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POLYTECHNIQUE MONTRÉAL

affiliée à l'Université de Montréal

| Multi-scale investigation of bank f | iltration: Investigating | g the province wide |
|-------------------------------------|--------------------------|---------------------|
| occurrence of bank filtration a | and methods to charac | terize recharge |

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Mémoire présenté en vue de l'obtention du diplôme de *Maîtrise en sciences appliquées*Génie minéral

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Ce mémoire intitulé:

Multi-scale investigation of bank filtration: Investigating the province wide occurrence of bank filtration and methods to characterize recharge

présenté par Marc PATENAUDE

en vue de l'obtention du diplôme de *Maîtrise en science appliquées* a été dûment accepté par le jury d'examen constitué de :

Félix GERVAIS, président

Paul BAUDRON, membre et directeur de recherche

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DEDICATION

I would like to dedicate this memoire to all those who inspired me to return to school including friends and family. I was inspired to focus on studying water through a podcast I heard several years ago. In this podcast, a man named Justin Wren described his work providing water to a forgotten group of people in the Congo who had been mistreated and who were seen as less than human. Through development of their water resources, he has brought hope to those communities and enabled them to develop beyond what they would have imagined before his time there. This demonstrates the power that clean water can have on changing the lives of communities throughout the world. Though this work did not have the same end goals as the work of Justin, I hope to use the knowledge I have acquired to make a more positive impact on the world than what I would have made staying in the mining industry.

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I would like to thank the members of the HydroPoly lab who have assisted me with field work over the last two years and with whom I have had many discussions on a variety of subjects relating to my research. They have been incredibly helpful and put up with my frequent questions. I would like to thank Manon Leduc and Jérôme Leroy, and Gabriel St.-Jean for help with laboratory work and sample analysis. Providing the short notice and impromptu support for our apparatuses, sample submissions and field preparation has made things run smoothly throughout this project. Finally, I would like to thank Paul Baudron, for considering me for a master's position when I had struggled to find a school that would accept me. You saw potential in me even though my academic history may not have been excellent. I would also like to thank Paul for providing the resources necessary to complete this project.

RÉSUMÉ

Les investigations des sites de filtration sur berge (FSB) au Québec se font depuis quelques années seulement, et le nombre de sites au Québec qui utilisent la filtration sur berge n'est pas connu. Dans les systèmes de filtration sur berge, les eaux de surface passent à travers les berges d'un lac ou d'une rivière vers un ou plusieurs puits de pompage grâce au gradient hydraulique naturel ou induit par pompage. Leur qualité est ainsi généralement améliorée comparé aux eaux de surface. Grâce à une abondance d'eau de surface dans la province, il est probable qu'un grand nombre de puits font de la filtration sur berge, c'est-à-dire qu'ils pompent un mélange d'eau souterraine et de surface. La réglementation au Québec encadre bien le développement et le suivi des sites d'eau souterraine et d'eau de surface, toutefois, ces sites hybrides ne sont pas spécifiquement discutés. D'autre part, certains paramètres clés dans la capacité d'atténuation des contaminants tels que le taux de mélange de l'eau infiltrée avec l'eau souterraine régionale et le temps de résidence de l'eau de surface au sein des berges sont difficiles à quantifier avec les méthodes actuelles du génie. Il est donc important i) de mieux caractériser l'occurrence de la filtration sur berge dans la province pour permettre d'évaluer la nécessité du développement de règlements propres à ces sites et ii) de développer des approches novatrices permettant de caractériser les écoulements aux sites de FSB.

La première partie de cette étude vise à proposer une première évaluation de la distribution spatiale des sites de FSB et de la population alimentée par ces sites. Pour ce faire, une structure SIG a été utilisée afin notamment de calculer la distance entre ceux-ci et les plans d'eau en proximité. Un regard plus rapproché sur certains bassins versant a ensuite permis d'améliorer la compréhension des interprétations en prenant en compte le type d'aquifère et certains autres facteurs régionaux. Une des trois régions investiguées a démontré un potentiel élevé de FSB (Laurentides), une deuxième nécessiterait des informations supplémentaires pour améliorer le niveau de confiance dans les interprétations (Nicolet), et dans une troisième région (Vaudreuil-Soulanges) il a été démontré que l'utilisation de la FSB est peu-probable. À partir de cette approche, il a été démontré que près de 1 million de québécois sont alimentés en eau potable à partir de puits municipaux situés à moins de 500 m d'un plan d'eau de surface, et que plus d'un demi-million d'habitants ont une probabilité élevée d'être alimentés par des puits de FSB.

La deuxième partie de cette étude s'est concentrée sur l'utilisation du ²²²Rn comme chronomètre naturel in-situ des temps de résidence de l'eau de surface infiltrée au travers des berges. Plus

précisément, l'objectif était d'encadrer les conditions nécessaires à l'obtention de résultats les plus représentatifs possibles. Tandis que les études existantes s'étaient centrées sur une seule ligne d'écoulement en régime permanent, cette étude a visé à investiguer l'utilisation du ²²²Rn pour déterminer les temps de résidence sur un site avec plusieurs puits et des conditions hydrauliques transitoires. La proximité des puits et l'estimation hydrodynamique des temps de résidence (2 à 7 jours) faisait du site à l'étude un candidat favorable pour l'utilisation du 222Rn comme traceur de temps. Des mesures de radon ont été faites pendant 1.5 ans et les temps de résidence ont été calculés pour chacun des échantillons en utilisant divers pôles de mélanges et valeurs à l'équilibre du ²²²Rn. Il a été déterminé que la proportion d'eau souterraine a un impact important sur les temps de résidence calculés. Ceci était particulièrement évident lorsque les proportions d'eau souterraine étaient faibles (<50%) et les temps de résidence calculés étaient typiquement courts (<5 jours). Aussi, la présence de stratification des traceurs dans l'aquifère complique les calculs de temps de résidence puisque certaines portions d'eau qui sont à l'équilibre avec le substrat ne sont pas visible avec cette approche. Malgré que cette méthode soit considéré bien développée par certains auteurs, des travaux sont encore requis pour établir les conditions exactes sur un site qui permette de produire des calculs de temps de résidence fiables. Des pôles stables, un niveau de confiance élevé dans la représentativité des mesures, et la présence de mélange binaire simple sont tous nécessaires pour que la méthode soit fiable.

Cette étude sensibilise à l'utilisation généralisée de la filtration sur berge pour l'approvisionnement de nombreuses municipalités au Québec. Elle met en évidence la nécessité de coupler divers outils de caractérisation. Ses résultats permettent des études plus ciblées basées sur les recommandations de ce manuscrit. Des études à l'échelle régionale utilisant des traceurs environnementaux pourraient aider à évaluer plus précisément l'occurrence de la filtration sur berge non seulement dans la province, mais aussi à une plus grande échelle au Canada et dans d'autres pays.

ABSTRACT

The study of bank filtration in the province of Quebec is still in its early stages, and an overall understanding of the prevalence of this type of groundwater extraction is unknown. During bank filtration, water is drawn towards a pumping well via a natural or induced gradient and water quality is improved relative to surface water. Due to the abundance of surface water in the province of Quebec, Canada, it is suspected that many groundwater wells are pumping a mixture of groundwater and surface water via induced bank filtration (IBF). The regulatory framework in Quebec provides comprehensive guidance for the development and monitoring of surface water and groundwater drinking water production systems. However, the regulations do not specifically address hybrid groundwater-surface water production systems such as IBF sites. Certain key parameters for attenuation of contaminants, notably residence times and mixing between regional groundwater and infiltrating surface water and the residence times of surface water are difficult to quantify using traditional methods that are the current standard practices. It is therefore important to i) better characterize the occurrence of bank filtration sites in the province in order to better evaluate whether new regulations should be adapted to these sites and, ii) develop innovative approaches that allow for characterizing groundwater flow at BF sites.

In order to provide a first evaluation of municipal wells potentially using IBF and the corresponding population served by these wells, a Geographic Information Science framework (GISc) was used to implement an IBF spatial database and calculate the distance from each well to the nearest surface water body. GISc is based on open-source GIS programs and openly available data, to facilitate the reproducibility of the works. From this provincial scale approach, we show that nearly one million people are supplied by groundwater from municipal wells located < 500 m from a surface water body and half a million would have a significant probability to be supplied by IBF wells. A more focused look at watershed scale distribution of wells allows us to improve our interpretations by considering the aquifer type and other regional factors. This approach reveals strong spatial variability in the distribution of wells in proximity to surface water. Of the three selected regions, one has high potential for IBF (Laurentides), one requires additional information do draw precise conclusions (Nicolet), and the third region (Vaudreuil-Soulanges) is unlikely to have widespread use of IBF. With this study, we demonstrate that extensive use of IBF is likely and that there is a need for improved understanding and management of these sites in order to properly protect the drinking water supply.

The second part of this study focuses on the use of ²²²Rn as a tracer for calculating residence times of infiltrating surface water. This study aims to incorporate the lessons learned from various studies into a more comprehensive understanding of the conditions when the ²²²Rn method is an effective method of evaluating residence times, and to test the impact of endmember choice on the calculations of residence time. Whereas the previous studies were typically investigating a single flow line under steady state flow conditions, this study was undertaken to evaluate the use of ²²²Rn as a tracer for calculating residence times in a multi-well bank filtration system under variable pumping and hydraulic conditions. On this site, the close proximity of pumping wells to surface water, and the residence times estimated in previous studies (2-7 days) made it a favourable candidate for use of the previously established method of residence time calculations using ²²²Rn. It was determined that the proportion of groundwater strongly impacts calculated residence times. This was particularly evident when the proportion of surface water was low (<50%) and the resulting residence times was typically short (<7 days). Additionally, aquifers with stratification of either ²²²Rn production or EC are not favorable for use of this method for determining residence times as there are portions of water at equilibrium that cannot be identified. Despite this method being considered well developed by some authors, additional work should be conducted to establish the exact hydrogeological conditions that are required for this approach to provide representative results. A clear stable radon end-member, high confidence in the representativity of samples and simple binary mixing are all essential for this. Several steps are recommended in order to improve this method and determine when and under what conditions it is appropriate.

This study raises awareness of the widespread use of bank filtration for supplying many municipalities within the province of Quebec. Its results enable more targeted studies based on the recommendations of this manuscript. Regional scale studies using environmental tracers could help to more precisely evaluate the occurrence of bank filtration not only in the province, but also at a wider scale in Canada and in other countries.

