Subject: Review of Manuscript hess-2017-184

Dear Editor of Hydrology and Earth System Sciences,

On behalf of all co-authors, I would like to thank you very much for the review of our Manuscript (hess-2017-184), entitled "Impacts of changes in groundwater recharge on the isotopic composition and geochemistry of seasonally ice-covered lakes: insights for sustainable management". Thank you for considering this revision and hoping you will find it suitable for publication in *Hydrology and Earth System Sciences*.

As suggested, a figure illustrating variations of Lake Lacasse isotopic composition and a table of mean lakes isotopic compositions have been added as supplementary material. The Manuscript, revised according to the comments, is attached, as are the responses to the reviewer's comments, in blue in the following.

Thank you,

Sincerely,

Marie Arnoux

Reviewer 1:

The manuscript presents the results of a model to simulate the isotopic composition of small groundwater-connected lakes in different locations under different future climate scenarios. The approach is not new, however is interesting for the projections and the considerations discussed for future climate scenarios and recharge conditions. In particular, it provides a useful tool for improving our understanding of catchment hydrological processes. Hence, this is a nice work and warrants publication in this journal. However, I have noted a few issues that need to be addressed before the manuscript is considered for publication. Please see my specific comments below

We thank Reviewer 1 very much for considering this manuscript, and for all of the helpful comments. Please find our responses to Reviewer 1's comments below.

The authors used a lake water budget where the inflow by runoff is considered negligible. This consideration is not explained and there are not geological and hydrogeological description in the paper that could justify this. So, I suggest the author to briefly justify this sentence.

The chosen kettle lakes do not have any surface stream inflow and are set in fluvioglacial deposits. Overland flow to lakes in the study areas is considered to be low because of the permeable nature of the sandy soils. Moreover, in such a particularly cold continental climate, runoff occurs mainly during the snow melt period as well as groundwater recharge. In previous study on Lacasse lake, we have seen that runoff is negligible face to precipitation and groundwater inflows. Moreover, considering a runoff to kettle lakes negligible has been used by other authors in similar climatic contexts (Isokangas et al., 2015; Krabbenhoft et al., 1990). However we agree with the reviewer that it has to be notified in the text and keep in mind in the uncertainties of the model. Sentences have been added in the method part and in the conclusion for this assumption.

The author assumed a steady state condition for the lakes, it could be under present condition but how it's not clear how this assumption could be true when the authors run future scenario. Under climatic changes and different recharge conditions, are these assumptions satisfied or there is a range in which they could be considered valid? Please, may the authors argument better this part.

A steady state can be considered because, in cold continental climate, lake water level does not vary significantly throughout a year, and water level variation is negligible on the considered yearly time steps. Moreover, considering a steady state lake has been widely used by other authors in such cold continental climates (see, among others, Gibson et al., 2015;Yi et al., 2008;Turner et al., 2010;Kluge et al., 2012;Kluge et al., 2007;Malgrange and Gleeson, 2014).

If we consider a transient state, the balance equations become:

 $\frac{dV}{dt} = I - E - Q = cste \text{ at the considered time step}$ and $V \frac{d\delta_L}{dt} + \delta_L \frac{dV}{dt} = I\delta_I - E\delta_E - Q\delta_Q$ which gives $\frac{d\delta_L}{dt} = (I_G\delta_G + P\delta_P - E\delta_E + \delta_L(P + I_G - E - 2cste)) / V$ at the considered time

step.

The use of this equation does not significantly change the results because $P+I_G-E>>dV/dt$. In the future, changes in fluxes of the yearly water balance will not be significant enough to modify this because all parameters (P, I_G and E) should increase between 0 to 50% on a year in considered future conditions (see Figure 2 and Rivard et al. 2014). Moreover, steady state is considered on a monthly time scale for lake Lacasse because it has been already shown that, for this lake $I_G>>dV/dt$ and this will not change in the future considering water balance parameters predictions (see Arnoux et al., 2017b for more details about lake Lacasse isotopic water balance). Considering these dynamics, we agree with Reviewer 1 that considering steady state impacts the results, but we consider this to be negligible on the considered time step and assume a steady state in the calculations.

In the eq.9 (L231P9) there is the term (-B/V dt), I think that it is not correct because if eq.9 is the solution of eq.8 it means that the eq. 9 is the solution (hence without dt). Please revise or better justify this passage.

Eq 9 is the expression of δ evolution in time, depending on δ at the time step before and the considered time step. We forgot the time in the A and B terms units, that is why it was probably confusing, units have been modified in the paper (L230 P9).

The authors do not report the isotopic data, but they say that samples were collected from the top of the epilimnion and from the base of ipolimnion, in case of lake water stratification. But it's not clear what values they use in the model? Average? But in this case for evaporation what values do they use? Please detail this. I suggest also to add a table with isotopic data of lake groundwater and rain water and for Lake Lakasse a figure illustrating the variation of isotopic composition monthly. This could better show the influence of melting periods; hence the authors say that in the 8.5 scenario the isotopic composition would decrease because of melting effect, but in the text, there are not data that support these (or references). Please add data or references.

All isotopic data (precipitation, groundwater, lakes) are available in Arnoux et al, 2017a for lakes average values used in the model and in Arnoux et al., 2017b for lake Lacasse monthly values. As suggested, a figure illustrating variations of Lake Lacasse isotopic composition and a table of mean lakes isotopic compositions have been added as supplementary material.

Evaporation used comes from climate models, and δ_E is calculated with the isotopic model (cf P8). For RCP 8.5, evolution of temperature, humidity, evaporation and precipitations are illustrated on Fig 2 and show increase in precipitation, evaporation and in temperature more pronounced than RCP 4.5. A description of monthly parameter evolution regarding scenarios and melting effect can be found in Rivard et al. 2014. The text has been improved to better explain from where data used come.

Do the authors test the sensitivity of the model to investigate the dominant controls on the lake isotope system (a good reference is: Jones et al., 2016. Quaternary Science Reviews, 131:329-340)?

Thanks to Reviewer 1 for this interesting reference. Sensitivity analyses has been done on the model in the two references related to the data Arnoux et al, 2017a and b and show that the model is more sensitive to E, h and δ_G . A sentence has been added in the method part about this purpose.

May the authors describe better how they calculate or estimate evaporation (E)? What values of humidity do the authors use? (ie. from meteorological station?) Evaporation and humidity come from climate model, as described P10 and 11 and illustrated on Fig 2.

The authors repeat in the abstract, in the introduction and in the conclusion that the paper illustrated the effect of future trend on lake geochemistry, but in the paper they discuss only the isotopic composition of water and some consideration about phosphorus load. There are not discussion or results about geochemical data (ie. pH, anions, cations, alkalinity, oxygen dissolved in water. . .), so I advise the authors to add these data or discussion or to delete the sentence.

We agree with the reviewer 1 that this paper focuses only on a part of lake geochemistry evolution, which are isotopic composition and phosphorous load, and does not treat the complete lake water chemistry which was not the paper aim. As suggested by the reviewer, sentences in abstract, introduction and conclusion have been modified. However, as this paper focuses still on lake geochemistry even if it is a part, we decided to keep the title.

In my opinion, the last paragraph about phosphorous is not well connected with the previous part dealing with isotopic model and future scenario. I suggest to link these two parts. Moreover, the phosphorous geochemical behaviour should be different in stratified lake with anoxic water at the bottom. It's not so easy to estimate the quality evolution along different lakes. Do the authors consider the lake geochemistry and thermal/oxygen stratification when they discuss about P load on different lakes?

We agree with Reviewer 1 that this part is more qualitative than the rest of the paper. How recharge changes can influence P load to lakes is not often taking into account in model studies and we thing that it can be an important aspect to consider. That is why, in this paper, where we talk about how lake geochemistry can change in the future regarding recharge changes, we propose a first estimation of how P load to lake could be affected by recharge change. It is a first step for a more complex model, based on P dynamics in lakes, to determine more precisely how lake will be affect by P load changes in future. Some sentences have been added in this part to better make the link with the rest of the paper and better explain the associated assumptions.

L183P7: Is the accuracy calculated in relation to deviation of international standard? And what are the international standards used? What is the reproducibility?

 δ values are deviations in per mil (‰) from the isotopic composition of the international standard which is Vienna Standard Mean Ocean Water (VSMOW). The measurement accuracy is ± 1 ‰ vs VSMOW for δ 2H and ± 0.2 ‰ vs VSMOW for δ 18O, considering reproducibility (P7).

