GROUNDWATER ORIGIN AND GEOCHEMICAL PROCESSES IN THE BASSES-LAURENTIDES SEDIMENTARY ROCK AQUIFER SYSTEM, ST. LAWRENCE LOWLANDS, QUÉBEC, CANADA

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ABSTRACT
A comprehensive hydrogeochemical study was carried out in the Paleozoic Basses-Laurentides sedimentary rock aquifer system in Québec. The hydrogeochemical data were analyzed by combining conventional geochemical and statistical methods, such as classification by water types and multivariate statistical analysis. The integration of hydrogeochemistry, including the origin, evolution and geochemical processes, within the hydrogeological and geological contexts allowed the division of the region into four main geochemical areas, providing a global picture of the aquifer system dynamic. With its integrated approach, this hydrogeochemical study brings an example of the long-term evolution of hydrogeological systems, as well as a contribution to the characterization and understanding of complex groundwater flow systems. This understanding is essential for the sound management and future development of groundwater resources in the region.

RÉSUMÉ
Une étude hydrogéochimique détaillée a été réalisée sur le système aquifère des Basses-Laurentides, dans les roches sédimentaires paléozoïques au Québec. Les données hydrogéochimiques ont été analysées en utilisant une combinaison de méthodes géochimiques classiques et statistiques, dont la classification en types d'eau et l'analyse statistique multivariable. L'intégration de l'hydrogéochimie, incluant l'origine, l'évolution et les processus, avec les contextes hydrogéologiques et géologiques a permis de diviser le territoire en quatre régions géochimiques, procurant un portrait global de la dynamique du système aquifère. Avec son approche intégrée, cette étude hydrogéochimique apporte un exemple d'évolution à long terme de systèmes hydrogéologiques, ainsi qu'une contribution à la caractérisation et la compréhension de systèmes d'écoulement complexes. Cette compréhension est essentielle pour la gestion éclairée et le développement futur de la ressource en eau souterraine dans la région.

1. INTRODUCTION

The groundwater hydrogeochemistry of the Basses-Laurentides aquifer system was studied extensively as part of a regional hydrogeological characterization project (Savard *et al*. in press). The study area is located in the St. Lawrence Lowlands, northwest of Montréal, and covers approximately 1500 km^2. The regional aquifer system consists of nearly horizontal Cambrian-Ordovician fractured sedimentary formations lying in unconformity on the crystalline basement of the Precambrian Grenville Province (Figure 1a). The geological units are mainly sandstone (Covey Hill and Cairnside formations), dolostone (Theresa, Beauharnois and Carillon formations), and limestone (Chazy, Black River and Trenton groups). In the area, the sedimentary formations rarely outcrop, as Quaternary sediments cover them. Unconfined conditions, characterizing areas of groundwater recharge, are located in areas of elevated topography, sometimes with rock outcropping or with thin to thick permeable surface sediments, generally till (Figure 1b). The aquifer system is confined in areas where thick low permeability Champlain Sea clays are present, such as in buried valleys. Regionally, the main groundwater flow paths are from North to South (Figure 1c). Figure 1d illustrates the conceptual groundwater flow model, showing the transition from unconfined to confined conditions, as well as the main hydrostratigraphic units adapted from Savard *et al*. (in press).

The objectives of the hydrogeochemical study were to determine the influence of hydrogeological and geological contexts on groundwater geochemistry, identify groundwater origin, and recognize the main processes controlling the spatial and temporal evolution of groundwater geochemistry. The global understanding of the aquifer system is required for the sound management and future development of groundwater resources.

Groundwater samples were collected at 153 sites, characterizing all geological and hydrogeological units to a maximum depth of 140 m. Groundwater was analyzed for major, minor and trace inorganic constituents, stable isotopes δ^2H, δ^{18}O and δ^{13}C of dissolved inorganic carbon (DIC), and some samples were analyzed for ^{87}Sr/^{86}Sr, ^{3}H and ^{14}C of DIC. A total of 146 groundwater samples were kept for the geochemical interpretation. The complete hydrogeochemical characterization program, including the sampling and analytical protocols, are described in Cloutier (2004) and Cloutier *et al*. (in prep a).