TABLE OF CONTENTS

| DEDICATION | III |
|--|--------|
| ACKNOWLEDGEMENTS | IV |
| RÉSUMÉ | V |
| ABSTRACT | VII |
| TABLE OF CONTENTS | IX |
| LIST OF TABLES | XII |
| LIST OF FIGURES | XIII |
| LIST OF SYMBOLS AND ABBREVIATIONS | XV |
| LIST OF APPENDICES | XVI |
| CHAPTER 1 INTRODUCTION | 1 |
| 1.1 State of knowledge | 1 |
| 1.1.1 Bank filtration overview | 1 |
| 1.1.2 General history of the study and use of bank filtration | 2 |
| 1.1.3 State of knowledge: Bank filtration in Quebec | 3 |
| 1.1.4 Characterization of the origin and residence times of infiltrating surface was pumping wells | |
| 1.2 Scientific questions | 6 |
| 1.3 Objectives of the study | 6 |
| CHAPTER 2 LITERATURE REVIEW | 8 |
| 2.1 Uses of GIS related to BF | 8 |
| 2.2 Assessing residence times of infiltrating groundwater | 11 |
| CHAPTER 3 DISCUSSION OF THE RESEARCH PROJECT AND C | ENERAL |
| ORGANISATION OF THE DISSERTATION | 13 |

| ARTICLE 1 EVALUATING BANK-FILTRATION OCCURRENCE IN THE F | PROVINCE OF |
|--|-------------|
| QUEBEC (CANADA) WITH A GIS APPROACH (PUBLISHED ARTICLE) | 14 |
| 3.1 Introduction | 15 |
| 3.2 Materials and Methods | 18 |
| 3.2.1 Data set | 18 |
| 3.2.2 Description of the GISc Approach | 20 |
| 3.3 Results and Discussion | 21 |
| 3.3.1 Overview of the Province | 23 |
| 3.3.2 Insights on Selected Sub-Basins | 27 |
| 3.4 Conclusions | 33 |
| CHAPTER 4 222RN AS A TIME TRACER OF SURFACE WATER IN | FILTRATION: |
| SENSITIVITY TO GEOCHEMICAL END-MEMBERS SELECTION | 35 |
| 4.1 Abstract | 35 |
| 4.2 Introduction | 35 |
| 4.3 Site description | 38 |
| 4.3.1 Hydrology | 38 |
| 4.3.2 Hydrogeology | 40 |
| 4.3.3 Pumping wells | 41 |
| 4.3.4 Transient flow regime | 43 |
| 4.4 Methodology | 43 |
| 4.4.1 Groundwater and Surface Water sampling | 43 |
| 4.4.2 Analytical techniques | 44 |
| 4.4.3 Quality control | 44 |
| 4.4.4 Residence time of infiltrating groundwater | 45 |
| 4.5 Results | 50 |

| 4.5.1 | Electric conductivity | 50 |
|---------|---|--------|
| 4.5.2 | Radon activities | 51 |
| 4.5.3 | Mixing ratios | 52 |
| 4.5.4 | Residence times of the bank filtrate | 54 |
| 4.6 I | Discussion | 57 |
| 4.6.1 | Impact of mixing rates on calculations of residence times | 57 |
| 4.6.2 | Impact of Rn end-member on residence time calculations | 57 |
| 4.6.3 | Evidence of spatial, vertical and temporal heterogeneity of the endmembers an | ıd the |
| impac | et on calculated residence times | 59 |
| 4.6.4 | Explaining the spikes in radon observed | 61 |
| 4.6.5 | Influence of pumping schemes | 61 |
| 4.6.6 | Implications of higher values measured in surface water | 62 |
| 4.6.7 | Potential evidence for a link between residence times and viscosity? | 63 |
| 4.6.8 | Recommendations | 63 |
| 4.7 | Conclusions | 67 |
| CHAPTER | S 5 GENERAL DISCUSSION | 69 |
| CHAPTER | 6 GENERAL CONCLUSION | 70 |
| REFEREN | ICES | 72 |

LIST OF TABLES

| Table 4.1: Summary of data available in the databases. | .19 |
|--|-----|
| Table 5.1: Hours pumped per month for each well | .42 |
| Table 5.2: Summary of all mixing ratio and radon scenarios tested. | .48 |

LIST OF FIGURES

| Figure 1.1: Figure taken from (Hiscock, K.M. & Grischek, 2002) Showing the processes that are |
|--|
| involved in attenuating contaminants in a bank filtration site. |
| Figure 1.2 Processes that occur in the hyporheic zone of the aquifer as surface water infiltrates toward a BF well (Tufenkji et al., 2002). |
| Figure 2.1: Criteria considered in the AHP analysis conducted by Lee and Lee 200910 |
| Figure 4.1: Spatial distribution of municipal drinking water source. The water sources are classified into three categories, i.e. groundwater (in blue), surface water (in red) and groundwater considered surface water (in green). |
| Figure 4.2: Distance of municipal wells from water surface for a) 0 to 2 000 m and b) 0 to 500 m |
| Figure 4.3: Cumulative population supplied by municipal groundwater wells with respect to the distance from surface water body |
| Figure 4.4: Location of the three selected watersheds (area#1: Laurentides, area #2 Nicolet, and area #3 Vaudreuil-Soulanges). |
| Figure 4.5: a) Spatial distribution of municipal wells located at less than 500 m from lakes and rivers in area #1 and distribution of wells according to b) the type of surface water and c) the type of aquifer. |
| Figure 4.6: a) Spatial distribution of municipal wells in the Nicolet area that are located at less than 500 m from lakes and rivers in area #2 and distribution of wells according to b) the type of surface water and c) the type of aquifer |
| Figure 4.7: a) Spatial distribution of municipal wells located at less than 500 m from lakes and rivers in area #3 and distribution of wells according to b) the type of surface water and c) the type of aquifer. |
| Figure 5.1: Study Site a) Location of the study site and drainage basins that feed the LDDM; b) all sample locations with bedrock geology; c) quaternary units surrounding the study site and setup of wells and observation wells |

| Figure 5.2: Water levels in LDDM (Blue), Pz15 level logger (Green), Pz15 point measurements |
|--|
| (red), calculated average value for LDDM since 1986 (orange). LDDM data from (Centre |
| d'expertise hydrique du Québec, 2019)41 |
| Figure 5.3: Schematic of the radon values used in the calculation of residence times |
| Figure 5.4: Boxplot of EC and ²²² Rn data for all sampling points based on measurements made |
| throughout the entire sample program. The number of data points used to generate the |
| boxplots are indicated above each boxplot and range from 1 point up to 16 data points50 |
| Figure 5.5: Mixing Ratios at all wells using each mixing ratio scenario. The dashed line represents |
| the average value throughout the sample campaign53 |
| Figure 5.6: Boxplot of residence times for all scenarios at each well |
| Figure 5.7: Calculated residence times for each of the residence time scenarios with residence times |
| exceeding 21 days removed. Note that scenarios A1, B1, and C1 have lower medians than A2, |
| B2, and C2, however they contain the most residence times exceeding 21 days56 |
| Figure 5.8: Time series of pumping hours, measured ²²² Rn, and measured EC for well 3. The EC |
| seems to have a direct correlation with pumping hours at this well62 |

LIST OF SYMBOLS AND ABBREVIATIONS

RBF Riverbank filtration IBF Induced bank filtration

BF Bank filtration

MAR Managed aquifer recharge AHP Analytical Hierarchical Process

LDDM Lac des deux montagnes

SW Surface water GW Groundwater

EC Electrical Conductivity

MR Mixing Ratio

LMWL Local meteoric water line

LIST OF APPENDICES

| APPENDIX A RESULTS | 82 |
|---|----|
| APPENDIX B CHLORIDE VS. EC | 90 |
| APPENDIX C IBF SITES GLOBALMAR DATABASE | 92 |

CHAPTER 1 INTRODUCTION

1.1 State of knowledge

1.1.1 Bank filtration overview

Bank filtration (BF) is a cost-effective method of improving the water quality of surface water (Hiscock, K.M. & Grischek, 2002; Tufenkji et al., 2002). As water passes through the banks of a lake or river towards a pumping well, either via an induced or natural gradient, a variety of processes can effectively remediate many contaminants that are present in surface water. These processes, which are both physical and chemical include adsorption, reduction, physicochemical filtration and biodegradation (Figure 1.1) (Hamann et al., 2016; Hiscock, K.M. & Grischek, 2002). BF benefits from ease of operation due to the generally shallow depths of exploitation, a comparatively higher production rate in relation to groundwater exploitations, and lower treatment costs (Maliva, 2020) In a well-designed and operated system, the water quality remains much more consistent than surface water and requires less treatment to ensure a safe drinking water supply (Maliva, 2020; Tufenkji et al., 2002).

Bank filtration can be conducted in a variety of settings, but it is most effective when conducted in alluvium or finer grained deposits that occur at the edges of lakes or rivers, as these types of deposits have favourable characteristics for the removal of contaminants (Tufenkji et al., 2002). These types of sediments generally have favourable hydrogeological characteristics (permeability and filtration efficiency) for effective bank filtration. The biogeochemical processes that govern the effectiveness of bank filtration depend on the mineralogy and shape of aquifer, the oxygen and nitrate concentrations in surface water, the types of organic matter present throughout the system, and the land use in the surrounding catchment (Hiscock, K.M. & Grischek, 2002).

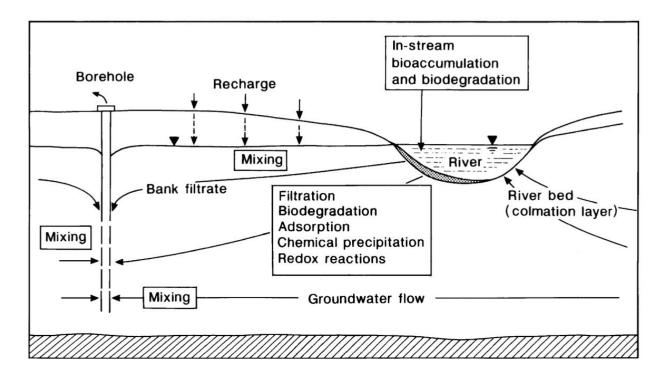


Figure 1.1: Figure taken from (Hiscock, K.M. & Grischek, 2002) Showing the processes that are involved in attenuating contaminants in a bank filtration site.

Despite its many benefits relative to surface water, water quality problems have been known to occur at many bank filtrations sites throughout the world. (Bertin & Bourg, 1994; Grischek, T. & Paufler, 2017; Paufler, S., Grischek, T., 2018; van Driezum et al., 2019).