Is the parameter B (L230P9) m3? I think that is should be a Volume/time. Thank you to Reviewer 1 for this helpful comment, parameters units have been modified.

L271P10: Flake? Is it a typo?

Flake is the name of the lake model used in the climate simulations (see Mironov et al., 2010;Martynov et al., 2012).

Fig.4: what does the box-whisker describe? (average/median and standard deviation/confidence range/non-outlier min and max?)

The bow-whisker describes median, first and third quartiles and maximum and minimum values, this has been added to Figure legend.

L474P20: "...significant relationship..." what does it mean statistically? Do authors perform statistical test? And what?

The relationship is highlighted by the Figure 8, not by statistical tests; the sentence has been modified regarding this comment.

Fig.9: It's not clear what this figure illustrates. Do they points represent P loads? Is it the results of the model? Please, explain better what the figure wants to describe.

The figure 9 is the result of what is explain in the paragraph and is here to illustrate lakes sensitivity regarding percentage of changes in recharge and in population and therefore in P loads to lakes. The description of the Figure has been improved: The shaded area represents the scenarios for which lakes may be under risk of too high P loading, and therefore at risk of a decrease in water quality. Dots represent lakes in the four study areas for three recharge scenarios.

Reviewer 2:

Interactive comment on "Impacts of changes in groundwater recharge on the isotopic composition and geochemistry of seasonally ice-covered lakes: insights for sustainable management" by Marie Arnoux et al. Anonymous Referee #2 Received and published: 16 July 2017

General comments: The authors present an interesting study of the variability of the isotopic composition and geochemistry in kettles lakes due to the future variability of recharge and climate. In this aim, the authors compare the measured δ 18O and δ 2H in several kettles lakes at annual and monthly intervals and the modeled δ 18O and δ 2H. The modeled isotopic composition of lake is estimated from climate and estimation recharge models. The modeling results are used to determine if the future evolution of the climate and the recharge could modify the isotopic signature of lake and if the isotopic monitoring in lakes could be an efficient tool to highlights the variability of water budget and quality.

The modeling results have be well analyzed and interpreted, and the authors explain well the assumptions and the limits of their results. The authors study also the water quality but only by the phosphorous. This part, for me, is not really on the topic of this article, less argue than the part about isotopic signature, and maybe not necessary.

Specific comments: The paper is relatively clear, well written, well structured. Nevertheless, some parts are too long and descriptive and has to modify for a better understanding, notably in the part of results and discussion.

We thank Reviewer 2 very much for considering this manuscript, and for all of the helpful comments. Please find our responses to Reviewer 2's comments below.

Abstract: The abstract is completed and structured, nevertheless the scientific problematic is not really highlighted, could you add a sentence explaining more clearly the problematic of the paper.

Thanks to the Reviewer 2 for this comment, a sentence has been added in the abstract.

Introduction: Line: 86-88: the interest of this sentence and the link with the end of this paragraph is not clear. Please modify this sentence. The study is based on kettle lakes, this methodological choice should be exposed in the introduction.

Thanks to the Reviewer 2 for this comment, the sentence has been modified and a sentence has been added about kettle lakes.

Methods: Line 187-190 : the sentence is not clear; please modify it. The sentence has been modified.

Water mass balance: several assumptions (Is=0, Ir=0) has not justified, could you please add a sentence to justify this hypothesis.

The chosen kettle lakes do not have any surface stream inflow that is why Is=0. Moreover they are set in fluvioglacial deposits, therefore overland flow to lakes in the study areas is considered to be low because of the permeable nature of the sandy soils. Moreover, in such a

particularly cold continental climate, runoff occurs mainly during the snow melt period as well as groundwater recharge. In previous study on Lacasse lake, we have seen that runoff is negligible face to precipitation and groundwater inflows (Arnoux et el. 2017b). Moreover, considering that runoff to kettle lakes is negligible has been used by other authors in such similar climatic contexts (see Isokangas et al 2015; Krabbenhoft et al 1990). However we agree with the reviewer that it has to be notified in the text and keep in mind in the assumption of the model. Sentences have been added in the method part and in the conclusion for this assumption.

Line 251-254: this sentence is not clear; please modify it. The sentence has been modified.

Paragraph evolution scenarios: an introductive sentence could allow a better understanding of this paragraph reminding the interest and using of these models in the study. Sentences have been added.

Line 296-297: Please explain the interest to work with two period, a reference period and future period. Indeed, the reference period is largely in the future. Please explain moreover the choice of 2040 for the transition between these two periods.

This choice has been made because of recharge predictions from Rivards et al. 2014 which are on a reference period, based on actual measurement, and a future 2041-2070 period, therefore to use these data it was necessary to work on a reference period close to present and on 2041-2070 for future period. Also, the reference period has been chosen to cross the two years 2015-2016 field campaign in order to calibrate the model. Furthermore, we decided to choice the same time duration for these two compared periods (to have same signification on means) and the same model for climate data (for the consistency of modelling), that is why we use the 2010-2040 period as the reference period.

Figure 2: what represent the dotted line? The dotted line is just a mark to facilitate the reading.

Fig. 3: It's difficult to understand which model is used, could you clarified this in the caption. In the text, we can suppose that the fig.3a is a result of the publication Arnoux et al., 2017b, if it is the case, could you add the citation in the caption? The Figure 3 caption has been clarified.

Results and discussion: Monthly evolution of lake isotopic composition

Line 373: please, remind quickly how the G-index is measured.

It has been added to the text.

Fig. 4: the interest of the close-up is relatively low, without its, the figure will be clearer.

We agree with the reviewer, however, we decided to keep this representation to show to the reader the range of variations of our results and on what are based the means.

Fig. 5, line 389-393: the link between the figure and the interpretation is not clear. We talk about on one hand of reference period on the other hand of the future period while in the figure, the difference between reference period and future period is illustrated.

As the reference period is the same for all future scenarios (S0, S1, S2 and NC), difference between reference and future with changes ($\Delta \delta^{18}$ O S0, S1 and S2) can be compared to difference between reference and future with no change ($\Delta \delta^{18}$ O NC) which is equivalent to a comparison scenarios regarding no change in future. But as suggested by the reviewer, the text has been modified to be clearer.

Annual isotopic signature evolution, isotopic signature evolution.

This paragraph is not clear. Indeed, first, line 456-458 the authors explains that lakes with a low G-index and a small volume have higher potential variability in isotopic composition than those with a high G-index and high volume but to illustrate the remark, they used two lakes with a similar mean G-index. Secondly, line 463 to 464, the authors write that "when lakes have a high G-index, the groundwater flux tends to buffer lake isotopic variations, and so they tend to be less sensitive to changes in climate data", but the authors don't give some arguments (results or figure). Please, be clearer. Furthermore, this sentence is not consistent with the figure 8, and the explanation line 476 to 477 " lake isotopic composition is more sensitive to changes in recharge for G-indices ranging from 50 to 80%, with a maximum of sensitivity observed for a G-index of around 65 %. Please clarified this paragraph.

Thanks to the reviewer 2 for this comment, this paragraph has been clarified: lakes with a low G-index and a small volume have higher potential variability in isotopic composition regarding climate variability (evaporation and precipitation) while lake with G-indices ranging from 50 to 80% have an isotopic composition more sensitive to changes in recharge. We explain first the variability regarding climatic parameters and then, regarding changes in recharge.

Lake quality evolution

This part of the article is disconnected of the other results, where the isotopic variability is analyzed. The scientific interest of the part about the P is really lesser than the rest of the article and not necessary.

We agree with the Reviewer 2 that this part is more qualitative than the rest of the paper but we decided to keep it in the paper because how recharge changes can influence P load to lakes is not often taking into account in model studies and we thing that it can be an important aspect to consider. That is why in this paper, where we talk about how lake geochemistry can change in the future regarding recharge changes, we propose a first estimation of how P load to lake could be affected by recharge change. It is a first step for a more complex model, based on P dynamics in lakes, to determine more precisely how lake will be affect by P load changes in future. Some sentences have been added in this part to better make the link with the rest of the paper.

Conclusion: This part is clear and well structured. Just, please highlied that when you talk about water quality you study only the evolution of P. Moreover, the sentence, line 573-575, underlines that the part about P is based on several assumptions (not exposed in the article) and that this part is maybe not necessary on this article.