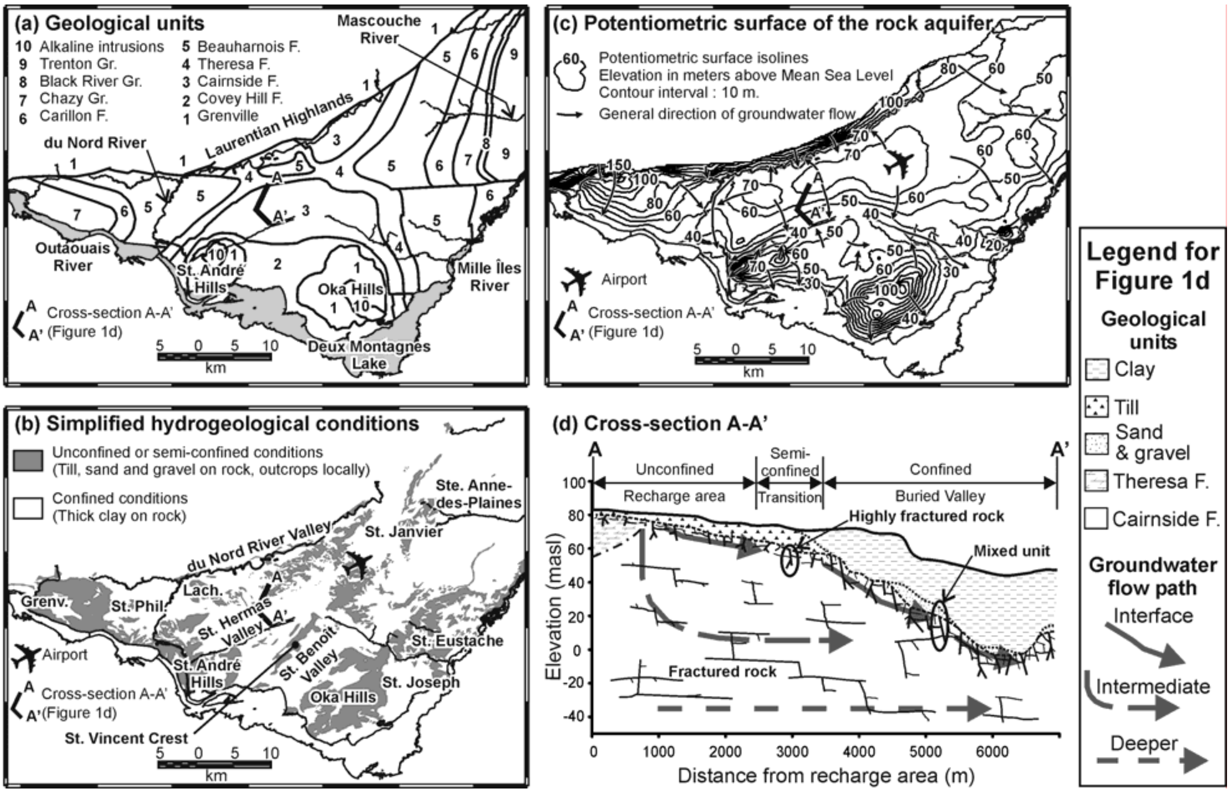

Figure 1. Context of the study area: (a) geological map (modified from Rocher *et al*. in press), (b) hydrogeological conditions map (modified from Hamel *et al*. 2001), (c) potentiometric surface map (modified from Paradis in press), and (d) groundwater flow conceptual model (modified from Cloutier *et al*. in prep a)

2. RESULTS AND INTERPRETATIONS

2.1 Classification and Zoning of Samples

The Basses-Laurentides sedimentary rock aquifer system has a highly variable groundwater geochemistry. An approach of dominant and mixed groundwater types, based on major ions concentrations, was used to classify the samples into six dominant and four mixed groundwater types (Cloutier *et al*. in prep a). The ten groundwater types were associated into four main groundwater groups, G1 to G4 (Table 1).

The distribution map of the groundwater groups for the 146 samples shows their relationships with the hydrogeological conditions (Figure 2). To help visualize the geographical distribution of the groundwater groups, the groundwater relative quality zones defined by Cloutier *et al*. (in press) are presented on Figure 2. The samples located in, or near, the unconfined or semi-confined areas are characterized by groundwater belonging to G1 and G4, particularly by the dominant groundwater types Ca-HCO_3 (water quality zones C and E). Groups G2 and G3 characterize the aquifer under confined conditions, dominated by the groundwater types Na-HCO_3 (water quality zones A, B and F) and Na-Cl (water quality zone G), respectively. Groundwater from G3 (Na-Cl) is also found locally in water quality zones A, B and F. The distribution of groundwater types in the study area, as well as the descriptive statistics show that the hydrogeological conditions exert an important control on the geochemistry of the groundwater (Cloutier *et al*. in prep a).

Although they are located in, or close to, unconfined areas, some samples collected along the main highways 15 and 117, particularly around St. Janvier, have G3 groundwater (Figure 2). Cloutier *et al*. (in prep b) demonstrated that these samples were contaminated by de-icing road salts, which gives their Na-Cl characteristic.

Table 1. Groundwater types and groups determined by Cloutier *et al*. (in prep a).