1.1.2 General history of the study and use of bank filtration

Bank filtration has been used for over a century in Europe (ex. Düsseldorf. est. 1870 Schubert (2002)), and is becoming increasingly popular in North America due to the aforementioned benefits and has been used in the US for the last 60 years (Dillon et al., 2019). Although the first known uses of bank filtration occurred in Europe roughly 150 years ago, many of these sites improved the water quality sufficiently that they were able to operate without additional treatments other than disinfection (i.e. Düsseldorf). The Düsseldorf waterworks, being one of the oldest known BF sites, has experienced changes throughout its operation that can provide insights into other BF waterworks. For example, during the 1950s, river water quality decreased. Most notably, oxygen concentrations decreased and led to more anoxic conditions which favoured the solubilisation of iron and manganese. This required that additional treatments be put in place to remove iron and

manganese as well as ammonium and organic micropollutants (Schubert, 2002). These same problems are still encountered in more modern systems, however the knowledge gained from these more long-lived systems has not been transferred to all operating bank filtration sites. More broadly, studies of bank filtration became much more prevalent in Europe following a contamination event in the Rhine known as the Sandoz accident in 1986 (Dillon et al., 2019). The transfer of the scientific knowledge requires a clear classification of what a bank filtration system is, and an awareness of the technique among the hydrogeological community within a given region. In Quebec, for example, this classification does not exist, and has not been incorporated into the general framework of the regulatory system of public waterworks. It is therefore difficult to use best practices for BF sites and transfer knowledge from the longest-lived systems to the unclassified BF sites and those where BF is being performed inadvertently.

1.1.3 State of knowledge: Bank filtration in Quebec

Until recently, the use of bank filtration (BF) had not been studied in the province of Quebec. Since, the study of BF in Quebec is in its infancy, there are still many unknowns. In fact, the number of municipalities that are performing BF and the proportion of the population that they supply is unknown. The only classification system that touches on to surface water-groundwater interaction is the ESSIDES (Eau souterraine sous influences direct des eaux de surface) classification (Ministère de l'Environnement et Lutte contre les changements climatiques, 2019b). This classification is similar to the Groundwater under the direct influence of surface water (GWUDISW) classification system that was introduced in the USA in the 1990s (Dillon et al., 2019). Wells are classified as ESSIDES based on bacterial and virologic contamination at the well during an initial characterization period following construction. Drinking water production systems that are considered ESSIDES are subjected to the same regulations as surface water, which are more stringent than for all groundwater wells. This system is not designed to determine whether BF is occurring and therefore does not provide a complete framework for protection of wells. There are many wells that are performing BF within the province that do not have frequent bacterial contamination and therefore their vulnerability may not be adequately assessed. By specifically considering BF sites in the Quebec inventory of water intakes, knowledge related to common BF issues could be better integrated into the collective management of drinking water resources in the province.

1.1.4 Characterization of the origin and residence times of infiltrating surface water toward pumping wells

In recent years, along with studies of bank filtration in general, a large number of studies have been focused on the processes that occur in the hyporheic zone of lakes and rivers where exchanges between surface water and groundwater occur (Bourke et al., 2014; Cranswick et al., 2014). The complex processes that occur in this zone make it a distinct biochemical environment. Within this zone, there are changes in temperature, pH, redox potential, oxygen and organic carbon, which impact the quality of bank filtrate (Lewandowski et al., 2019; Tufenkji et al., 2002). These changes are most often caused by microbial activity related to the degradation of organic matter which occur in the early period following infiltration (Tufenkji et al., 2002) (Figure 1.2)

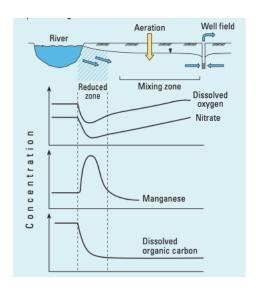


Figure 1.2 Processes that occur in the hyporheic zone of the aquifer as surface water infiltrates toward a BF well (Tufenkji et al., 2002).

Besides the biogeochemical conditions in the hyporheic zone, hydraulics exert a major control on the functioning of the underground reactors that represent BF sites. Indeed, the proportion of surface water in the bank and its residence time directly affect the location within the banks, type, kinetics and extent of such reactions, further complicating the interpretation of data, and leading to periods of greater vulnerability to certain contaminants. Characterizing the sources of water and their residence times at a BF site is therefore a crucial aspect in understanding its overall functioning. Under transient flow conditions, that are often experienced at BF sites, flow paths can vary significantly throughout the year since hydraulic conditions in surface water masses and neighbouring aquifers are variable, as well as pumping rates that vary greatly throughout the day and in different times of the year.

There is no scientific consensus of what proportion of surface water or residence time is required for a system to be classified as a BF system (Maliva, 2020). During the characterization of pumping wells by professionals in the field of hydrogeology in Quebec, the proportion of surface water and residence times are generally assessed through hydrodynamic modelling. The assumption commonly applied, like limited heterogeneity and steady state hydrodynamics, lead to results that may represent neither the high transience of groundwater flow nor the existence of preferential flow paths that can cause breakthrough of contaminants. Isotopic and geochemical tracers may provide significant insights on groundwater-surface water exchanges. Still, the transient pumping and hydraulic conditions are rarely taken into account and the representativity of the isotopic and geochemical end-members used for such characterization is rarely discussed. The province of Quebec, where BF sites face highly variable hydrology and are influenced by recharge from several sources such as flood-water, snowmelt, and rainwater, offers a great opportunity to investigate the use of isotopic and geochemical tracers.

Determining the potential sources of water can therefore provide insights into how the flow paths are changing as a result of these variations in hydraulic conditions. In ideal cases, the groundwater and surface water end-endmembers are sources that remain static throughout the year. Due to the complexity of the site presented within this study, its extremely important to evaluate whether the water sources are static in time and in space. Installation of push-point piezometers was used to determine whether there was stratification within the aquifer, and also to determine whether there are different groundwater signatures at either end of the well field. This is also important since such a large well field, could certainly have spatial variation. It is also widely known that infiltration of flood-water, snowmelt, and rainwater can lead to chemical stratification within the aquifer (Gilfedder et al., 2019).

In addition to understanding the changes in the different sources at the pumping wells, it is also important to understand the residence time of infiltrating surface water. The attenuation of many microbiological, virological, and pharmaceutical contaminants impacted by physical and chemical processes. Residence times have a significant impact on the removal of turbidity and bacteria (Sahu et al., 2019). The temperature and the abundance of dissolved oxygen can have a significant impact on the removal rate of organic micropollutants which generally have higher removal rates under warmer than colder temperature in an oxic environment (Burke et al., 2014), impacting the rate and stability of chemical reactions in the bank.

1.2 Scientific questions

In order to properly integrate the scientific knowledge of BF into the provincial water management strategy, it is first essential to determine whether bank filtration is taking place in the province and to what extent.

Question: Is bank filtration being used to supply municipal waterworks with drinking water in the province of Quebec?

Is the use of ²²²Rn appropriate for quantifying residence times in a multi-well bank filtration site with non-stationary flow conditions?

What is the impact of end-member selection on the results of a residence times calculations using well developed method of calculating residence times using ²²²Rn?

1.3 Objectives of the study

- Estimate the occurrence of BF within the province of Quebec using a GIS approach
- Evaluate the potential of ²²²Rn as a tracer for calculating residence times of infiltrating surface water in order to better assess the sites vulnerability
 - Evaluate the possibility of using ²²²Rn at this site for monitoring of residence times
 - Evaluate the impact of end-member choice for calculating mixing ratios and the ²²²Rn activity in surface water and groundwater on the results of the ²²²Rn method

 Determine the reliability of the ²²²Rn and evaluate the conditions required for the method to provide meaningful results.

CHAPTER 2 LITERATURE REVIEW

This literature review drew from other literature reviews within the introductions of many studies and on literature reviews that compiled large quantities of information whenever these were available in order to benefit from the work of the colleagues in the field of managed aquifer recharge (MAR), a broad term that encompasses BF and many other groundwater management strategies. Recharge in a hydrogeological context is defined as the proportion of precipitations that are not intercepted by natural processes such as evapotranspiration and runoff, and reach the water table. MAR on the other hand is recharge that is not related to precipitations but is rather generated by anthropic processes. In the case of recharge during BF, the recharge being discussed is the anthropogenic recharge that is induced by the drawdown generated by pumping in proximity to surface water.

Due to the varied subjects covered throughout this project, the literature review was separated into several sections. For the initial study of widespread use of BF throughout the province, the study of the use of GIS in BF was conducted. The second section which involved using tracers for characterizing the site involved extensive study of the use of radon as a tracer of residence times, and a complimentary review of the use of other tracers. Finally, a review of regional studies was done to gain a more thorough understanding of the site history, and the geology of the site as well as to gain a better understanding of the history of quality related problems at the site.

2.1 Uses of GIS related to BF

Geographic Information Systems (GIS) and Multi-Criteria Decision Analysis (MCDA) approaches are being developed for site suitability mapping for the development of managed aquifer recharge (MAR) sites (Sallwey et al., 2019) and IBF (Lee & Lee, 2010; Wang, L.X. et al., 2016). The use of suitability maps has been widespread and has seen a variety of techniques developed using different types of spatial data for an assortment of types of MAR (Sallwey et al., 2019). The use of GIS-MCDA involves combining and weighing geospatial data which is used to meet the studies objectives by characterizing the area in question. These studies are designed to evaluate land at various scales in order to develop future sites for MAR use. They are not designed for pinpointing specific locations for well placement but rather are designed to more broadly evaluate which

locations within a given area have favourable hydrogeological conditions or unfavourable characteristics such as proximity to contaminant sources (Sallwey et al., 2019).

A few studies of varying degrees of complexity have been performed for site suitability development of riverbank filtration sites. The simplest of these studies was completed by (Razak et al., 2015) along the Langat River. This study involved use of remote sensing data to classify land usage. Following this, land that was classified as barren land or range land, was combined, using the Boolean AND operators, with locations within 10-20% of the width of the river and 15 meters from built up areas to produce a suitability map with a single layer that met all three criteria.

A second, more elaborate site suitability study was developed to evaluate the potential for BF along a portion of the Second Songhua River basin (China) and involved the development of a spatial index for site suitability mapping (Wang, L.X. et al., 2016). This study took into consideration large scale interpolated data for water quantity (hydraulic conductivity, aquifer thickness, and surface runoff), water quality (status of groundwater quality, status of surface water quality), interaction intensity of surface water and groundwater (groundwater hydraulic gradient, width of interaction zone between surface water and groundwater, permeability of riverbed) and the exploitation condition of the groundwater resource (depth to groundwater). These data were then combined with various weights to produce a final map of the river basin with 5 classes with varying degrees of suitability. A second study was also performed on the Songhua River by a different group of researches with the same approach (Yin et al., 2018).

A thesis was also written on site suitability for RBF in Columbia (Uribe, 2015) in which they studied the impact of stream morphology on bank filtration sites. In this study, successful RBF sites in the US and Europe were examined from a geomorphological perspective and then compared to potential sites in Columbia. The specific characteristics that were considered when classifying stream morphology in this study were the slope of the reach and basin area. An additional step to evaluate stream reach features involved calculating the width, sinuosity, radius of curvature, the amplitude, and bend wave length. The risk of clogging was evaluated using stream power.