The assumptions about lake quality evolution have been added in the lake quality evolution part.

Technical corrections: Line 188 : two weeks Line 205: avoid that the (δp) is not at the same line that precipitation. Line 211: the equation is in subscript. Line 263: two time-levels Line 333: add parenthesis for Rivard et al., 2014, same line 343. Line 364: check the English Figure 6: be careful the indicated period is different between the text and the caption. Line 462: be careful for the reading of the lake volume. Same line 466 Thanks to the reviewer, the technical corrections have been done.

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Impacts of changes in groundwater recharge on the isotopic composition and geochemistry of seasonally ice-covered lakes: insights for sustainable management

Marie Arnoux^{1,2}, Florent Barbecot¹, Elisabeth Gibert-Brunet², John Gibson³, Aurélie Noret²

¹ GEOTOP, Université du Québec à Montréal, Montréal, Québec, Canada H3C 3P8
² GEOPS, UMR 8148, CRNS-Université Paris Saclay/Paris-Sud, Orsay, France
³ Alberta Innovates Technology Futures, 3-4476 Markham Street, Victoria, BC V8Z 7X8, Canada

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Corresponding author:

Marie Arnoux (marie.arnoux@u-psud.fr) +33 6 79547616 GEOTOP, Université du Québec à Montréal, Montréal, Québec, Canada H3C 3P8 GEOPS, UMR 8148, CRNS- Université Paris-Saclay/Paris-Sud, Orsay, France

Florent Barbecot (barbecot.florent@uqam.ca) GEOTOP, Université du Québec à Montréal, Montréal, Québec, Canada H3C 3P8

Elisabeth Gibert-Brunet (elisabeth.gibert@u-psud.fr) GEOPS, UMR 8148, CRNS- Université Paris Saclay/Paris-Sud, Orsay, France

John Gibson (jjgibson@uvic.ca) Alberta Innovates Technology Futures, 3-4476 Markham Street, Victoria, BC V8Z 7X8, Canada

Aurélie Noret (aurelie.noret@u-psud.fr) GEOPS, UMR 8148, CRNS- Université Paris Saclay/Paris-Sud, Orsay, France

<u>ABSTRACT</u>

Lakes are under increasing pressure due to widespread anthropogenic impacts related to rapid development and population growth. Accordingly, many lakes are currently undergoing a systematic decline in water quality. Recent studies have highlighted that global warming and the subsequent change in water use may further exasperate eutrophication in lakes. Lake evolution depends strongly on hydrologic balance, and therefore on groundwater connectivity. Groundwater also influences the sensitivity of lacustrine ecosystems to climate and environmental changes, and governs their resilience. Improved characterization of groundwater exchange with lakes is needed today for lake preservation, lake restoration, and for sustainable management of lake water quality into the future. In this context, the aim of the present paper is to determine if the future evolution of the climate, the population and the recharge could modify the geochemistry of lakes (mainly isotopic signature and quality via phosphorous load) and if the isotopic monitoring in lakes could be an efficient tool to highlights the variability of water budget and quality.

Small groundwater-connected lakes were chosen to simulate changes in water balance and water quality expected under future climate change scenarios, namely Representative Concentration Pathways (RCP) 4.5 and 8.5. Contemporary baseline conditions, including isotope mass balance and geochemical characteristics, were determined through an intensive field-based research program prior to the simulations. Results highlight that future lake geochemistry and isotopic composition trends will depend on four main parameters: location (therefore climate conditions), lake catchment size (which impacts the intensity of the flux change), lake volume (which impacts the range of variation), and lake G-index (i.e., the percentage of groundwater that makes up total lake inflows), the latter being the dominant control on water balance conditions, stable isotopes appear to be especially useful for detecting changes in recharge to lakes with a G-index of between 50% and 80%, but response is non-linear. Simulated monthly trends reveal that evolution of annual lake isotopic composition can be dampened by opposing monthly recharge fluctuations. It is also shown that changes in water quality in groundwater-connected lakes depend significantly on lake location and on the intensity of recharge change.

1. INTRODUCTION

For decades, climate change, combined with rapidly expanding urban, industrial, and agricultural water needs, has placed increasing stress on water resources and on groundwater resources in particular. Future pressure on these resources is likely to be even more pronounced, as groundwater is likely to be increasingly exploited to enhance water supply and to alleviate the worsening drought situation in some arid regions (Dragoni and Sukhija, 2008). Many studies have suggested that sustainable groundwater use has to be based on, among other things, a reliable assessment of recharge, which largely controls its evolution. Aquifer recharge refers to the quantity of water reaching the saturated zone of an aquifer, and therefore replenishing the water table. Unfortunately, in many parts of the world, recharge rates are often not well-known at the regional scale (Rivard et al., 2013). While aguifer recharge is crucial to supporting sustainable management of regional groundwater resources, it is difficult to accurately estimate, owing mainly to limited data availability, as well as limitations inherent to estimation methods and field measurements (Rivard et al., 2013). Recharge rates are controlled by geology, soil characteristics, topography, land cover, land use and climate (Rivard et al., 2014). Thorough literature reviews of the various techniques that exist to quantify groundwater recharge are provided in Scanlon et al. (2002) and Healy (2011). Many methods can be used to estimate groundwater recharge, such as water budget methods, modelling methods, tracer methods, and methods based on surface water interaction studies. The latter is based on the estimation of groundwater discharge to surface water, mainly by streambed seepage determination, stream flow duration curves, or stream flow hydrograph separation (Scanlon et al., 2002). The recharge amount (in mm.yr⁻¹) is then typically obtained by dividing measured or estimated discharge flow by the surface drainage area at the measurement site. This procedure assumes that aquifer boundaries coincide with watershed boundaries, and consequently that the area of the aquifer that contributes to groundwater discharge is equal to the surface drainage area (Kuniansky, 1989; Rutledge, 1998, 2007). However, this assumption must be considered carefully, as groundwater basins and watershed boundaries can differ drastically (Tiedeman et al., 1997). Miscalculation of the aquifer contributing area will lead to a proportional error in recharge estimate.

Although the groundwater inflow to streams is often taking into account in water budgets, it is less commonly considered for surface water bodies, probably due to the greater difficulty of quantifying groundwater discharge in these settings. However, in recent years some studies have proven that groundwater flow into lakes can be reliably quantified. Interactions between lakes and groundwater depend on geology, soil and sediment properties, and also on hydraulic gradient, which is strongly

dependent on climatic conditions and recharge (Winter, 1999). Therefore, variation in groundwater fluxes may indicate a change in recharge in the lake catchment (Meinikmann et al., 2013).

In Quebec (Canada), more than ten percent of the surface is covered by freshwater, with more than one million lakes known to exist. In many cases, these are connected to underlying aquifers. However, lake-groundwater interactions are highly dynamic throughout the year, and, even if it now possible to quantify groundwater inflow with a reasonable degree of confidence, it is difficult to determine how and to what extent lakes can be sensitive to changes in groundwater recharge. The lake water isotopic composition has been proven to be particularly useful for determining water balance parameter controls under changing conditions. For example, as shown in Turner et al. (2010), lake isotopic composition can highlight that (i) reduced winter precipitation could cause snowmelt-dominated lakes to become rainfall-dominated lakes, or that (ii) during longer ice-free seasons, mainly rainfall-dominated, but also potentially snowmelt-dominated lakes, may turn into evaporation-dominated lakes. Moreover, among all the methods used to quantify groundwater flux variations on seasonal and yearly time scales (Arnoux et al. 2017a). Water stable isotopes are therefore expected to be very useful for monitoring seasonal and inter-annual variations in the water budget under changing recharge conditions.

The impact of climate change on groundwater recharge is not easy to determine, because of the complexity of interactions and processes evolved, and can varies vastly depending on regions (Rivard et al. 2014; Crosbie et al., 2013). In addition, it is predicted to shift differentially under various climate scenarios and models (Jyrkama and Sykes, 2007; Levison et al., 2014). In Canada, highly variable recharge rates have been proposed in previous studies; for example, for the 2050 horizon (mainly the period 2041-2070) relative to modern (2000-2015) or past recharge rates (1950-2010), depending on study site, scenario, and model: +10 to +53% in the Grand River watershed, Ontario (Jyrkama and Sykes, 2007), -41 to +15% in the Chateauguay River watershed, Quebec (Croteau et al., 2010), –6 to +58% in the Otter Brook watershed, New Brunswick (Kurylyk and MacQuarrie, 2013), -4 to +15% at Covey Hill, Quebec (Levison et al., 2014), +14 to +45% in the Annapolis Valley, Nova Scotia (Rivard et al., 2014), and -28 to +18% for the Magdalen Islands, Quebec (Lemieux et al., 2015).