Groundwater group (N[1])	Dominant water type (N[1])	Mixed water type (N[1])
G1 (68)	Ca-HCO_3 (59) Mg-HCO_3 (4)	Alkaline earth-HCO_3 (5)
G2 (47)	Na-HCO_3 (35)	Mixed cations-HCO_3 (12)
G3 (28)	Na-Cl (25)	Mixed cations-Cl (2) Na-Mixed anions (1)
G4 (3)	Ca-SO_4 (2) Na-SO_4 (1)	

[1] number of samples

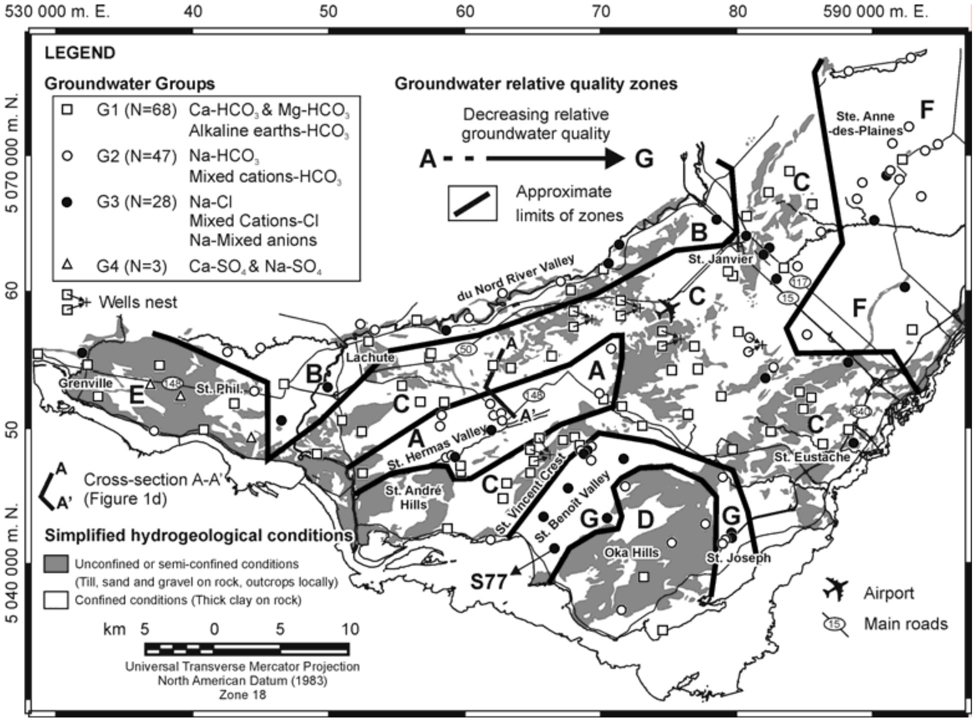

Figure 2. Regional distribution map of groundwater groups (hydrogeological conditions map modified from Hamel *et al*. 2001; groundwater relative quality zones from Cloutier and Bourque in press)

2.2 Groundwater Origin

Qualitative interpretation of ^3H data shows that preferential recharge areas, characteristic of G1 (Ca-Mg-HCO_3) and G4 (Ca-SO_4, Na-SO_4), have modern, tritiated groundwaters, and that the confined aquifer areas, characteristic of G2 (Na-HCO_3) and G3 (Na-Cl), have submodern groundwaters. Groundwater G2 results from the evolution of G1, and groundwater G3 from the mixing of G2 with Pleistocene Champlain Sea water, as well as diffusion of solutes from marine clay aquitard (Figure 3).

Samples obtained in the till belong to groundwater group G1, and were interpreted by Cloutier *et al*. (in prep a) to represent the major ions geochemistry of the recharge water and thus, it is a geochemical groundwater end-member of the aquifer system. Sample S77, located in the buried valley of St. Benoît (see Figure 2), has the highest TDS of the wells sampled in the course of this project (calculated TDS = 11 337 mg/L), and a Na-Cl geochemistry very similar to the marine clay pore-water extracted in the middle of clay accumulations. Both of these were interpreted to represent Champlain Sea water, a second geochemical end-member to the aquifer system (Cloutier *et al*. in prep a).

The relation between groundwater groups and end-members is particularly noticeable in St. Hermas Valley, where modern meteoric G1 water in the recharge area evolves to G2 water (Step 3), to finally G3 water (Step 4) in the center of the valley (Figures 2 and 3).