A study in South Korea was conducted using an Analytical Hierarchical Process (AHP), which is used to select the optimal alternative via a hierarchical classification of various attributes

followed by a measure of the importance of each attribute (Lee & Lee, 2010). The analysis was based on the parameters shown in Figure 2.1

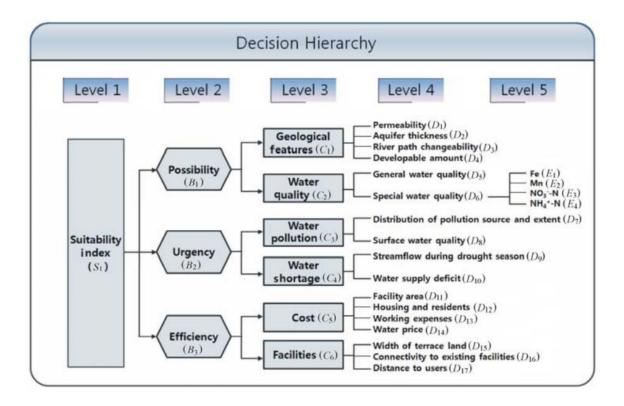


Figure 2.1: Criteria considered in the AHP analysis conducted by Lee and Lee 2009.

These studies were all developed for the site selection phase and therefore parameters that would make for a favourable BF site were considered. Since the study that was undertaken herein was focused on identifying existing sites, these parameters were not considered. A similar approaches was attempted with certain layers of DRASTIC (Aller et al., 1985) data however the lac of variability of the parameters did not allow for distinctions between wells to be made.

The specifics of the various datasets are presented in section 3.2.1. There is no clear definition of a minimum residence time required for surface water to be considered as groundwater (Dillon et al., 2019). There is also no specific classification that exists to determine what proportion of surface water is required to be classified as BF (Maliva, 2020) and the existing sites throughout Quebec are not likely to be optimally designed for use as BF sites since during their initial conception, they were not developed with that specific purpose. For this reason, a simple GIS

method, and broad characterization was used during the initial classification of potential BF sites in Quebec.

Although several studies have been done to evaluate potential for BF during the site selection phase, to our knowledge, this is the first study that uses a Geographic information science (GISc) approach to identify the number of existing IBF sites. The objective of this study is to provide a first overview of the potential extent of IBF in the province of Quebec. The framework is based on the concept of Geographic Information Science (GISc), described in (Singleton, 2016) and (Malczewski, 2015). GISc is based on the use of open-source GIS programs and openly available data. This concept facilitates reproducible research to other areas of study and data sets and could improve the transparency of research using GIS.

2.2 Assessing residence times of infiltrating groundwater

There exists a number of tools to investigate the residence times of infiltrating groundwater. Models calibrated using hydraulic head measurements have been used to estimate residence times however they cannot reliably reproduce mixing ratio or residence times in BF systems (Schilling 2017). Various chemical and physico-chemical tracers also have been used including: temperature (Constantz, 2008; Wang, W.-s. et al., 2020), and EC (Cirpka et al., 2007; Vogt et al., 2010). 3 H/ 3 He (Massmann, G. et al., 2008; Massmann, Gudrun et al., 2009), time series of stable isotopes of water (Małoszewski & Zuber, 1982; Massmann, G. et al., 2008; Moeck et al., 2017; Sprenger, Christoph, 2011), 37 Ar (Schilling et al., 2017) each of which provides information on a certain temporal gap. More recently studies have combined both models and tracers to gain additional insights into the behaviour of groundwater (Gilfedder et al., 2019; Liu & Yamanaka, 2012; Maloszewski & Zuber, 1996). This study, is focused on investigating the use of 222 Rn, which has been used to assess residence times of infiltrating groundwater (Bertin & Bourg, 1994; Hoehn & von Gunten, 1989; Schilling et al., 2017; Stellato et al., 2013).

A combination of factors makes ²²²Rn a particularly favourable tracer for quantifying relatively short residence times with a cited resolution of up to 21 days in certain studies depending on the precision of measurements (Gilfedder et al., 2019). The properties that make ²²²Rn an ideal tracer are:

- In general, ²²²Rn is present in much greater concentrations in the groundwater than in surface water (Stellato et al., 2013) since it will degas rapidly in turbulent surface water as well as experiencing radioactive decay.
- ²²²Rn is a noble gas (Freyer et al., 1997), and therefore it will not react with the substrate as it infiltrates.
- As ²²²Rn originates as part of decay of ²³⁸U, it is ubiquitous throughout all geological environments (Lefebvre et al., 2015; Stellato et al., 2013).
- ²²²Rn follows a typical radioactive in-growth curve when infiltrating into an aquifer (Hoehn & von Gunten, 1989) and activities in water will increase until secular equilibrium with the substrate is reached, which takes roughly 20 days.

When using ²²²Rn to determine residence times of infiltrating surface water, one of the steps that is required is determining the value at secular equilibrium of the substrate which occurs after 5 half-lives or roughly 21 days (Gilfedder et al., 2019). This allows for the determination of the emanation rate of the sediments

CHAPTER 3 DISCUSSION OF THE RESEARCH PROJECT AND GENERAL ORGANISATION OF THE DISSERTATION

Since the study of bank filtration is still in its early stages in the province of Quebec, work is required to understand not only how frequent this technique of groundwater management is but also to develop cost-effective techniques for understanding these sites. As such, the first portion of this dissertation takes a simple approach using GIS data throughout the province of Quebec to provide an overview of the number of sites that are potentially using bank filtration. The material presented has been published in the journal Water (Patenaude et al., 2020).

The second portion of the dissertation focuses on evaluating the potential use of ²²²Rn as a cost-effective tracer for quantifying and understanding residence times of infiltrating surface water at a complex bank filtration site. A separate literature review as done for each of these aspects. The use of ²²²Rn has been examined periodically to quantify residence times of infiltrating groundwater in a variety of contexts, however its potential as a tracer in a hydraulically complex bank filtration site remains uncertain.

Additional work was done using additional tracers although it is not presented in this document.

ARTICLE 1 EVALUATING BANK-FILTRATION OCCURRENCE IN THE PROVINCE OF QUEBEC (CANADA) WITH A GIS APPROACH (PUBLISHED ARTICLE)

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Journal: Water

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Abstract: Due to the abundance of surface water in the province of Quebec, Canada, it is suspected that many groundwater wells are pumping a mixture of groundwater and surface water via induced bank filtration (IBF). The regulatory framework in Quebec provides comprehensive guidance for the development and monitoring of surface water and groundwater drinking water production systems. However, the regulations do not specifically address hybrid groundwater-surface water production systems such as IBF sites. More knowledge on the use of IBF in the province is thus needed to adjust the regulations with respect to the particularities of these systems. In order to provide a first evaluation of municipal wells potentially using IBF and the corresponding population served by these wells, a Geographic Information Science framework (GISc) was used to implement an IBF spatial database and calculate the distance from each well to the nearest surface water body. GISc is based on open-source GIS programs and openly available data, to facilitate the reproducibility of the works. From this provincial scale approach, we show that nearly one million people are supplied by groundwater from municipal wells located < 500 m from a surface water body and half a million would have a significant probability to be supplied by IBF wells. A more focused look at watershed scale distribution of wells allows us to improve our interpretations by considering the aquifer type and other regional factors. This approach reveals strong spatial variability in the distribution of wells in proximity to surface water. Of the three selected regions, one has high potential for IBF (Laurentides), one requires additional information do draw precise conclusions (Nicolet), and the third region (Vaudreuil-Soulanges) is unlikely to have widespread use of IBF. With this study, we demonstrate that extensive use of IBF is likely and that there is a need for improved understanding and management of these sites in order to properly protect the drinking water supply.

Keywords: managed aquifer recharge (MAR); induced bank filtration (IBF); geographic information science (GISc); drinking water supply; guidelines

3.1 Introduction

Induced bank filtration (IBF) is a widely used method of managed aquifer recharge (MAR) (Umar, 2017). In an IBF system, surface water is drawn through the banks and/or bed of a lake or river towards a pumping well by an induced hydraulic gradient. This results in a pumped mixture of both groundwater and infiltrated surface water. This process reduces the risk of intensive use (Llamas & Custodio, 2002)of the aquifer and improves water quality relative to surface water (Gillefalk, 2018). The drawback of this method however is that there is a higher risk of contamination from certain sources in comparison to standard groundwater exploitations (Hellauer, 2018; Hiscock, K., Grischek, T., 2002; Sprenger, C et al., 2011a; Tufenkji et al., 2002), which makes it important to develop a specific regulatory framework adapted for this type of drinking water supply system.

In recent reviews of the international use of bank filtration (Dillon et al., 2019; Sprenger, C. et al., 2017; Stefan, C. & Ansems, 2016; Stefan, C., Ansems, N., 2018), it is clear that IBF has been used worldwide and in particular in Europe for more than 100 years. In the last decades, the use of IBF also became relatively popular in USA (Gillefalk, 2018 and references therein). More recently, few IBF systems have also been implemented in developing countries (Gillefalk, 2018 and references therein). The use of this technology is however completely overlooked in Canada and the province of Quebec in particular. As of this point in time, no inventory of IBF sites exists for the province and the extent of its use throughout the province remains unknown. There is however a high probability that IBF is widely used. Indeed, Quebec is a province rich in both surface water and groundwater with 22% of its surface covered by water either in the form of lakes, rivers, or wetlands (MDDELCC, 2014). It is estimated that Quebec contains 3% of the Earth's renewable freshwater resources (Boyer, 2008). In addition, Quebec was settled through its waterways which has had long term effects on its population distribution. The abundance of water and the distribution of the population mean that there is high probability that pumping wells are located in close proximity to surface water. The process of unintentional IBF is thus likely to be occurring in many municipalities throughout the province.

In addition to the number of IBF sites throughout Quebec being unknown, the current regulations and guidelines in Quebec do not specifically address wells that pump a mixture of surface water and groundwater. In Quebec, the *Règlement sur le prélèvement des eaux et leur protection* (RPEP) provides comprehensive guidelines for protecting surface water and groundwater extractions from contamination (Government of Quebec, 2019). This regulation provides a framework for monitoring contaminants that allows for appropriate intervention when groundwater resources contain surface microbiological contaminants, which ensures that the population is not likely to be affected by changes in water quality. Additionally, drinking water production systems that are considered Groundwater under the Direct Influence of Surface Water (GWUDI) are subjected to the same regulations as surface water, which are more stringent than for all groundwater wells. For instance, in such a case, the raw pumped water is tested weekly for microbiological parameters, whereas groundwater be tested monthly can (Ministère de l'Environnement et Lutte contre les changements climatiques, 2019b). Despite its name, the GWUDI classification does not aim at characterizing infiltration from surface water bodies. Rather, what is referred to as "surface water" within this classification system is in reference to any source of contamination from the surface (including septic tanks) that might provide recurrent microbial or viral contamination to a well. There is therefore no correlation with surface water bodies that are hydraulically connected to an aquifer unless said surface water bodies is considered a potential source of contamination or if there is persistent bacteriologic or virologic presence in the pumping well during initial characterization. Existing non-GWUDI IBF sites are therefore necessarily treated as standard groundwater extractions. This means that even if the groundwater wells are located within a few meters of a water body, and there is a hydraulic connection between them, there is a lack of protection guidelines for the nearby surface water.