Recharge fluctuations can also impact lake water quality by changing groundwater fluxes, which are closely linked to phosphorous (P) loading to lakes. It is known that lake water quality is mainly driven by variations in P load, since this plays a critical role in limiting lake primary productivity and algal biomass, which in turn regulate lake trophic status. Increasing P concentration in the water column is the primary factor responsible for accelerated eutrophication and associated algae blooms (Schindler, 1977; Wang et al., 2008). At sites without urban drainage or point P sources, such as sewage treatment

plants, domestic waste from septic systems may represent the largest anthropogenic source of P to lakes on the Canadian Shield (Dillon and Evans, 1993). Increases in shoreline development and population, combined with groundwater fluxes variations, can clearly impact lake quality, but still remain to be quantified.

For the present study, ten lakes in southern Quebec were sampled to quantify their yearly groundwater inflows (see Arnoux et al., 2017a for more details), and one of these lakes was sampled over the course of a year to quantify its monthly groundwater inflows (see Arnoux et al., 2017b for more details). Small kettle lakes without surface inlets set in fluvioglacial deposits, and that are most likely well connected to shallow unconfined aquifers, are specifically targeted. The main objectives of this study were (i) to determine how future groundwater recharge changes might affect lake water balance and geochemistry, and (ii) to assess whether stable isotopes might be an effective tool for identifying lakes that are susceptible to change or are undergoing changes in water balance and water quality. To address these objectives, seasonal models of water and isotopic budgets were established for several lakes, and the models were then forced with future yearly and monthly time scale climate data from predictive global models to simulate anticipated conditions. Climate outputs of the Canadian Regional Climate Model were used, based on scenarios RCP 4.5 and RCP 8.5 (Moss et al., 2010; IPCC, 2014). It is assumed that recharge fluctuation is the main parameter influencing groundwater fluxes into lakes, and thus a percentage of recharge change will lead to the same percentage of change of groundwater fluxes to lakes. Different recharge scenarios, which translate into changes in groundwater inflow, were then tested to determine changes in water budget and isotopic evolution of the lakes. Predicted changes in recharge were then compared to predicted population growth in the study areas to discuss lake quality evolution. After determining the evolution of the lake geochemical signature, how lakes connected to groundwater can be used to identify changes in groundwater recharge can be determined, as can whether or not the isotopic composition of lakes can serve as an effective indicator of change or variability.

2. <u>Method</u>

2.1. Study sites

The ten lakes chosen are located in four regions of southern Quebec characterized by contrasting climatic conditions: Laurentides (LAU), Outaouais (OUT), Abitibi-Témiscamingue (AT), and Saguenay-Lac –Saint-Jean (SAG). These kettle lakes, set in coarse-grained (sand/gravel) fluvioglacial deposits, are specifically targeted in this study, because they (i) are small enough to be sensitive to environmental changes on a short time scale, (ii) do not have permanent surface inflow streams, and so are largely groundwater dependent, (iii) are generally characterized by predictable and uniform geomorphological features, and (iv) are likely connected to shallow, unconfined aquifers (Arnoux et al. 2017a; Isokangas et al., 2015). Kettle lakes originate as depressions in the landscape formed following the melting of ice blocks buried in the ground after glacial retreat of the Late Glacial to Holocene transition period (from -12 to -7 kyr). These kettle holes, becoming kettle lakes when they are filled with water, are mainly found in fluvioglacial deposits, such as outwash plains, deltas, eskers, and kame terraces (Benn and Evans, 2011). Figure 1 shows the locations of the ten lakes analyzed here. Their main characteristics are described in Table 1.



Fig. 1. Locations of the study lakes (circles) and sources of climate data (triangles)

2.2. Lake isotopic composition

2.2.1.Sampling

Water samples from each lake were retrieved during two field campaigns, in June-July and October-November 2014. When physicochemical parameters, measured *in situ* along the water column, revealed a well-mixed lake, the lake was considered to be homogeneous, and only one sample was collected, from close to the lake bottom, at its greatest depth. Otherwise, for stratified periods, two samples were collected: one from the top of the epilimnion and one from the base of the hypolimnion, in order to obtain the complete range of isotopic composition variation. Whenever possible, groundwater was sampled from private wells located in the vicinity of the studied lakes. Untreated groundwater samples were collected from residential wells from the tap after purging approximately three times the well volume.

Samples were transported in a cooler, and subsequently stored at 5°C until analyses were performed. Water stable isotopic compositions were measured with a Laser Water Isotope Analyser (OA ICOS DLT, Los Gatos Research, now ABB) at the GEOPS Laboratory (University of Paris-Sud/Paris-Saclay, France). The measurement accuracy is $\pm 1 \%$ vs VSMOW for δ^2 H and $\pm 0.2 \%$ vs VSMOW for δ^{18} O. Results are reported in δ values, representing deviations in per mil (‰) from the isotopic composition of the international standard (Vienna Standard Mean Ocean Water, VSMOW), such that δ^2 H or δ^{18} O=((R_{sample}/R_{VSMOW})-1)×1000, where R refers to ²H/H or ¹⁸O/¹⁶O ratios.

One of the lakes, Lake Lacasse, was sampled in more detail throughout 2015-2016. Water samples were collected from the lake at two weeks to one month intervals, mainly from the deepest part of the lake, and at 1 to 2 meter depth intervals in order to monitor the vertical heterogeneity of the water column. Groundwater was sampled twice from eight private wells in the vicinity of the lake (see Arnoux et al, 2017b for more detail).

2.2.2. Water mass balance

The lake water budget is defined as:

$$\frac{dV}{dt} = I - E - Q$$
 Eq. (1)

where V is the volume of the lake (m³); t is time (days); E is evaporation (m³.day⁻¹); I is the instantaneous inflow (m³.day⁻¹), corresponding to the sum of upstream surface inflow (I_S; zero for the studied lakes because they do not have surface inlets), runoff (I_R; considered negligible because of the permeable nature of the sandy soils of kettle lakes), groundwater inflow (I_G), and precipitation on the lake surface (P); Q is the outflow (m³.day⁻¹), which is the sum of surface (Q_S) and groundwater (Q_G)

outflows. Under constant atmospheric and hydrologic conditions, steady state is assumed (Gibson et al., 2016), implying that dV/dt=0. Therefore $I_G = Q_S + Q_G + E-P$ for the entire lake.

2.2.3. Stable isotopic mass balance

Considering water stable isotopes, the lake isotopic mass balance is:

$$V\frac{d\delta_L}{dt} + \delta_L \frac{dV}{dt} = I\delta_I - E\delta_E - Q\delta_Q \text{ Eq. (2)}$$

where δ is isotopic composition of: the lake (δ_L ; equals to the mean if the lake is stratified - see Arnoux et al. 2017a and b for more details about lake isotopic compositions used in the model), total inflow (δ_I), which include runoff (δ_R), precipitation (δ_P), surface inflow (δ_S) and groundwater inflow (δ_G), and total outflow (δ_Q), which include surface (δ_{QS}) and groundwater (δ_{QG}) outflows. The isotopic composition of evaporating water (δ_E) was estimated using the Craig and Gordon (1965) model, expressed by Gonfiantini (1986) as:

$$\delta_E = \frac{(\delta_L - \varepsilon^+) / \alpha^+ - h \delta_A - \varepsilon_K}{1 - h + 10^{-3} \varepsilon_K} \quad \text{Eq. (3)}$$

where h is the relative humidity at the lake surface; δ_A is the local isotopic composition of the atmospheric moisture (‰); α^+ is the equilibrium isotopic fractionation;

 $\varepsilon^{+} = (\alpha^{+} - 1) * 1000$ is the equilibrium isotopic separation (‰);

 $\varepsilon_{\kappa} = C_{\kappa} (1-h)$ is the kinetic isotopic separation (‰), with C_K being the ratio of molecular diffusivities between heavy and light molecules (Gibson et al., 2016).