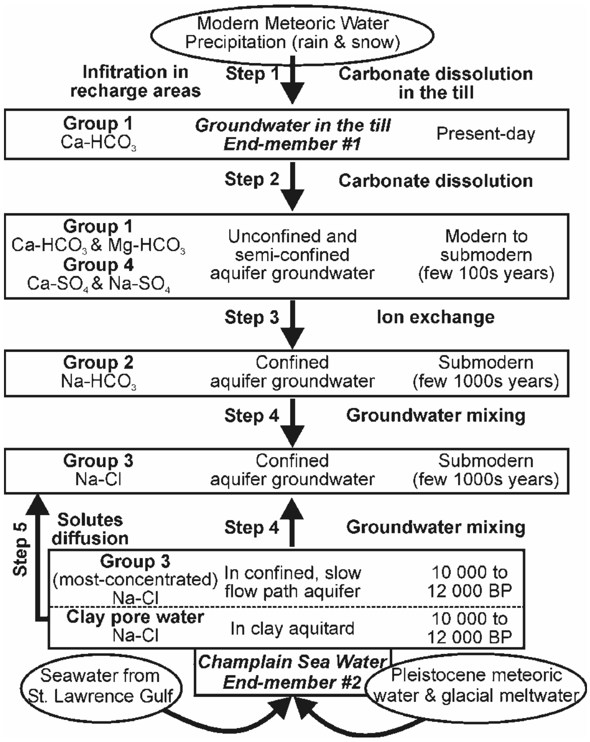

Figure 3. Conceptual model of groundwater origin and relationships between groundwater groups and end-members (modified from Cloutier *et al*. in prep a)

2.3 Geochemical Processes and Evolution

Numerous geochemical processes contribute to the highly variable groundwater geochemistry of the aquifer system (Table 2; Figure 4). Cloutier *et al*. (in prep b) identified that dissolution of carbonates, calcite and dolomite, dominates in the preferential recharge areas resulting in Ca-Mg-HCO_3 (G1) groundwater. In the aquifer system under confined conditions, Ca^{2+}-Na^+ ion exchange is the cause of groundwater evolution to the Na-HCO_3 type (G2). Groundwater mixing with Pleistocene Champlain Sea water and diffusion of solutes from the marine clay aquitard are the causes of the occurrence of Na-Cl groundwater (G3) also under confined conditions.

The marine invasion that resulted in the Champlain Sea is the cause of the salinization of the aquifer system (Cloutier *et al*. in prep b). The conservative tracers Cl^- and Br^-, as well as $\delta^{18}O$, indicate that the original Champlain Sea water, in the study area, was a mixture of about 34% seawater and 66% freshwater (Pleistocene meteoric water and glacial meltwater). The current groundwater geochemistry indicates that the aquifer system is at different stages of desalinization, ranging from the original Champlain Sea water still present in hydraulically stagnant areas of the aquifer to fully flushed conditions in the more active parts of the aquifer system, especially in recharge zones. A modern salinity source, de-icing road salt, was identified in a recharge area, leading to the occurrence of Na-Cl (G3) groundwater. This interpretation of groundwater groups within the hydrogeological contexts defined the main geochemical processes, thus providing the meaning to these groups in terms of groundwater evolution.

Table 2. Main geochemical processes, water types and synthesis of the pieces of evidence (Cloutier *et al*. in prep a; in prep b).

1. Carbonate dissolution
Resulting WT[1] : $G1_{Till}$ (Ca-HCO_3) & G1 (Ca-Mg-HCO_3)
- $[Ca^{2+}]$ and $[HCO_3^-]$ increase linearly
- Mg/Ca ratios between 0.6 and 0.9
- pH increases relative to precipitation
- elevated pCO_2 (between $10^{-1.5}$ to $10^{-2.5}$ atm)
- undersaturated to near equilibrium calcite & dolomite SI[2]
- GW[3] $\delta^{13}C_{DIC}$ in the till unit = -15.0‰
- availability of calcite and dolomite in the till matrix
- GW[3] $^{87}Sr/^{86}Sr$ (0.7082-0.7091) in dolostone & limestone
- elevated $[Ca^{2+}]$ and $[Mg^{2+}]$ in dolostone
2. Ion exchange
Resulting WT[1] : G2 (Na-HCO_3)
- negative $\log(Ca+Mg)/Na^2$
- increasing trend between pH and $[Na^+]$
- increasing $[Na^+]$ coupled to a decrease in $[Ca^{2+}]$
- increase in $[Na^+]$ not really coupled to a $[Cl^-]$ increase
3. Groundwater mixing
Resulting WT[1] : G3 (Na-Cl)
- $[Na^+]$ and $[Cl^-]$ increase linearly, and towards seawater
- $[Cl^-]$ and TDS increase linearly, and towards seawater
- geological history: Champlain Sea invasion
- geochemical similarities between S77 & clay pore water
- sample S77 plots on the seawater Cl/Br line
- mixing model based on tracers Cl^-, Br^- & $\delta^{18}O$
- downward diffusion of Cl^- and Na^+ from the aquitard