Due to the lack of regulations specific to hybrid groundwater-surface water systems, many problems common to IBF sites may be overlooked or unanticipated during the planning and operation of these sites. IBF sites are sensitive to changes in surface water quality (Sprenger, C et al., 2011a, 2011b; Stuyfzand, P. J., 1989), changing redox conditions, and changes in the hydraulic conditions of the site (Masse-Dufresne et al., 2019). Additionally, there are many undesirable chemical components and contaminants that can be found at pumping wells in IBF sites including Mn (Paufler, S. & Grischek, 2018; Paufler, S. et al., 2018; Paufler, S et al., 2018), Fe (Grischek, T., Paufler, S, 2017; Romero-Esquivel, 2017), NO₃, organic micropollutants (Dragon et al., 2018;

Moeck et al., 2017; Trásy, 2018; van Driezum et al., 2019), cyanobacteria (Grützmacher, 2010; Pazouki, 2016; Rose, 2018), coliforms (Kumar, 2018). In order to have a more resilient water supply the risk posed by these contaminants should be identified as early on in the development of the site as possible in order to minimise unforeseen costs and develop plans to reduce the risk. By identifying the potential contaminants at the sites related to changing hydraulic and chemical conditions, the risks associated with them would be reduced by allowing for strategic planning when the conditions associated with poorer quality are encountered. For example, well configuration and pumping schemes could be modified to increase transit times from surface water to pumping wells producing a more consistent water quality.

Geographic Information Systems (GIS) and Multi-Criteria Decision Analysis (MCDA) approaches are being developed for site suitability mapping for the development of MAR (Sallwey et al., 2019 and references therein) and IBF (Lee & Lee, 2010; Wang, L., Ye, X., Du, X., 2016). As an example, Jamarillo Uribe (Uribe, 2015) has studied the impact of stream morphology on bank filtration sites. Nonetheless, to our knowledge, this is the first study that uses a GISc approach to identify the number of existing IBF sites. The objective of this study is to provide a first overview of the potential extent of IBF in the province of Quebec. The framework is based on the concept of Geographic Information Science (GISc), described in (Singleton, 2016) and (Malczewski, 2015). GISc is based on the use of open-source GIS programs and openly available data. This concept facilitates reproducible research to other areas of study and data sets and could improve the transparency of research using GIS. First, we have processed and homogenized the sources of information from three different agencies. Secondly, we have carried a pre-quantification of municipal wells with a higher likelihood of providing drinking water through IBF using easily available government data. Following this pre-selection, zones with varying characteristics are considered in greater detail and the likelihood of IBF taking place in these areas are discussed.

3.2 Materials and Methods

3.2.1 Data set

3.2.1.1 Well data

Well data in the province of Quebec is contained in a variety of databases. All private wells require that various information is reported following drilling such as coordinates, ownership, well design (i.e. type of tubing and depth of well) and stratigraphic sequence. This data is compiled in the Système d'Information Hydrogeologique (SIH) database (MELCC, 2018) which has previously been described by Sterckx (Sterckx, 2013). The SIH database consists of information largely provided by well drilling companies during destructive drilling. This can lead to variability in the geological descriptions and the precision of the coordinates. This database is however considered a reliable source of information concerning the depth of the contact between overburden and bedrock (INRS-ETE, 2012).

The database with the most extensive information is available through a series of studies entitled "Programme d'Acquisition de Connaissances sur les Eaux Souterraines" (PACES) (Ministère de l'Environnement et Lutte contre les changements climatiques, 2009-2015). These projects were initiated in 2008 and aimed at improving knowledge of groundwater resources in the southern regions of the province of Quebec in order to protect them and ensure their sustainability. These studies have led to a number of subsequent publications (Beaudry et al., 2018; Gagné et al., 2018; Ghesquière et al., 2015; Nadeau, 2018). As of today, the PACES studies have been completed on a total of 13 regions throughout the province and led to the compilation of varied information (i.e. depth of well, depth of screen, depth of water table, type of aquifer, type of well and coordinates) on a total of roughly 180,000 wells. Supplemental information, including geochemistry, geology and hydrogeological data (e.g. hydraulic conductivity), was also compiled for a small subset of wells (n=15,162). The PACES database contains significant overlap with the SIH database and therefore some issues with the reliability of the coordinates and geological descriptions also affect this database.

The third database is comprised of municipal wells and surface water extraction points (MELCC, 2017). This database is typically used by decision makers for public health and land-use planning. The version of the database used in this study was acquired in 2017 (i.e. prior to the most recent

update in November 2018). The information contained in this database is centered on geographic coordinates, population served and types of potabilization treatment. It contains information on 2,116 individual wells and surface water extraction points. The geographic coordinates are generally more recently acquired and derived from official declaration documents, resulting in a better precision of the localization of the wells than the two other databases.

The municipal database formed the basis of this study with complimentary information pulled from the other sources. The decision to focus on the municipal well database was made since i) IBF requires sufficiently high pumping rates to induce a hydraulic gradient from the surface water to the well, ii) the localization of the wells is precise and iii) all these wells are supplying drinking water to the population. The PACES database was used to extract complementary information and draw some general conclusions about the distribution of wells within the province. Table 0.1summarizes the data sets and the variables used in this study.

Table 0.1: Summary of data available in the databases.

| Information | SIH | PACES* | Municipal* |
|-------------------|----------|----------|------------|
| Number of wells | ~216,000 | ~180,000 | ~2,000 |
| Depth of well | X | X | |
| Depth of screen | | Χ | |
| Geology | (X) | (X) | |
| Chemistry | | (X) | |
| Population Served | | | X |
| Type of aquifer | (X) | X | |
| Type of well | | Χ | X |
| Type of treatment | | | X |
| Coordinates | X | X | X |

^{*}databases used for subsequent calculations; (X) not systematically compiled.

3.2.1.2 Surface Water Bodies and other Data

Natural Resources Canada's (NRCAN) website contains a variety of vector data in the Canvec portion of the site (NRCAN, 2016). The surface water files are subdivided into two categories, "water courses" and "water bodies". Many of the features in the "water courses" files are drainage ditches and ephemeral streams that are not likely to be supplying bank filtrate to wells throughout the year. The features in the "water bodies" files correspond to water bodies of larger size and more permanent nature, which are more likely to contain a sufficient volume of water to support a municipal water supply. Considering the above, it was decided to conduct our province-wide study with the "waterbodies" files only. The named rivers from the "water courses" files were added to

the regional studies. These files contain two types of data, i.e. polygons and polylines, respectively representing the water bodies and shorelines.

3.2.2 Description of the GISc Approach

In this study, we used a GISc approach in order to calculate the minimal distance between each municipal pumping well and the nearest surface water body (i.e. lake or river). Throughout this study, the distance to surface water is calculated with respect to the "waterbodies" files retrieved from NRCAN website, as mentioned above. The work process has been carried out with the Quantum GIS program (QGIS) (Team, Q.D., 2020). In this subsection, we list a number of steps that were taken in order to perform the spatial analysis.

3.2.2.1 Processing and homogenization of spatial data sets

• Removal of duplicate wells

The work process is initialized with spatial data debugging. The database of municipal wells contained a number of duplicated geometries, wells with identical coordinates which caused issues when performing the calculations. The duplicate data most often resulted when wells were supplying water to multiple municipalities. In order to remedy this, the duplicate geometries were automatically identified and removed from the spatial database. In cases where discrepancies were found in the attribute table, the correct points were identified and retained.

Homogenization of the spatial reference system

Coordinates needed to be converted into a common projected coordinate system in order to do subsequent distances calculations. Spatial data was reprojected from original coordinate system (i.e. NAD 83) to Lambert EPSG: 32198. The Quebec Lambert was selected since it is suitable for use in Quebec (Canada) and for applications with an accuracy of < 2 m (Geomatic Solutions, 2018).

• Conversion of geometries

First, the geometries were converted from multipart to single part. This step was necessary in order to ensure that the conversion to lines in the following step inserted all the necessary points. Without this step, only the existing nodes of the polyline file were converted to points which led to an uneven and wide spread of points along the shorelines. Second, waterbody files were converted from lines to points with 2m spacing with the function *Convert lines to points* in the SAGA (Conrad

et al., 2015) toolbox. This function was run as a batch process for all of the 1:50,000 National Topographic System (NTS) zones individually.

3.2.2.2 Performing distance calculation

Calculate distance from each well to the nearest point

Using the smaller 1:50,000 zones allowed for the direct calculation from each well to the nearest point that was generated along the edge of the water body in the previous step. This calculation was completed using the *Distance to nearest hub* function. This algorithm identifies the nearest feature to each point and the Euclidean distance between all points. This however led to the creation of roughly 220 different files each containing one distance value for each well. This avoided some problems that occurred when the calculation was done in the reverse order, but it created extremely large files that were not easy to work with in QGIS especially for the larger PACES files. Each of the calculations therefore required a significant time to be realised ranging from 45 minutes to over 2 hours depending on the size of the files. With the number of iterations that were required to polish and develop the procedure, this required several months of computation.

Calculate the minimum value for each well

The subsequent step was to merge all of these roughly 220 files and calculate the minimum value for each well. This was done in QGIS using the *merge* function and the minimum selection was made using the *Select by expression* dialogue box and the following line of code HubDist=minimum (HubDist, Well ID). This could only be done in QGIS for the smaller municipal well files. For the larger PACES files, the merge and distance calculations were completed using R (Team, R.C., 2019) programming language. Once the distance was calculated, manipulation of the data was done manually using filters in QGIS and R. Additional data was also joined to the well files either manually, or using common fields or spatial relationships using a variety of functions in QGIS. This process was repeated for the regional studies with named rivers from the watercourse file and the water bodies used in the province-wide study.