In this study, C_{K} values were considered to be representative of fully turbulent wind conditions and a rough surface for both oxygen (C_{K} =14.2‰) and hydrogen (C_{K} =12.5‰), based on experimental data (Horita et al., 2008). For calculating equilibrium fractionation factors, experimental values of Horita and Wesolowski (1994) were used:

$$\alpha^{+}(^{18}O) = \exp(-7.685/10^3 + 6.7123/T - 1666.4/T^2 + 350410/T^3)$$
 Eq. (4)

$$\alpha^{+}(^{2}H) = \exp(1158.8 \times T^{3} / 10^{12} - 1620.1 \times T^{2} / 10^{9} + 794.84 \times T / 10^{6} - 161.04 / 10^{3} + 2999200 / T^{3}) \text{ Eq. (5)}$$

where T is temperature (K). The isotopic composition of atmospheric moisture (δ_A , ∞) was calculated assuming equilibrium isotopic exchange between precipitation and vapor:

$$\delta_{A} = \frac{\delta_{P} - \varepsilon^{+}}{1 + 10^{-3} \varepsilon^{+}}$$
 Eq. (6)

where δ_{P} (‰) is the mean annual isotopic composition of precipitation. Assuming well-mixed conditions in the lake, the combination of Eq. (3) and Eq. (2) yields:

$$V\frac{d\delta_L}{dt} + \delta_L\frac{dV}{dt} = P\delta_P + I_G\delta_G - Q\delta_L - \frac{E}{1 - h + 10^{-3}\varepsilon_K} \left(\frac{\delta_L - \varepsilon^+}{\alpha^+} - h\delta_A - \varepsilon_K\right) \text{ Eq. (7)}$$

A steady state was assumed, such that dV/dt=0. Equation (7) can therefore be simplified to:

$$V\frac{d\delta_L}{dt} = P\delta_P + I_G\delta_G - (P + I_G - E)\delta_L - \frac{E}{1 - h + 10^{-3}\varepsilon_K} \left(\frac{\delta_L - \varepsilon^+}{\alpha^+} - h\delta_A - \varepsilon_K\right) \text{Eq. (8)}$$

Resolving this calculation therefore allows isotopic composition of the lake water at time t+dt to be determined, expressed as a function of its value at the previous time step, t, and two established parameters, A (∞ .m³/yr) and B (m³/yr):

$$\delta_L^{t+dt} = \frac{A}{B} + (\delta_L^t - \frac{A}{B}) \exp(-\frac{B}{V} dt) \text{ Eq. (9)}$$

with

$$A = P\delta_{P} + I_{G}\delta_{G} - \frac{E}{1 - h + 10^{-3}\varepsilon_{K}} \left(-h\delta_{A} - \varepsilon_{K} - \varepsilon^{+} / \alpha^{+}\right) \text{ Eq. (10)}$$
$$B = P + I_{G} - E\left(1 - \frac{1}{\alpha^{+}(1 - h + 10^{-3}\varepsilon_{K})}\right) \text{ Eq. (11)}$$

The monthly mean isotopic composition of precipitation (δ_P) was assessed in the four regions from the Global Network of Isotopes in Precipitation (GNIP) and Program for Groundwater Knowledge Acquisition (PACES) datasets. Future δ_P trends are uncertain; however, they have been shown to be mainly dependent on temperature evolution and local factors (Stumpp et al., 2014), and a recent study in Siberia showed that a long term increase in precipitation $\delta^{18}O$ is close to the detection limit of the tracers (<1‰ per 50 years) (Butzin et al., 2014). Monthly current means were therefore used in the current simulations. The mean value of groundwater isotopic composition (δ_{Gi}) was determined from the mean groundwater isotopic composition measured in wells, located in the same region and presenting no enrichment due to evaporation. The mean isotopic values used for groundwater are presented in Table 2.

The uncertainties associated with the Craig and Gordon (1965) model in the estimated isotopic composition of evaporating moisture (δ_E) can be substantial, especially if relative humidity is greater than 0.8 (Kumar and Nachiappan, 1999). Moreover, a sensitivity analysis of ¹⁸O isotopic balance of a small lake in Austria (Yehdegho et al., 1997) indicates that for flow-though, groundwater-dominated systems with limited evaporation, the isotopic composition of the lake water and the inflow water are the parameters critical to the overall uncertainty. Horita et al. (2008) recommended using time-averaged values of the parameters in the calculation of δ_E for the given period of interest. Moreover atmospheric parameters should be preferably evaporation-flux weighted whereas liquid fluxes to a lake should be amount-weighted (Gibson, 2002; Gibson et al., 2016). Therefore, on an annual time step, δ_P is monthly

precipitation-flux weighted, except when it is used to estimate δ_A ; in this case, δ_P is monthly evaporation-flux weighted. At a monthly time scale, monthly values are used for each parameter of the model, and evaporation is considered to be null during the ice-covered period. Moreover, in winter, when monthly mean temperature is below zero, precipitation is assumed to be zero in the model. Then, when monthly temperature becomes equal to or higher than zero, accumulated precipitation and amount-weighted δ_P are added to the calculation during the melt period. Moreover, sensitivity tests on this model have performed in Arnoux et al. 2017a and b and show that it is mostly sensitive to E, h and δ_G .

2.3. Evolution scenarios

2.3.1. Climate models

Climatic parameters used in this study (evaportation, humidity, temperature and precipitation) come from climate models. RCMs allow the downscaling of large-scale information from GCMS to scale closer to watershed scale, leading to a better representation of surface forcings. In the present study, the fifth version of the Canadian RCM (CRCM5) was chosen, which has a 0.44° horizontal grid resolution (approx. 50 km; Sushama et al., 2010; Martynov et al., 2013; Šeparović et al., 2013). The CRCM5 is a grid-point model, based on a two time-level, semi-Lagrangian, (quasi) fully implicit time discretization scheme (Alexandru and Sushama, 2015). The model includes a terrain-following vertical coordinate based on hydrostatic pressure (Laprise, 1991; Alexandru and Sushama, 2015), and an horizontal discretization on a rotated latitude-longitude, Arakawa C grid (Arakawa and Lamb, 1977; Alexandru and Sushama, 2015). Following CRCM4, changes that have been introduced into CRCM5 include, for example, evolution in the planetary boundary layer parameterization to suppress both turbulent vertical fluxes under very stable conditions and the interactively coupled one-dimensional lake model (Flake; Mironov et al., 2010; Martynov et al., 2012; Šeparović et al., 2013). CRCM5 uses the Canadian Land-Surface Scheme (CLASS, version 3.5; Verseghy, 1991; Alexandru and Sushama, 2015). This model is described in detail in Martynov et al. (2013) and Šeparović et al. (2013).

The CRCMs were driven by the second-generation Canadian Earth System Model (CanESM2, improved from CanESM1; Arora et al., 2011), developed by the Canadian Center for Climatic Modeling and Analysis (CCCma). As explained in Šeparović et al. (2013), it consists of a fourth-generation atmospheric general circulation model CanAM4, coupled with (i) the physical ocean component OGCM4 developed from the NCAR CSM Ocean Model (NCOM; Gent et al., 1998), (ii) the Canadian Model of Ocean Carbon (CMOC; Christian et al., 2010), and (iii) Canadian Terrestrial Ecosystem Model (CTEM; Arora and Boer, 2010). The CanAM4 is a spectral model employing T63 triangular truncation with

physical tendencies calculated on a 2.81 linear grid and 35 vertical levels (Arora et al., 2011; Šeparović et al., 2013).

2.3.2. Climate data

Four greenhouse gas concentration scenarios (Representative Concentration Pathways, RCP) have been adopted by the IPCC in its fifth Assessment Report (AR5) in 2014: RCP 2.6, RCP 4.5, RCP 6.0, and RCP 8.5. The scenarios selected for the present study are RCP 4.5 and RCP 8.5, for which predicted climate data are available until 2100 for the study regions. The RCP 4.5 scenario considers that long-term global emissions of greenhouse gases and land-use-land-cover stabilize radiative forcing at 4.5 W.m⁻² (approximately 650 ppm CO₂-equivalent) by the year 2100, without ever exceeding that value. The RCP 8.5 scenario corresponds to the highest greenhouse gas emissions pathway scenario, with gas emissions and CO₂ concentrations increasing considerably over time, and thus leading to a radiative forcing of 8.5 W.m⁻² by the end of the century (approximately 1370 ppm CO₂ equivalent). The defining characteristics of these scenarios are enumerated in Moss et al. (2010).