[1] water type [2] saturation index [3] groundwater

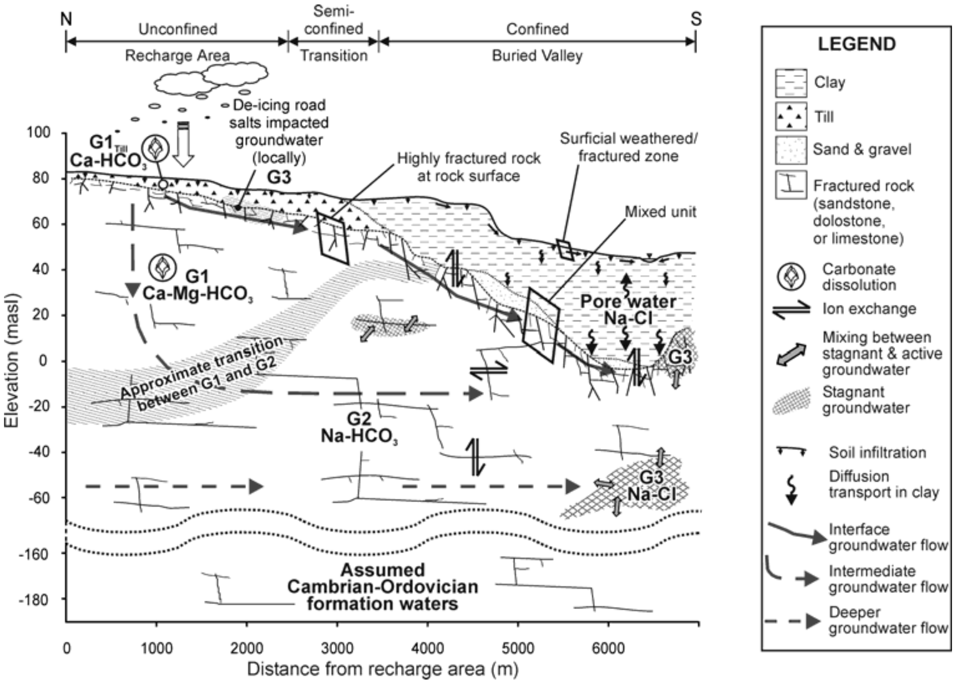

Figure 4. Groundwater flow conceptual model with the relative position of the groundwater groups within the Basses-Laurentides sedimentary rock aquifer system (modified from Cloutier *et al*. in prep b)

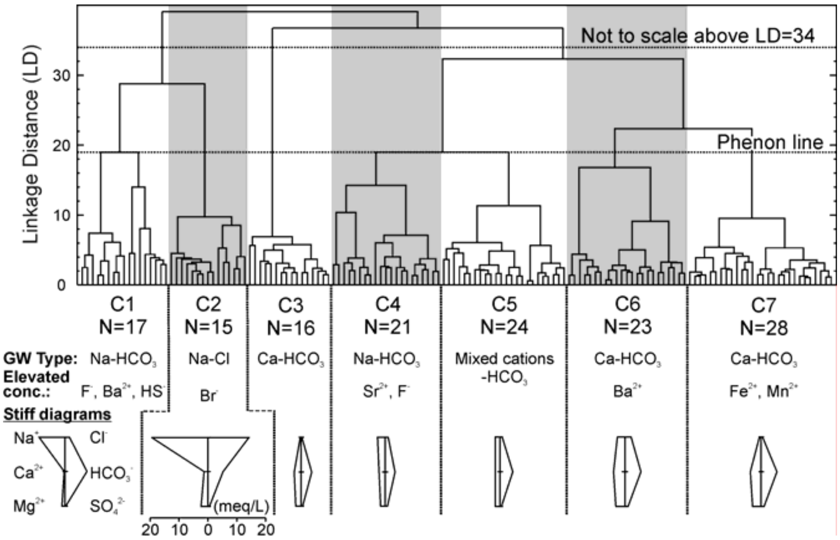

Figure 5. Dendrogram for the 144 samples, showing the division into 7 clusters and the median Stiff diagram of each cluster (N: Number of samples, GW: Groundwater, conc.: concentrations) (modified from Cloutier *et al.* in prep c)

2.4 Multivariate Statistical Analysis

To further refine the geochemical interpretation, multivariate statistical methods, hierarchical cluster analysis (HCA) and principal components analysis (PCA), were applied to a subgroup of the dataset that consisted of 144 samples and 14 parameters (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, HCO_3^{-}, Cl^{-}, SO_4^{2-}, Fe^{2+}, Mn^{2+}, Br^{-}, Sr^{2+}, F^{-}, Ba^{2+}, HS^{-}) (Cloutier *et al.* in prep c). The HCA was performed using the Euclidean distance between the sampling sites and Ward’s method as linkage rule. The resulting dendrogram classified the groundwater samples into seven geochemically distinct clusters, C1 to C7 (Figure 5). Three clusters, C1, C2, and C5, characterize samples from the aquifer system under confined conditions. Samples that belong to the other four clusters, C3, C4, C6, and C7, are located mostly in preferential recharge areas (Cloutier *et al.* in prep c). In addition to recognizing the importance of hydrogeological conditions on groundwater geochemistry, the distribution of clusters also identified the importance of the geological formations on minor and trace elements, such as Fe^{2+}, Mn^{2+}, Sr^{2+}, F^{-} et Ba^{2+} (Figure 5).