3.3 Results and Discussion

There are two main conditions that will determine whether a well is pumping a mixture of surface water and groundwater. First, a hydraulic connection between the surface water and the aquifer is

necessary. Second, assuming groundwater typically discharges into surface water bodies in the study region (Larocque et al., 2010), it is necessary to pump a sufficient volume to reverse that natural gradient and draw surface water toward the pumping well. The reversal of the hydraulic gradient will depend on a range of factors as well, including the hydraulic conductivity of the aquifer, the proximity of the wells to the surface water, the pumping rates, the duration of pumping, the natural hydraulic gradient, and the recharge from precipitations. In order to conduct a study at the scale of the province, we used the municipal well database (description available in section 2.1.1), which reports the geographical coordinates and the population served by each of the 2,116 municipal wells and surface water extraction points since these wells are likely to be pumping a volume that is high enough to meet the second condition required for IBF to take place. Since the type of aquifer is not reported within this database, the distance from the surface water bodies was deemed to be the most important criteria for estimating the number of IBF sites available for a province-wide scale. Then, we selected three areas with distinct characteristics in order to conduct regional analyses including integrating the type of aquifer and other hydrogeological information. The results of both province and regional scale approaches are presented and discussed in the next subsections.

Bank filtration sites can be located at various distances from surface water bodies (Kumar, 2018; Stuyfzand, Pieter J. et al., 2006) from a few meters to greater than one kilometer. In this study, as a starting point, distances > 500 m are considered less likely to be performing IBF, as wells were located < 500m from surface water in several studies on IBF. Groundwater will discharge from aquifers towards rivers and lakes in most of Quebec for the majority of the year, especially during drier months (Larocque et al., 2010), therefore wells must be able to reverse that natural gradient in order to perform IBF. In addition, most of the sites have been operational for at most a few decades, are not likely to have experienced intensive use (Llamas & Custodio, 2002) and therefore large scale flow regime reversals, as seen in the Netherlands (Stuyfzand, Pieter J. et al., 2006), are not likely. These combined factors make it unlikely that wells at a distance > 500 m are performing IBF since, reversal of a gradient over those distances would be more difficult. In addition, wells that are located at a distance < 500m, are not necessarily conducting IBF as they require certain conditions for the surface water to be drawn through the banks towards the well which are outlined above. This choice for the cut-off could potentially omit some bank filtration sites from the province, however this is considered to have minimal impact on the overall conclusions of this

study. A similar evaluation of BF sites that are inventoried in the GlobalMAR database (Stefan, C. & Ansems, 2018) developed in the present work illustrate that the bulk of the IBF sites are located within the first 500m. This information is presented in Appendix C.

3.3.1 Overview of the Province

3.3.1.1 Municipal Drinking Water Supply: Surface Water vs Groundwater

Municipal drinking water sources can be classified into three categories, i.e. groundwater, surface water, and groundwater considered surface water, as shown in Figure 0.1. The first category (in red) includes all municipal drinking water sources which rely on one or multiple groundwater wells. Contrastingly, the second category (in blue) refers to municipal drinking water sources supplied by surface water. The third category (in green) corresponds to the few municipal drinking water sources relying on groundwater wells which are documented as GWUDI according to the protocol detailed in the Guide de conception des installations de production d'eau potable described in section 1 (Ministère de l'Environnement et Lutte contre les changements climatiques, 2019b). Of the 2,075 municipal extraction points, 87% (n=1,799) are groundwater pumping wells, whereas 13% (n=276) are directly extracting surface waters. This results in approximately 15% of the population of Quebec relying on groundwater resources for drinking water, representing roughly 1 260 000 citizens. A similar estimate (i.e. 20%) was also reported by the *Ministère de l'environnement* lutte les changements et contre climatiques (Ministère de l'Environnement et Lutte contre les changements climatiques, 2009-2015). discrepancy between these two estimates is likely due to the proportion of the population supplied by private wells rather than a municipal distribution network.

In the more densely populated areas, surface water is the main water source for drinking water supply systems. This is likely a consequence of the greater drinking water demand in the cities, and the larger population's ability to support the more costly water treatment required for surface water. In smaller municipalities, groundwater sources are preferred as they are less costly to operate. In fact, in rural areas of the province, 90% of the population is served by groundwater sources (Rousseau et al., 2004). As explained above, the important number of surface water bodies and the population's distribution results in a high likelihood of having a groundwater well near a surface

water body, suggesting that many municipal drinking water systems may be benefiting from IBF processes.

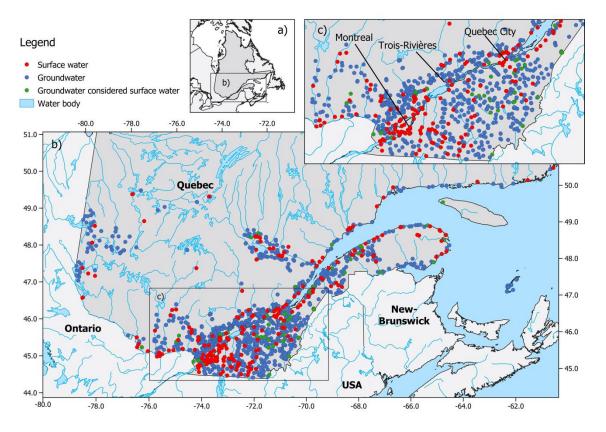


Figure 0.1: Spatial distribution of municipal drinking water source. The water sources are classified into three categories, i.e. groundwater (in blue), surface water (in red) and groundwater considered surface water (in green).

3.3.1.2 Water Bodies Distribution around Wells

Among the municipal wells, almost all (97% of the cases; n=1,749) are located at < 2,000 m from a surface water body. As illustrated by Figure 0.2a), it is evident that the closest water body to most municipal wells are lakes (72%; n=1,262). This is likely due to the extremely high number of lakes in Quebec which means that there is likely always a lake within a reasonable distance from any given point in the province, resulting in a geographically homogeneous distribution of municipal wells near lakes. The distribution of wells in close proximity to rivers (28%; n=487) is less homogeneously distributed throughout the province compared to those in proximity to lakes (see Figure 0.2). Many municipalities are located in close proximity to a river due to the settlement of the province through its waterways. Secondly, the area around rivers can often have more favorable

properties for larger scale water extraction due to the higher likelihood of containing sandy granular deposits compared to the more common glacial deposits that cover the rest of the province (Ministère de l'Environnement et Lutte contre les changements climatiques, 2019a).

Figure 0.2b) also reveals the same trend of homogenous distribution of wells in proximity to lakes for each bin of 10 m width. In fact, 14% (n=245) of the municipal wells are located at < 200 m from a river. The overall distribution of wells shows a significant decrease in the density of wells at around the 120 m mark. Since the number of wells in proximity to lakes remains relatively constant in all 10 m bins, and the distribution of wells near rivers decreases around the 120 m mark, the overall trend in this graph of more wells in the first 120 m is controlled mostly by the greater number of wells in proximity to rivers.

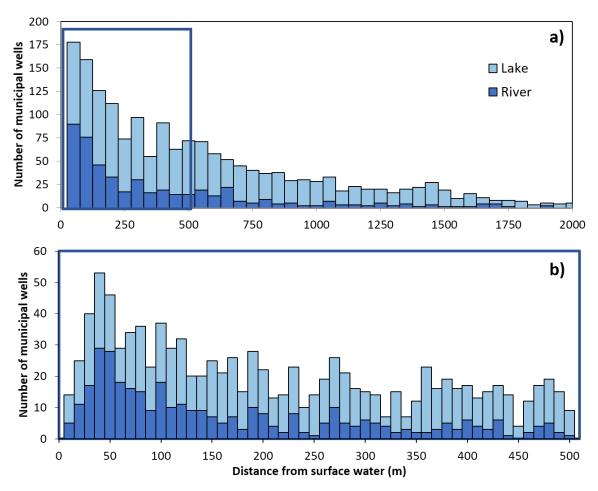


Figure 0.2: Distance of municipal wells from water surface for a) 0 to 2 000 m and b) 0 to 500 m.

3.3.1.3 Insights into the Potential Population Supplied by IBF

Figure 0.3illustrates the cumulative population served by municipal wells that are located at a distance of 0 m to 2,000 m from a surface water body. It shows that approximately 1,200,000 people are served by those municipal wells. It also reveals that outside of the major cities, wells located at less than 500 m from a surface water body account for 74% of the population (n=920,000) whose drinking water is supplied by groundwater from municipal wells. In fact, there is a rapid increase in population served by wells within the first few hundred meters of a water body. Wells within 250 m and 100 m account for 57% (n=720,000) and 34% (n=410,000) of the population served by municipal wells respectively. Although many of these sites may not be using IBF due to various factors, these results reveal a significant probability that half of the groundwaterfed population connected to a municipal drinking water supply in Quebec, i.e. more than half a million citizens, might depend of IBF. This initial overview demonstrates the great opportunity of a better assessment of IBF occurrence in the province of Quebec in order to better protect our resources. This high proportion of close-to surface water wells is likely an indication that planners and developers are in fact selecting pumping sites in close proximity to surface water. Still, as aforementioned, the existing knowledge of IBF is not fully integrated into the planning process when developing and managing water abstraction plants.

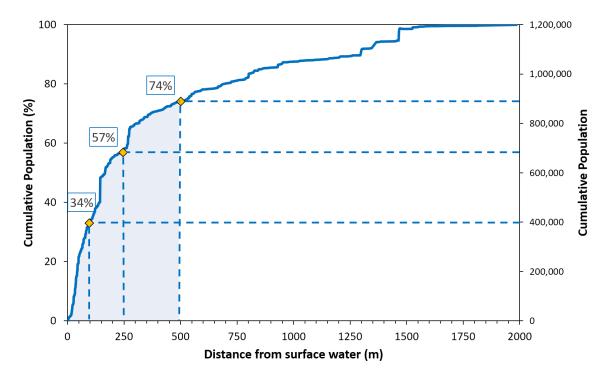


Figure 0.3: Cumulative population supplied by municipal groundwater wells with respect to the distance from surface water body.

3.3.2 *Insights on Selected Sub-Basins*

In light of the results presented in the previous subsection, it was determined that making widespread generalizations about the province would not be straightforward. We opted to focus on a selection of areas with diverse population sizes, diverse geological settings, a variety of distances from surface water, a variety of surface water body types, and a variety of land coverage types. By cross-checking the ID and localisation of wells in the available databases, it was possible to manually assign the type of aquifer (fractured bedrock, unconfined granular, confined granular) to 101 municipal wells within the 3 areas, namely Laurentides (area #1), Nicolet (area #2) and Vaudreuil-Soulanges (area #3). These areas correspond to watersheds "du Nord", "Nicolet", and "Vaudreuil-Soulanges" respectively. It is important to note that the "Vaudreuil-Soulanges" watershed also extends into the neighbouring province of Ontario to the West, but the investigation is limited to the portion within the Quebec border. The localization and the extent of each area is illustrated in Figure 0.4. The main geological contexts are also reported on this figure. The Grenville Province (area #1) is mainly composed of Archean autochthonous rocks dominated by highly metamorphosed gneissic complexes. The Appalachian Province (area #2) is composed of

various types of strongly deformed rocks (i.e. sedimentary, volcanic and ophiolitic rocks), whereas the St-Lawrence Platform (area #3) corresponds to sedimentary rocks. The criteria used to evaluate each region are the nature of quaternary deposits, the type of nearest water body in proximity, the population of the municipalities, distance of wells, as well as certain qualitative factors that are mentioned in the PACES reports for each region (Gagné et al.; Institute de recherche et de développement en agroenvironnement, 2009; Lajoie, 1960; Larocque, Marie et al., 2015).