In order to connect these RCP forecasts to our study and to visualize trends, yearly mean data are presented in Fig. 2. Based on previous literature on recharge changes (see part 2.2.3.), a reference period (2010-2040) is compared to a future period (2041-2071). It is noted that both evaporation and temperature display increases between the reference and future periods for both scenarios, although it is more pronounced for RCP 8.5. Moreover, precipitation and relative humidity do not show clear trends. However, it seems that precipitation variability will increase overall for both scenarios, although this is more pronounced for RCP 8.5. Moreover, the southern regions (i.e., OUT and LAU) have higher temperatures than the northern regions (i.e., AT and SAG), and precipitation is higher in LAU than in the other three regions. On a monthly time scale, surface temperatures in LAU show an increasing monthly trend, whereas evaporation increases mainly during summer and stays relatively constant the rest of the year (data not shown). Meanwhile, precipitation does not show any clear trend. However, as temperatures increases in winter, melt periods likely will shift more frequently occur earlier in the year.



Fig. 2. Climate data for the reference (Ref; 2010-2040) and future (Fut; 2041-2071) periods, obtained from CRCM5 – CanESM2, with RCP 4.5 (left) and RCP 8.5 (right) scenarios for the four different study areas. The variables are: a) surface air temperature, b) surface water evaporation (obtained from surface heat flux), c) surface relative humidity (obtained from surface specific humidity), and d) precipitation (Martynov et al., 2013; Šeparović et al., 2013); bow-whiskers describe median, first and third quartiles and maximum and minimum values.

2.3.3. <u>Recharge evolution</u>

The mean annual recharge for each lake basin was obtained by dividing the lake drainage area by the calculated mean annual groundwater inflow to the lake (Meinikmann et al., 2013). In this study, recharge evolution is thus expressed in terms of changes in groundwater inflow to the lakes.

In the first step, recharge is assumed to be constant for the 2006-2014 period. Over this period, recharge is adjusted to fit the calculated lake isotopic compositions to those measured. In the second step, the results of Rivard et al. (2014) was chosen for the simulation of recharge scenarios, since this study focusses on the Annapolis Valley (Nova Scotia, Canada), not far from southern Quebec and with a similar latitude, geology, and climate. Therefore, the future recharge dynamics determined for the Annapolis valley are assumed to be similar to those of the present study sites. Rivard et al. (2014) found that all scenarios predict an annual recharge to the aquifer within the range of +14 to +45% higher than at present by 2041-2071. They also predict, on a seasonal basis, that recharge will undergo (i) a marked decrease in summer (from 4 to 33%), and (ii) a spectacular increase in winter (more than 200%), due to an earlier melt period starting date.

The following section focussed firstly on monthly lake isotopic composition evolution (Part 3.1.) and secondly on yearly lake isotopic composition evolution (Part 3.2.). Monthly and yearly values are compared for the two standard periods (i.e., for reference (2010-2040) and future (2041-2071) periods).

For the first part of the study, Lake Lacasse, located in the LAU region, has been chosen, since it was subject to continuous monitoring (Arnoux et al., 2017b). Its groundwater inflow and variability has therefore already been well-constrained throughout the year 2015-2016 (Fig. 3 b). For this lake, the model was run from 2006 to 2071, and four different recharge evolution scenarios were applied to the 2041-2071 period, following the predictions of Rivard et al. (2014) for scenarios S1 and S2, as described below.

- NC: no change in recharge (groundwater inflow follows the pattern described in Fig. 3, obtained from Arnoux et al., 2017b);
- S0: a recharge decrease of 33% during the summer period (from June to October);
- S1: a 200 % increase in recharge during the melt period (from January to March), and a 4% decrease in the summer period;
- S2: a 200 % increase in recharge during the melt period, and a 33% decrease during the summer period.

For the second part, three annual recharge evolution scenarios were tested, following the predictions of Rivard et al. (2014): no change (NC), a 14% increase (Low), and a 45% increase (High) in mean annual recharge.

2.4. Population growth

Variations in the quantity and/or quality of groundwater feeding lakes can obviously impact the geochemistry, and thus the water quality of lakes, especially for lakes displaying a high G-index (the percentage of groundwater comprising the total lake inflow; Arnoux et al., 2017a). Moreover, in rural areas of Quebec, lake and groundwater quality is likely to be influenced by changes in population density. The population of Quebec is aging, and many seasonal residences (e.g., cottages) around lakes in rural areas are expected to become year-round residences. Furthermore, these residences are not connected to waste water treatment plants; rather, owners have their own private wells for drinking water and private septic tanks with subsurface seepage beds for waste water. The predicted population changes are summarized in Table 3. Population is mainly expected to increase in the southern regions (OUT and LAU), with a mean increase of 24 and 28% respectively (ISQ, 2014; Table 3). Scenarios of population growth are compared with scenarios of recharge evolution for each lake to assess their future quality evolution.

3. RESULTS AND DISCUSSION

3.1. Monthly evolution of lake isotopic composition

Figure 3 shows the measured (see Arnoux et al. 2017b for more details about measured values) and modelled isotopic compositions of Lake Lacasse. It can be observed that the modelled values are more variable than the measured ones, undoubtedly due to the higher evaporation rate in the climatic model (459 mm) than that measured during the field monitoring period (204 mm). It is also shown that the model attributes greater weight to the contribution of the depleted snow value than in reality. This is probably due to the snow column (which is close to 0°C during the snow melt) being less dense than the lake surface water (which has a mean temperature of close to 4°C), and therefore bypasses the lake, flowing rapidly out of the lake outlet. In such a case, the snow does not influence the lake isotopic composition as much as the model predicts. Since similar results are obtained for δ^2 H values, only the δ^{18} O results from the model will be presented in the following sections.



Fig. 3. (a) Isotopic composition of Lake Lacasse between June 2015 and May 2016, measured and modelled from stable isotopic mass balance model using climate data from climate model CRCM5 –CanESM2 and scenario RCP 4.5; (b) the pattern of groundwater inflow (I_G) to Lake Lacasse (Arnoux et al., 2017b).

Lake Lacasse has a mean G-index (i.e. the percentage of groundwater in total lake inflows) of 69% during the reference period. Results for monthly simulations, with RCP 4.5 climate data, are illustrated in Fig. 4. Lake isotopic compositions are not significantly different between the reference and future periods if no change is applied to the recharge pattern (Fig. 4). Under scenarios S1 and S2, it can be observed that future δ^{18} O is nearly 100% different from reference conditions during the two first months of the year (Fig. 4). It is at least 75% different for the month of March, but this month shows important variation during the future period. Throughout the rest of the year, ranges of variation are not completely different, but increasing or decreasing trends can be observed, depending on the season.

Indeed, Fig. 5 shows the monthly differences between mean lake δ^{18} O in the reference period (which is the same for all scenarios) and mean lake δ^{18} O in the future period, for the four recharge evolution scenarios.

On the year:

regarding the reference period, the highest variation is observed in March for S1 (-1 ‰), S2 (-1 ‰), and NC (-0.4 ‰), after the melt period. For S0, the greatest change regarding the

reference period is observed in September and October (+0.4 ‰), after the evaporation period;

 regarding the NC future period, the greatest difference between winter recharge is in February (-0.6 and -0.5 ‰ for S1 and S2 respectively). This suggests that future changes in lake isotopic composition associated with recharge may be highest in February.

During the summer:

- regarding the reference period, the highest variation will be in August for NC (+0.2 ‰), while it will be in September and October for S0 (+0.4% for both months) and S2 (+0.2 and +0.3 ‰ in September and October respectively). S1 do not show any variation;
- regarding the NC future period, the greatest change will be in October for S0 (+0.3 ‰) and S2 (+0.2 ‰), and in September for S1 (-0.1 ‰).

Results of scenario S2, characterized by the greatest changes in recharge, in both summer and winter, highlights that the impact of decreased recharge during summer attenuates the substantial impact of increased recharge during winter. Indeed, during winter, S1 shows more depleted values than S2 (-0.5 versus -0.4 ‰ in January, and -0.8 and -0.7 ‰ with respect to the reference period for S1 and S2 respectively). Therefore, the more recharge decreases in the summer, the more lake isotopic composition increases in the summer, due to increased future evaporation. Meanwhile, the more recharge increases in the winter, the more lake isotopic composition is depleted in the winter. If both phenomena occur in a given year, the mean annual lake isotopic composition evolution will therefore not be expected to shift much, since their opposing impacts on lake isotopic composition will cancel each other out. As such, S1 is the scenario showing the highest variation in annual mean, of -3 ‰, compared with -2 ‰ for S2 and +2‰ for S0.