The PCA was performed by extracting the principal components on the correlation matrix. The Varimax normalized rotation was applied after keeping the components with eigenvalues greater than 1. The first five components of the PCA account for 78.3% of the total variance in the dataset. Figure 6 shows the position of the loadings of chemical parameters in the plane defined by the axes of components 1 and 2. Component 1 explains the greatest amount of the variance, and is characterized by highly positive loadings in Na^{+}, Cl^{-}, and Br^{-}, and thus, is defined as the "salinity" component. Component 2 is characterized by highly positive loadings in Ca^{2+} and Mg^{2+}, and thus, is defined as the "hardness" component. Loading is also high for SO_4^{2-}. The first two components account for 40.6% of the variance in the hydrogeochemistry, indicating the regional importance of salinity and hardness in this aquifer system. Components 3 to 5 are related to more local and geological effects.

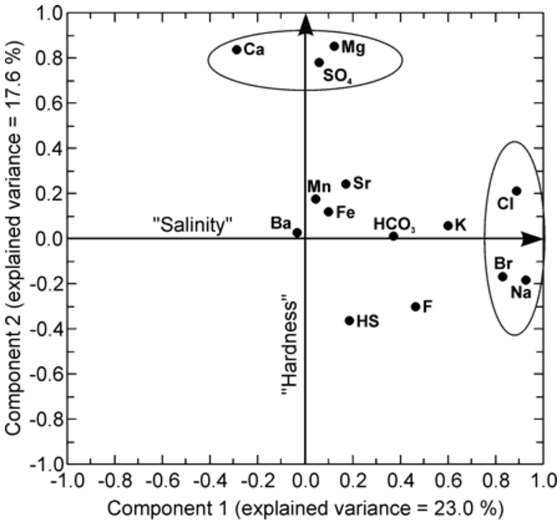

Figure 6. Loadings for the first two components (from Cloutier *et al.* in prep c)

The Piper diagram shows the relationships between the seven groundwater clusters and the four groundwater groups (Figure 7). The majority of the samples from C3, C6, and C7 belong to G1 ($Ca-Mg-HCO_3$), the majority of the samples from C1 belong to G2 ($Na-HCO_3$), and the majority of the samples from C2 belong to G3 (Na-Cl). Samples from C4 and C5 have a majority of samples from G2, with samples belonging to G1 as well.

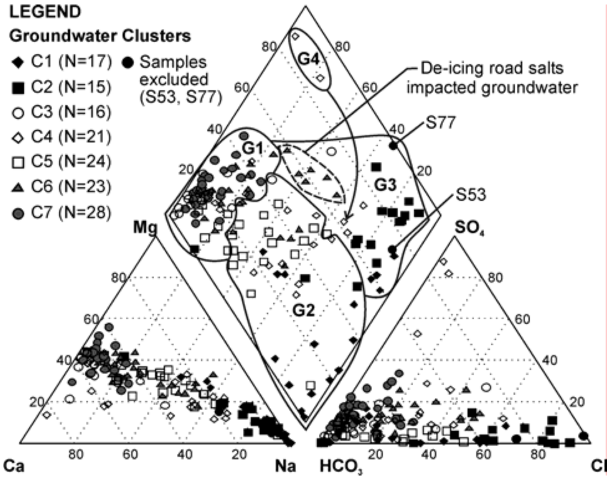

Figure 7. Piper diagram of the groundwater clusters with the groundwater group envelopes (modified from Cloutier *et al*. in prep c)

2.5 Global Understanding of the Aquifer System

Figure 8 presents the principal component scores for the first two components for all groundwater samples. The scores represent the influence of the component on the groundwater samples. Grouping of samples is possible into this plane defined by the axes of components 1, "salinity", and component 2, "hardness". Samples from unconfined and semi-confined areas, as well as from surface sediments and springs, dominate the upper-left quadrant of the diagram. They are characterized by elevated hardness and lower salinity. On the other hand, samples from confined areas dominate the lower-right quadrant of the diagram, and are characterized by elevated salinity and lower hardness. The scores of the samples indicate that both first components, the "salinity" and the "hardness", are to some extent dependant, or related to, the hydrogeological context.

A Confinement Index (CI), based on the distribution of the scores of components 1 and 2, is defined to integrate both components into a unique parameter. CI ranges from unconfined conditions, at CI=0, to confined conditions, at CI=10. The index gives an "apparent" level of confinement for each sampling site. Confinement was chosen to relate components 1 and 2, as it is a parameter based on field knowledge of each sampling site. The "apparent" unconfined conditions dominate at a CI lower than 2.5, and the "apparent" confined conditions dominate at a CI above 5.5. From a CI of 2.5 to 5.5, the sampling sites are defined as "apparent" semi-confined.