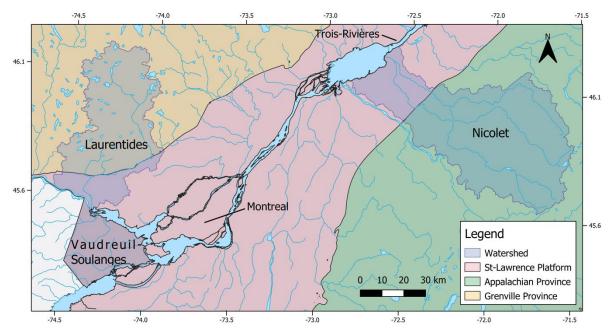


Figure 0.4: Location of the three selected watersheds (area#1: Laurentides, area #2 Nicolet, and area #3 Vaudreuil-Soulanges).

3.3.2.1 Area #1: Laurentides

The area of Laurentides has a mixture of forested areas and urban areas and is underlain by the hilly Grenville Province. It has not been covered by regional PACES studies at this time, and therefore a detailed description of the surficial deposits underlying the region are somewhat dated. Portions of the area along the Saint Lawrence and Ottawa Rivers were described by Lajoie (Lajoie, 1960). Bedrock in this area is overlain by till that varies in thickness from 7 to 12 m. Deposits of sand and gravel can also be found within this region, that were deposited by glacial rivers. Along existing rivers, including the Rivière-du-Nord, recent fluvial deposits composed mainly of loam are present (Institute de recherche et de développement en agroenvironnement, 2009).

Supplemental material containing the map of the quaternary geology and well distribution for each area is provided with this paper.

In this area, 31 municipalities (of a total of 58) rely on groundwater to produce drinking water from a total of 136 municipal wells and 91% (n=124) are located < 500 m from a surface water body. A total of 63 municipal wells (51%) are located at < 500 m from a lake, the remaining part (49%; n=61) being at < 500 m from a river. As shown in Figure 0.5, a significant number (31%; n=39) of municipal wells are found along the main river (i.e. Rivière-du-Nord). These wells are often serving municipalities with a population > 1000 people and are typically found in unconfined granular aguifers. There are three other rivers with wells in close proximity, namely, Red River with 4 wells serving two municipalities, Ottawa River with 4 wells serving three municipalities, Two Mountains Lake 12 wells serving one municipality, Achigan River with 2 wells serving one municipality. Further from the Rivière-du-Nord, the population generally decreases, and the wells are more frequently found in proximity to lakes and in fractured bedrock aquifers. The type of aquifer could be compiled for a total of 56 wells. Of these wells, 73% (n=41) are found in an unconfined granular aquifer, with the majority located < 100 m from a surface water body. Since the largest municipalities are more likely to be pumping sufficiently to induce a hydraulic gradient forcing the surface water to infiltrate the aquifer, the municipalities with the highest likelihood of performing IBF are the more populated ones in proximity to the Rivière-du-Nord. Meanwhile, wells located further from surface water and installed in fractured bedrock present a lower confidence level in the probability that IBF is taking place. It is important to note that the above-mentioned results include some wells outside of the selected watershed, as they shared a similar geological setting.

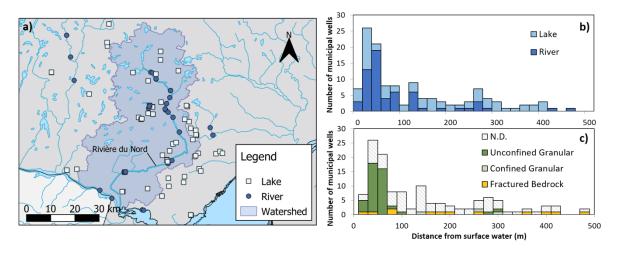


Figure 0.5: a) Spatial distribution of municipal wells located at less than 500 m from lakes and rivers in area #1 and distribution of wells according to b) the type of surface water and c) the type of aquifer.

3.3.2.2 Area #2: Nicolet

The Nicolet area straddles two geologic provinces, the Saint-Lawrence Platform in the lower altitude portion to the North and the Appalachian Orogen in the higher altitudes to the South. This area is principally covered by agricultural land. A total of 84 municipal wells are actively producing drinking water from groundwater resources to serve a total of 35 municipalities (of a total of 39). These municipalities, are generally smaller than those in other regions with many serving < 500 people. As illustrated by Figure 0.6, in area #2, a high number of wells are in the vicinity of different rivers and tributaries, similarly to area #1. In fact, a total of 48 municipal wells (57%) are in the 0-500 m range from a surface water body. Of these wells, most are located close to rivers (71%; n=34) and the remaining 29% (n=14) are located near lakes. However, the distance of these municipal wells varies more widely than in area #1. The Nicolet River and its tributaries are the rivers with the largest number of wells in close proximity. There are 5 rivers in the Nicolet river system with wells in close proximity. In addition, the Saint-Francois River has three wells in proximity serving 2 municipalities. Within this region, there are many more rivers whose quality could impact drinking water quality than in the other regions.

The Nicolet region has a distinctly different sequence of quaternary deposits when compared to the other two zones. As reported in the PACES study (Larocque, M et al., 2015), thick quaternary deposits exceeding 100 m in certain areas overly the bedrock in certain parts of the region. A

roughly 20 km wide band along the Saint-Lawrence River is underlain by a significant thickness of marine clays which can be partially or completely overlain by marine and lacustrine sands. The central portion of the basin located between elevations of 80 m and 120 m is dominated by aeolian and coastal sands, underlain by impermeable till around which peatlands can form. Thick glacio-fluvial sand and gravel deposits can lie directly on the bedrock in certain areas. Superficial deposit thicknesses at the municipal well locations calculated from the raster database that accompanies the PACES (Larocque, M et al., 2015) study ranged from < 5 m up to 30 m. Supplemental material containing the map of the quaternary geology and well distribution for each area is available.

Geological information could be compiled for a total of 26 municipal wells located < 500 m from a water body. In this region, 50% (n=13) of the municipal wells procure water from unconfined granular aquifers. Of these wells, 77% (n=10) are located < 200 m from a surface water body. These results suggest that there are favorable aquifers for pumping near rivers, while the regions more distant from rivers are less favorable. This region, although it consists mostly of smaller municipalities, has a high probability of IBF taking place, especially in wells within the first few hundreds of meters from a river. More information relative to the pumping rates for these wells, combined with a geochemical and isotopic approach would be needed to better estimate the potential of IBF in this region.

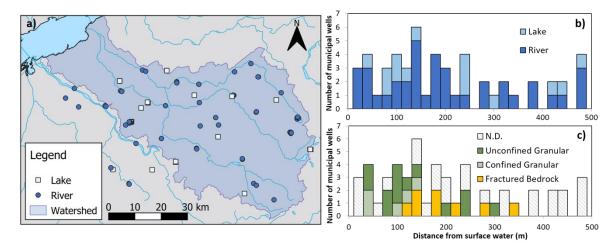


Figure 0.6: a) Spatial distribution of municipal wells in the Nicolet area that are located at less than 500 m from lakes and rivers in area #2 and distribution of wells according to b) the type of surface water and c) the type of aquifer.

3.3.2.3 Area #3: Vaudreuil-Soulanges

The area of Vaudreuil-Soulanges is located near Montreal and within the St-Lawrence Platform. The main types of land uses are agricultural and residential. This area, unlike the Nicolet area, is underlain solely by more recent quaternary deposits that were deposited uniquely during the last glacial cycle. The region is predominantly covered by marine clays (64% of the surface area). Only a small number of areas, along the Ottawa River and higher relief areas (i.e. till deposits, Mount Rigaud, "butte Saint-Lazare", and "butte de Hudson"), remain uncovered by these clays. Certain uncovered areas are composed of glacio-fluvial deposits that host productive granular aquifers. Examples of these deposits can be found along the Ottawa River in the northern portion of the zone and also in the Saint-Lazare region. Supplemental material containing the map of the quaternary geology and well distribution for each area is available.

In this area, 36 wells are in operation and produce drinking water from groundwater resources for 8 municipalities (of a total of 13) with populations typically > 1,000 people. As illustrated in Figure 0.7, the area contains a distribution of wells mostly in proximity to lakes. In fact, 66% (n=24) of the municipal wells are located at < 500 m from a lake. Among these wells, 92% (n=22) are located along a 6 km-long North-South transect near the region of Saint-Lazare. The PACES report highlights the presence of a thick and unconfined sandy deposit with a very productive aquifer in this zone (Larocque, M et al., 2015). Wells in proximity to rivers, for the most part, are located along the periphery of this zone near the more major water bodies. Another zone of concentrated wells (20%; n=10) is found in the region of Mount Rigaud, the majority of these wells are located in fractured rock or clay deposits (Larocque, M et al., 2015), making the probability of IBF unlikely. It can also be seen in Figure 0.7that there is a limited number of wells (33%; n=16) located within the first 200 m of a water body, which sharply contrasts with the other two regions. In this region, wells are located in proximity to two rivers only. The Viviry River with 4 wells serving one municipality, and the Saint Lawrence River with 4 wells serving one municipality are the two most prominent ones with some smaller streams making up the difference. The type of aquifer could be compiled for 22 municipal wells in area #3. From these wells, the majority is found in confined granular aquifers (31%; n=7) and fractured bedrock aquifers (40%; n=9). The fractured bedrock aquifers are known to be confined (Larocque, M et al., 2015), except in the regions with the highest concentrations of wells (i.e. St-Lazare, Hudson and Rigaud).

Overall, the greater distance from surface water in this area seems to indicate that IBF in most of the municipalities in this area is not likely. We identified only two municipalities with a total of 3 wells in close proximity to surface water. However, these wells are in confined granular or fractured bedrock aquifers and are thus presenting limited potential for IBF. Moreover, there is a strong possibility that the permeable areas of the Saint-Lazare region are influenced by infiltrating water from the spring thaw and precipitations.

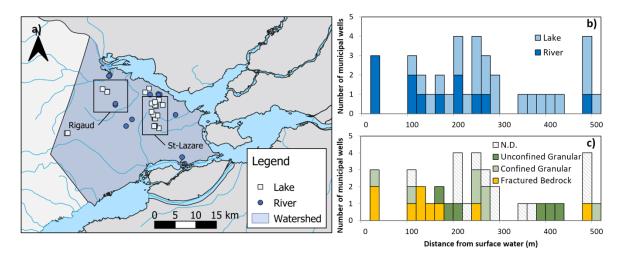


Figure 0.7: a) Spatial distribution of municipal wells located at less than 500 m from lakes and rivers in area #3 and distribution of wells according to b) the type of surface water and c) the type of aquifer.

