the study region. The geochemical method is sensitive to in-lake geochemical processes, and the isotopic method is more reactive to weather conditions. The ^{222}Rn results suggest that 38% of the studied lakes have a high proportion of groundwater in their balances. The results also show that ^{222}Rn activity in several lakes is below the detection limit, even for lakes where the other tracers suggest significant input from groundwater. This suggests that the ^{222}Rn method provides information that is limited in space and time and that one sampling point is not necessarily representative of the entire lake. Moreover, the ^{222}Rn balance appears to be very sensitive to heterogeneities in ^{222}Rn distribution in groundwater, which needs to be well constrained. Finally, the studied kettle lakes are characterized by a relatively high proportion of groundwater in the total inflows (G-index >30% in 19 cases), and short to medium groundwater flushing time ($T_f < 10$ years in 18 cases). As a result, these lakes can be considered to be sensitive to environmental and climate changes affecting groundwater. To improve upon this work, it would be interesting to carry out detailed monitoring of the tracers to better understand the internal dynamics of the tracers and better constrain and adapt the lake models presented here. Moreover, a similar study on a greater number of kettle lakes would be useful for testing the statistical reliability of the current conclusions. Although the G-index describes the groundwater dependency of the studied lakes rather unambiguously, lakes with moderate G-index values could also suffer if the water table were to drop as a result of climate and/or land-use changes. Finally, the kettle lakes are, in general, sensitive to changes in both quantity and quality of groundwater, especially those characterized by a high proportion of groundwater in their balance (i.e., a high G-index value) and a short flushing time.

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