Cluster ellipses, drawn for each groundwater cluster at a coefficient of 0.5, are reported on Figure 8 (Cloutier *et al*. in prep c). The CI can thus be calculated for each cluster ellipse. Figure 8 is efficient at separating the various clusters in the plane defined by the axes of components 1 and 2, associated to "salinity" and "hardness" respectively. This diagram also provides a good visualization of the various groundwater types found in the Basses-Laurentides, as well as insight into the geochemical processes responsible of these variations.

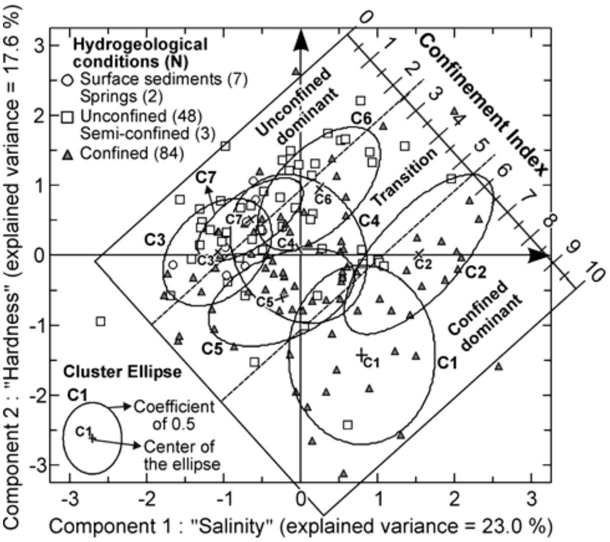

Figure 8. Principal component scores for the first two components with the definition of the Confinement Index and the groundwater cluster ellipses (modified from Cloutier *et al*. in prep c)

The integration of all classification methods within the geological and hydrogeological contexts allowed the division of the region into four main geochemical areas, and these were used to refine the interpretation of the hydrogeochemical evolution of groundwater of the Basses-Laurentides aquifer system (Figure 9; Table 3). Every geochemical area can be distinguished by the geological formations, hydrogeological contexts, hydraulic gradients, groundwater groups and clusters, CI, as well as main geochemical processes. Thus the following factors were recognized as influencing the evolution of groundwater identified in all geochemical areas: 1) geological characteristics including sedimentary rock type and till mineralogy; 2) hydrogeological characteristics represented by the level of confinement and the hydraulic gradient; and 3) geological history by the latest glaciation as well as Champlain Sea invasion. The main geochemical processes identified for this aquifer system are all apparent in the Central Crests and Valleys area. This hydrogeochemical evolution includes carbonate dissolution in recharge areas, to ion exchange in the confined areas, as well as groundwater mixing with Champlain Sea water, and solute diffusion from the marine clay aquitard in the deepest portions of the aquifer system. When interpreted within the hydrogeological and geological contexts, the division into main geochemical areas contributes significantly to the global understanding of the hydrogeochemical evolution of the Basses-Laurentides aquifer system while recognizing the significant impact of geological formations.