Based on these observations, it appears that isotopic signatures measured at the end of February and in September or October will provide information on the greatest changes during the winter and summer periods respectively. The greatest changes in lake isotopic composition are likely to be at the end of the melt period.



Fig. 4. (a) Monthly Lake Lacasse isotopic composition, calculated using RCP 4.5 climatic data, for different periods and various recharge patterns: no change (NC), -33% in the summer (from June to October; S0), +200 % during the melt period (from January to March) and - 4% in the summer (S1), and +200 % during the melt period and -33% in the summer (S2); (b) close-up of the winter months; c) close-up of the summer months; bow-whiskers describe median, first and third quartiles and maximum and minimum values.



Fig. 5. Differences between mean Lake Lacasse δ¹⁸O in the reference period and mean Lake Lacasse δ¹⁸O in the future period, for the RCP 4.5 climate scenario and four scenarios of recharge evolution: no change (NC), -33% in the summer (from June to October; S0), +200 % during the melt period (from January to March) and -4% in the summer (S1), and +200 % during the melt period and -33% in the summer (S2).

Moreover, simulation results show that RCP 4.5 and 8.5 models provide similar results for Lake Lacasse isotopic composition evolution. Figure 6 shows the comparison of lake δ^{18} O composition for both RCP climate scenarios, from 2010 to 2071, assuming the NC recharge scenario. In Fig. 6, it can be observed that there is a small trend toward δ^{18} O enrichment due to a higher evaporation rate, which is more pronounced for the RCP 8.5 than for the RCP 4.5 scenario. However, on a yearly time scale, the impact of evaporation increase in the summer seems to be attenuated by a precipitation increase throughout the rest of the year, likely implying that these climate changes result in a nearly non-measurable impact on lake isotopic composition evolution.

Finally, all these results show that extreme caution is required when interpreting trends in lake isotopic composition, and that their interpretation requires (i) a minimum background knowledge – at least one year of data – of lake isotopic composition evolution in relation to its hydrological balance, and (ii) an accurate evaluation of weather data variability in the year of monitoring, with respect to their annual means for the study lake. A long term change in recharge will definitely impact lake isotopic composition, but the lake is also sensitive to changes in other water budget parameters. It may therefore still be difficult to definitively isolate the effect of recharge over long time periods. As such, it is also important to consider evolution in the yearly mean lake isotopic composition.



Fig. 6. Comparison between monthly results in δ^{18} O for both scenarios RCP 4.5 and 8.5 for the 2010-2071 period.

3.2. Annual evolution of lake geochemistry

3.2.1. Isotopic signature evolution

The model was run for the ten study lakes, including Lake Lacasse (Table 1 for main lake characteristics). Figure 7 illustrates differences in δ^{18} O in the reference period compared to the future period for lakes which have a range of G-indices (see Arnoux et al. 2017a for more details about lakes measured values). It can be observed that, if the recharge is set as constant from 2010 to 2071 (NC recharge scenario), there is no significant difference between the reference and future period (Fig. 7), although evaporation shows a significant increase with time. The lack of a trend is probably mitigated by concurrent shifts in precipitation (Fig. 2). Without considering changes in groundwater inflow, it appears that lake isotopic composition will be at least as much impacted by changes in precipitation as by changes in evaporation.

Fig. 7 illustrates that the range of lake isotopic composition variation depends significantly on climate conditions, lake volumes, and their associated G-indices. It can be observed that lakes with a low G-index and a small volume have higher potential variability in isotopic composition regarding climatic variations than those with a high G-index and high volume. For example, for two lakes with a similar mean G-index, such as Lake Ludovic (SAG; G-index=51%) and Lake Lacroix (OUT; G-

index=53%), the former is expected to have a greater spread in isotopic compositions than the latter, even though the SAG region will likely undergo less evaporation increase compared with the OUT region (Fig. 2). This difference is due to the lower volume of Lake Ludovic (V=400000 m³), compared with Lake Lacroix (V=1080000 m³; Table 1). In addition, when lakes have a high G-index, the groundwater flux tends to buffer lake isotopic variations, and so they tend to be less sensitive to changes in climate data. The dominant control on lake isotopic variability therefore appears to be the G-index. Another example is Lake Lanthier, which has a smaller volume (V=125000 m³) and a higher G-index (G-index=94%), and therefore shows a limited range of isotopic variation compared with Lake Lacroix, although both are located in the OUT region (Fig. 7).

If a changing recharge scenario is applied, a decreasing trend in lake isotopic composition is clearly observed (Fig. 7). However, it is also shown that lakes are sensitive to large changes in annual recharge (+45%), but the differences are not significant if a smaller change (+14%) occurs. Moreover, as the percentage of recharge change applied in the model is the same for all lakes, it can be observed that the trend intensity will depend on four main parameters: lake catchment size (which controls the intensity of the flux change), the region (which underlies climate condition), lake volume (which impacts the range of variation), and the G-index. However, a relationship is only found with the latter.

Figure 8 illustrates variations in mean lake δ^{18} O versus G-index in both reference and future periods. As shown, lake isotopic composition is more sensitive to changes in recharge for G-indices ranging from 50 to 80%, with a maximum of sensitivity observed for a G-index of around 65 %. It can also be observed that RCP 8.5 predicts a more depleted isotopic composition than does RCP 4.5. This implies that for the same recharge scenario, variations in precipitation and melt period (duration and time in the year) may impact the lake isotopic evolution more than precipitation. Finally, the polynomial relationship between the two variables in Fig. 8 highlights that the G-index drives the response of lake isotopic composition to changes in recharge.



Fig. 7. Reference period (Ref; 2010-2040) lake δ¹⁸O composition and that corresponding to three different future period (2041-2071) recharge scenarios: no change (NC), +14% (Low), and +45% (High), for RCP 4.5 (top) and RCP 8.5 (bottom) scenarios. The values in brackets correspond to the mean G-index (percentage of groundwater flow in the total inflow) for each lake calculated for the reference period; left panels show OUT and LAU regions, middle panels AT and rights panels SAG; bow-whiskers describe median, first and third quartiles and maximum and minimum values.



Fig. 8. Differences between mean lake δ¹⁸O in the reference period (2010-2040) and future period (2041-2071), for the higher recharge change scenario, versus lake G-indices. RCP 4.5 (black dots) and 8.5 (grey dots) scenarios are represented.

3.2.2. Lake quality evolution

As it has been shown previously, isotopic composition can be sensitive to future change in recharge. This part is now a discussion about how lake water quality could be impacted by future changes in recharge depending on lake location. In the study regions, one of the principal concerns about lake water quality, today and in future, is to prevent blue-green algae blooms in limiting P loads to lakes. This study does not take into account several parameters that can impact blue-green algae blooms in lakes, such as the lake water biogeochemistry, chemical threshold processes, thermal/oxygen stratification and the warming of the water column. The purpose here is to show in which case, lakes could be under risk of too high P loading, and therefore at risk of a decrease in their quality, depending on their catchment evolution as a function of recharge and population evolution. In such cases it will be important to prevent P loads and therefore changes on the lake catchment.

Turning to the predictions of population growth summarized in Table 3, population is predicted to increase mainly in the southern regions, OUT and LAU, with a mean increase of 24 and 28% by 2036 respectively (ISQ, 2014). Assuming an identical per capita P load, total P load in groundwater originating from waste water should increase by the same percentage.

Domestic sewage is the main contribution of anthropogenic sources to the total P load for most of Canadian lakes (Dillon and Evans, 1993; Paterson et al., 2006). The total P load from sewage systems is a function of (i) the population and (ii) the annual P consumption per capita (Paterson et al., 2006). As done by Paterson et al. (2006), assuming an effluent concentration of 9 mg.L⁻¹ (considering reductions in the phosphate content of detergents) and a daily water usage of 200 L.capita⁻¹.day⁻¹, the P contribution is estimated to be 0.66 kg.capita⁻¹.yr⁻¹. Investigated lakes in the OUT and LAU regions collect sewage from 4 (Lake Lachigan), 53 (Lake Lanthier), 117 (Lake Lacroix), and 17 houses (Lake Lacasse) within their catchments respectively. If two habitants per house are assumed, P loading to groundwater will be increased from 1 to 39 kg.yr⁻¹ in the studied lakes in these areas.