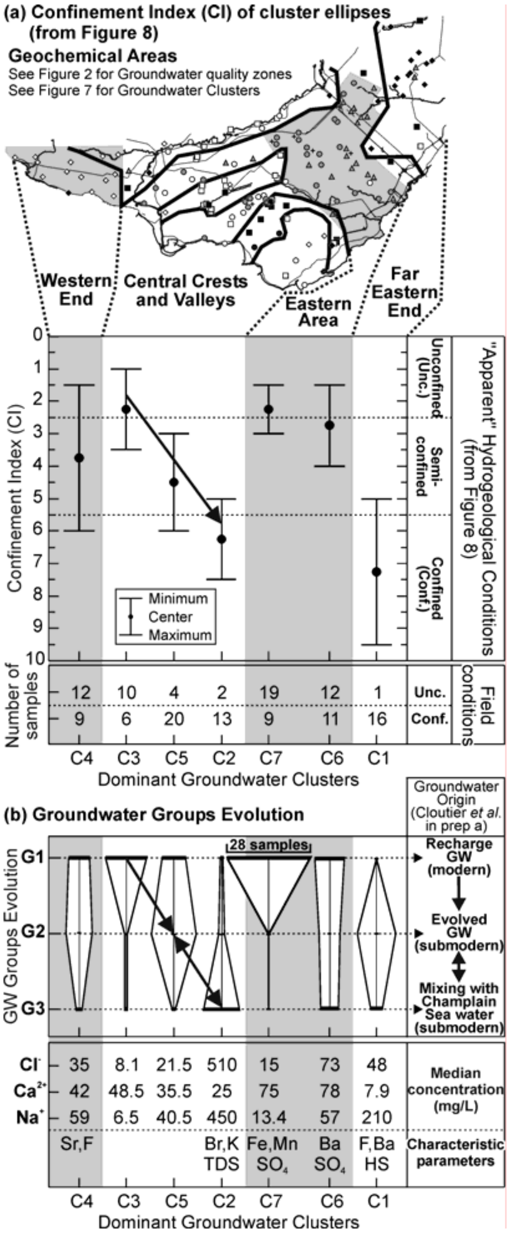

Figure 9. Groundwater evolution within the geochemical areas: (a) Range plot of CI for the cluster ellipses, and (b) relationship between clusters and groundwater groups evolution (modified from Cloutier *et al*. in prep c)

3. GROUNDWATER RESOURCE MANAGEMENT

Regionally, the water quality of the aquifer system is variable, but not too affected by human activities (Cloutier *et al*. in press). The low number of samples exceeding the water quality guideline (Health Canada 2003) for maximum acceptable concentrations (MAC) indicates a good groundwater quality in the area. Only one exceeding related to anthropogenic effects was identified for NO_3^- in a recharge area. The other parameters exceeding the MAC are Ba^{2+} and F^-, and have natural origin. Numerous concentrations exceeding for aesthetic objectives (AO) were observed, but these are also related to natural processes. Only the few Na^+ and Cl^- concentrations exceeding the AO in a recharge area along a main highway can be related to the use of de-icing salts.

The integration of the groundwater relative quality zones in a quantitative study of the groundwater resource identified three favourable zones for groundwater exploitation (Nastev *et al*. in press). These zones coincide with areas of best groundwater potential obtained from the integration between hydrogeochemical and hydraulic properties (Savard *et al*. 2002). The presence of modern groundwater in the recharge areas, identified by the distribution of Ca-Mg-HCO_3 water G1, tritium as well as the clusters, indicate that an important area of the aquifer system is vulnerable to contamination. The high NO_3^- concentration at a site in a recharge area, and the detection of de-icing road salts, confirmed this vulnerability. Even though anthropogenic effects are limited so far, it is essential to protect the recharge areas to prevent a potential degradation of groundwater quality.

4. CONCLUSION

This paper presented the interpretation of results obtained from a regional and comprehensive hydrogeochemical study, allowing the assessment of groundwater quality, the determination of groundwater origin and geochemical processes, as well as the development of an integrated model of the geochemical evolution of groundwater within the Basses-Laurentides aquifer system. By showing that this system is still influenced by a major Quaternary geological event, this study presents an example of the long-term evolution of hydrogeological systems, and shows that groundwater is an important geological agent. Tóth (1999) demonstrated the role of groundwater as geological agent in a regional, gravity-driven flow system. The present study identifies several processes discussed by Tóth (1999). These processes contribute to the evolution of groundwater, from the recharge areas and along the flow system. A distinctive feature of this aquifer system is that it was affected by a major geological event, rather than following a steady and continuous evolution. The Champlain Sea water invaded the aquifer system, thus imposing new hydrogeological and hydrogeochemical conditions to the system about 10 000 years ago. These conditions are still present in some hydraulically stagnant parts of the system in buried valleys, and in marine clay pore water. These saline waters will continue to influence the geochemistry of the underlying aquifer for a long time.

This hydrogeochemical study brings independent and new informations on the aquifer system dynamics. The resulting geochemical database and interpretation can be used to follow the evolution of the water quality due to human activities, identify areas vulnerable to contamination (Ross *et al*. 2003), elaborate management strategies for the resource, and evaluate the impact of future development on the groundwater resource.

Table 3. Main geochemical areas, geological and hydrogeological contexts related to identified groundwater clusters (modified from Cloutier *et al.* in prep c).

Geochemical Area (see Fig. 9)	Western End	Central Crests & Valleys	Eastern Area	Far Eastern End
Main groundwater clusters	C4	C3, C5, C2	C7, C6	C1
Geological context	- Dolostone - Limestone	- Sandstone - Dolostone	- Dolostone	- Dolostone - Limestone
Hydrogeological conditions	Mainly unconfined	Unconfined to confined	Mainly unconfined	Confined
Relative hydraulic gradient	Strong	Strong to moderate	Small	Small
GW Evolution : GW Group				
Recharge GW : G1	C4 (unconfined)	C3 (unconfined)	C7/C6 (unconfined)	
Evolved GW : G2	C4 (confined)	C5 (confined)	C6 (confined)	C1 (confined)
Ancient Champlain Sea water : G3		C2 (confined)	C6 (contaminated)	
Characteristic parameters	Sr, F	Br, K, TDS (C2)	SO_4 (C7/C6) Fe, Mn (C7); Ba (C6)	F, Ba, HS

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