The impact of this P load increase on lakes can then roughly be estimated based on the ratio of change in annual P load versus change in annual recharge, as illustrated in Fig. 9. For an increase in recharge, if Δ_P/Δ_R <1, the change in recharge over the catchment, and thus the evolution of the groundwater inflow to the lakes, will greater than the P variation. In such a case, the lake water quality may not be impacted by this P variation. On the other hand, if Δ_P/Δ_R >1, the lake water quality will be impacted, and precaution should be taken to minimize the risk of blue-green algae blooms and consequent eutrophication. For the study regions (Fig. 9), if recharge increases 14% by 2036, as estimated by Rivard et al., 2014, lakes in the LAU and OUT areas will experience a decrease in their water quality. However, if the recharge change is closer to +45% (Rivard et al., 2014), lake water quality

should not be worse than today, providing all other things remain equal and assuming the population growth forecasts are accurate.



Fig. 9. Population growth predictions versus changes in recharge. The shaded area represents the scenarios for which lakes may be under risk of too high P loading, and therefore at risk of a decrease in water quality. Dots represent lakes in the four study areas for three recharge scenarios.

4. <u>CONCLUSION</u>

The main objectives of this study were to determine how future trends groundwater recharge can affect lake geochemistry, and to assess whether stable isotopes might be an effective tool for identifying lakes that are susceptible to change, or are undergoing changes, in their water budget and quality.

Firstly, climate predictions from both RCPs 4.5 and 8.5 scenarios and their impacts on future lake isotopic composition have been considered. By 2050, temperature and evaporation are expected to increase, and precipitation to exhibit a slightly increasing trend, all trends being more intense under the RCP 8.5 scenario. On a monthly time step, it has been highlighted that future lake isotopic signatures will be more depleted with respect to the reference period, mainly in March and February, because of an earlier melt period. In the summer, lake isotopic composition will be more enriched, mainly in August, due to the higher evaporation rate expected. However, future variations with respect to the reference period are smaller in the summer than in the winter. Scenario RCP 8.5 induces more intense monthly variations, but no significant difference in future lake isotopic signatures is observed on a yearly time step between the two scenarios. This means that enrichment caused by increased evaporation compensates for depletion induced by precipitation variation. It is therefore unclear whether lakes will be

impacted more by increased evaporation or precipitation changes. Caution is therefore recommended in the interpretation of isotopic trends in lakes where background knowledge – for at least one year – of their isotopic composition evolution with respect to weather data and their hydrologic balance is lacking.

It has then been demonstrated that future lake isotopic composition will also depend on recharge fluctuations, in addition to climate conditions. On a monthly basis, the highest impact of recharge evolution on future lake isotopic composition will be in February. Moreover, if recharge decreases during the summer, the main difference will be observed at the end of the summer, after the evaporation period and before recharge stops decreasing, in September or October. Therefore, to clearly identify future changes in recharge through the lake isotopic signature evolution, sampling only at the end of February and in September or October will provide information on the greatest changes for the winter and summer periods respectively.

On an annual time step, modelled evolutions of lake isotopic composition can clearly be sensitive to both +45% and +14% changes in recharge, less so, nevertheless, to the latter. The intensity of the future trend of lake isotopic composition will depend on four main parameters: lake catchment (which controls the intensity of the flux change), the region (which drives climate conditions), lake volume (which impacts the range of variation), and the G-index (which is the dominant control on water balance conditions). Based on these model simulations, stable isotopes appear to be especially useful for detecting changes in recharge to lakes with a G-index of between 50% and 80%.

It is important to keep in mind that if both a winter increase and summer decrease in recharge occur during the same year, the trend in mean annual lake isotopic composition will be nullified, because seasonal variation is impacted it in opposing directions, cancelling out the signal at the yearly time step. Consequently, if no clear annual trend is observed, it does not mean that recharge is not changing. Nevertheless, mean annual lake isotopic compositions will be observed to be impacted by recharge evolution only if it evolves in the same way throughout the year for the most part (i.e., consistently decreasing or increasing). In light of these results, it is a monthly time step is strongly suggested in such investigations, since seasonal recharge fluctuations can be cancelled out in the yearly signal. Moreover, it is important to note that runoff has been considered negligible for our studied lakes but can be important for other lakes and, in these cases, this model could underestimated the effect of spring melt on future lake isotopic composition.

It is also shown that changes in water quality in groundwater-connected lakes depend substantially on lake location and on the intensity of recharge change. For the studied lakes, in the case of a +14% recharge increase by 2036, lakes in LAU and OUT regions may experience altered water quality (driven by phosphorous loading), but no change is expected in the case of a +45% recharge intensification. If the percentage of recharge increase is at least equal to the percentage of population

growth around the lake, lake quality should not become degraded, but if not, recharge evolution should be considered in lake management. Lakes water quality in the SAG and AT areas may not decrease when considering population growth predictions. However, this study does not take into account several parameters that can impact blue-green algae blooms in lakes, such as the lake water residence time, chemical threshold processes, and the warming of the water column (Planas and Paquet, 2016).

Finally, even if small groundwater-fed lakes will be sensitive to climate, and especially to recharge and anthropogenic changes, it is still difficult to predict how their geochemistry will be impacted, as it is very reactive to each slight variation in water balance parameters. However, more indicators are now available to predict lake geochemistry evolution, mainly depending on their location and their G-index. To go further, a recharge model adapted to lake catchments and coupled with melt dynamics, closely dependent on climate forecasts, could provide more details on lake geochemical evolution, for more sustainable lake management.

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Table 1. Main lake characteristics

Region	ID	Lake name	Lake surface area 10 ³ m²	Lake volume 10 ³ m ³	Catchment Area 10 ³ m ²
AT	1	Clair	115	695	2646
AT	2	Paix	41	97	796
AT	3	Sauvage	44	142	89
OUT	4	Lachigan	33	142	336
OUT	5	Lacroix	236	1080	772
OUT	6	Lanthier	25	125	1134
LAU	7	Lacasse	27	67	148
SAG	8	Beau Portage	42	271	364
SAG	9	Girard	67	679	211
SAG	10	Ludovic	94	400	1829

Table 2. Mean isotopic composition of groundwater obtained for the four regions; in ‰ vs VSMOW.

Region	δ ¹⁸ Ο	δ²H
AT	-14.00	-101.3
OUT	-11.56	-81.6
LAU	-11.71	-80.9
SAG	-14.06	-103.1

Table	3. Pred	dicted p	population	growth in	the differ	ent study	regions i	in 2036	relative to	o 2011	numbers,	according to	o three	different	scenarios
(ISQ,	2014)														

Region	Scenarios							
	Reference (%)	Low (%)	High (%)					
OUT	24	13	36					
AT	5	0	10					
LAU	28	21	34					
SAG	0	-4	4					

Supplementary material:

Table 1. N	Mean I	ake i	isotopic	composition	measured	in	June-July	and	October-November	[·] 2014; ii	n ‰ vs
VSMOW.											

Region	ID	Lake name	δ ¹⁸ O in June-July 2014	δ²H in June-July 2014	δ ¹⁸ O in October-November 2014	δ ² H in in October-November 2014
AT	1	Clair	-8,41	-73,43	-8,23	-72,95
AT	2	Paix	-7,66	-72,19	-7,34	-70,07
AT	3	Sauvage	-9,11	-79,32	-8,55	-76,31
OUT	4	Lachigan	-6,67	-60,05	-6,28	-57,84
OUT	5	Lacroix	-6,69	-60,26	-6,77	-60,39
OUT	6	Lanthier	-10,95	-77,64	-11,09	-78,24
LAU	7	Lacasse	-10,73	-76,57	-8,57	-66,23
SAG	8	Beau Portage	-7,85	-72,82	-7,53	-70,42
SAG	9	Girard	-12,82	-97,77	-12,54	-96,39
SAG	10	Ludovic	-10,00	-80,24	-9,06	-76,25

Figure 1. Mean volume-weighted isotopic composition of lake Lacasse and isotopic composition of its outlet, measured in 2015-2016; in ‰ vs VSMOW.