3.4 Conclusions

This research has provided a reliable starting point for determining the impact of IBF on the population of Quebec. A simple process based on GISc was used by incorporating several open-source program QGIS, SAGA and R and openly available data. The initial use of distance from a surface water body as well as additional information extracted from the government database revealed that nearly one million people in the province of Quebec are supplied drinking water via wells in close proximity (i.e. < 500 m) to a river or a lake. This first overview has also demonstrated that there is a high degree of regional variability with regards to the probability that IBF is being performed. One area has the greatest potential use of IBF (area #1: Laurentides), a second one requires more information to draw precise conclusions (area #2: Nicolet) and the third one has the lowest probability of IBF (area #3: Vaudreuil-Soulanges).

There are a number of shortcomings that are evident in the current development, management and understanding of hybrid groundwater surface water extraction sites in the province of Quebec. The current regulations do not contain general insights into the protection or management of these sites. The existing GWUDI classification is not designed to assess risk at an IBF site as it does not expressly consider the contribution of surface water to a pumping well, and only under certain circumstances are IBF wells considered GWUDI. With mounting pressures on our water resources from climate change and population growth, we hope that this work demonstrates the need to better understand and improve regulatory framework to specifically address hybrid groundwater-surface systems such as IBF in order anticipate problems common to IBF sites and ensure that our water resources are well protected and exploited sustainably.

It will be important to precisely develop guidelines for determining what municipalities should be targeted within future regulations. The policy changes would require the definition of a new category of wells for IBF sites, that requires short, medium, and long-term assessment of risk related to the permanent or intermittent contribution of surface water bodies to wells.

In order to determine more precisely which wells are using IBF, a possible approach would be based on a dedicated sampling program for specific dissolved tracers. These tracers would be used to quantify the spatio-temporal evolution of transit times (e.g. stable isotopes of water) and relative proportions of groundwater and surface water (e.g. electrical conductivity). A longer characterization period would allow for minimization of unforeseen costs over the life of the water treatment plant.

CHAPTER 4 222RN AS A TIME TRACER OF SURFACE WATER INFILTRATION: SENSITIVITY TO GEOCHEMICAL END-MEMBERS SELECTION

4.1 Abstract

This study aims to incorporate the lessons learned from various studies into a more comprehensive understanding of the conditions when the ²²²Rn method is an effective method of evaluating residence times, and to test the impact of endmember choice on the calculations of residence time. Whereas the previous studies were typically investigating a single flow line under steady state flow conditions, this study was undertaken to evaluate the use of ²²²Rn as a tracer for calculating residence times in a multi-well bank filtration system under variable pumping and hydraulic conditions. On this site, the close proximity of pumping wells to surface water, and the residence times estimated in previous studies (2-7 days) made it a favourable candidate for use of the previously established method of residence time calculations using ²²²Rn. It was determined that the proportion of groundwater strongly impacts calculated residence times. This was particularly evident when the proportion of surface water was low (<50%) and the resulting residence times was typically short (<7 days). Additionally, aquifers with stratification of either ²²²Rn production or EC are not favorable for use of this method for determining residence times as there are portions of water at equilibrium that cannot be identified using this method. Despite this method being considered well developed by some authors, additional work should be conducted to establish the exact hydrogeological conditions that are required for this approach to provide representative results. A clear stable radon end-member, high confidence in the representativity of samples and simple binary mixing are all essential for this. Several steps are recommended in order to improve this method and determine when and under what conditions it is appropriate. Also, some general guidelines to consider when selecting an end-member sampling location were outlined.

4.2 Introduction

Bank filtration is a widely used technique for pre-treatment of drinking water throughout Europe (e.g. Dillon et al., 2019; Hiscock & Grischek, 2002) and to a lesser extent in Canada (Patenaude et al., 2020). This process involves installation of wells in close proximity to surface water. The

surface water is drawn through the banks of a water body toward the pumping wells either by a natural (e.g. Baudron et al., 2016) or artificial gradient (Masse-Dufresne et al., 2019). The infiltrating water rapidly recharges the aquifer and benefits from natural attenuation of contaminants that are often present in surface water (Masse-Dufresne et al., 2019; Tufenkji et al., 2002). The efficiency of bank filtration is affected by a variety of factors such as the reactivity of the substrate, chemical composition, grain size, hydraulic conductivity and residence times of infiltrating water (Ray, 2002). These factors affect the capacity of the substrate to attenuate contaminants. Residence times, which are potentially affected by seasonal variability of surface water levels, are an important factor in the removal of contaminants (Kuehn & Mueller, 2000) and residence times can have an impact on the removal of bacteria, turbidity (Sahu et al., 2019) and organic micropollutants (Oberleitner et al., 2020).

There exists a number of tools to investigate the residence times of infiltrating groundwater. Physically based models calibrated using hydraulic head measurements have been used to model SW-GW systems however they cannot reliably reproduce mixing ratio or residence times in BF systems (Schilling 2017). For this reason, various chemical and physico-chemical tracers also have been investigated either as environmental tracers or as tools to constrain flow models. The various parameters that have been used in studies of residence times in bank filtration sites include: temperature (Doussan et al., 1994; Wang, W.-s. et al., 2020), 3 H/ 3 He (Massmann, G. et al., 2008; Massmann, Gudrun et al., 2009), time series of EC (Cirpka et al., 2007; Vogt et al., 2010), time series of stable isotopes of water (Massmann, G. et al., 2008), 37 Ar (Schilling et al., 2017) each of which provides information on a certain temporal gap. 222 Rn has also been used in a variety of studies to determine residence times of infiltrating surface water (Bertin & Bourg, 1994; Close et al., 2014; Gilfedder et al., 2019; Hoehn & von Gunten, 1989; Stellato et al., 2013).

Calculations of the residence times of infiltrating surface water are achieved by determining the emanation rate of the sediments. Since secular equilibrium with the aquifer occurs after roughly 5 half-lives, the resolution of the method is considered to be between 10 (Schilling et al., 2017) and 21 days (Gilfedder et al., 2019) in most studies. Therefore, measuring radon activity in the aquifer at a location that would have residence times greater than 21 days can form the basis of emanation rate calculation.

Throughout the research conducted on the use of ²²²Rn as a tracer in groundwater, no standard procedure has been found for determining a representative groundwater end-member. It is typically based on a measurement at a significant enough distance along the investigated flow line to have theoretically achieved equilibrium. Hoehn and von Gunten (1989) used a value measured in a well near the surface that a 20% higher value observed at the same distance in a deeper well. The steady state values used by Bertin and Bourg (1994) and Macheleidt et al. (2006) were measured in neighbouring hills and in a remote borehole respectively. Stellato et al. (2013) chose a value from a well which was not significantly influenced by the infiltrating river water, even though it had lower values than one situated on a similar flow path. Other methods have also been used, for example, Close et al. (2014) determined the end-member by fitting all radon data to the decay equation, which was transformed into a distance relationship. Other studies have used laboratory methods such as Cranswick et al. (2014) where equilibrium values were determined by leaving samples in jars for two months with no overlying air, and then measuring the activity in the pore water using a mini piezometer. Other studies, Dawe and MacQuarrie (2005) and Kobierska et al. (2015) simply took the highest measured values in the vicinity of the study area. As can be seen quite clearly, no standard procedure is used for determining the groundwater end-member, although it has an enormous impact on the residence time calculations.

Determining the proportion of the radon measured in the pumped water that is contributed by infiltrated surface water requires a calculation of mixing ratios. A single value is typically used in studies that incorporate mixing (Bertin & Bourg, 1994; Stellato et al., 2013). In more complex sites where both surface and groundwater end-members may fluctuate, and flow lines are more uncertain, the end-member choice is less evident, and can impact the calculated residence times. EC was chosen for mixing ratio calculations as it was considered a conservative tracer in the context of the study site where transit times are relatively short. A graph comparing EC to Cl concentrations is presented in Appendix B. Chloride is considered a conservative tracer of groundwater (Bertin & Bourg, 1994; Turner & Townley, 2006), and since the relationship between these two parameters (EC and Cl⁻) is linear, it can be considered that EC is also a conservative tracer for the purposes of this study. EC variability was observed for each of the end-members over the course of the sampling period on the study site is related to recharge events following the spring thaw, which can take several months to return to normal levels. The variability of end-members was considered in one of the mixing ratio calculation scenarios.

Finally, transience is rarely considered when using ²²²Rn to calculate residence times during BF. Studies that calculate residence times typically investigate a single flow line under steady state flow conditions. Recently, Gilfedder et al. (2019) compared several methods and found underestimates of the apparent ages using ²²²Rn when infiltrating rainwater mixes with regional groundwater along the advective front of a floodwater pulse and the radon activities increase above ingrowth alone.

This study was undertaken to evaluate the use of ²²²Rn as a tracer for residence times in a multi-well bank filtration system under variable pumping and hydraulic conditions. It was decided to test several seemingly logical end-member choices similar to those used in other studies. The close proximity of pumping wells to surface water at one bank filtration site in the province of Quebec, Canada, the short estimated residence times (2 to 7 days), and the expected high (90%) proportion of lake water estimated by the consulting firm AGEOS (Richard et al., 2009), made the study site a favourable candidate for testing the previously established method of residence time calculations using ²²²Rn. The groundwater endmembers were varied through time (EC) and space (²²²Rn). By considering many possible scenarios the objective was to evaluate the impact of these choices on the interpreted residence times. These various scenarios could then be used to discuss the important factors to consider during endmember selection and provide some guidelines for further studies. This site also provided an opportunity to evaluate whether the ²²²Rn method is effective on sites with intermittent pumping.

4.3 Site description

4.3.1 Hydrology

The study site is located on the shores of and strongly influenced by the adjacent Lac-des-Deux-Montagnes (LDDM; Figure 4.1). The LDDM is a large lake that is located at the outlet of the Ottawa River, where it joins the Saint-Lawrence river (Figure 4.1). The water level in the lake varies significantly throughout the year, as does the distance between the wells and the lake. Since 1986, the lake has reached a maximum elevation of 24.77 m and a minimum of 21.19 m, with an overall average elevation of 22.1 m (Centre d'expertise hydrique du Québec, 2019). Seasonal variations occur in the LDDM with water levels in the lake increasing during the period from the

end of March to the end of June reaching seasonal maximums during this period. Typically, there are two peaks in the water level of the LDDM, one in April, and one in May, which correspond to thaw in the lower and upper portion of the watershed respectively (Parc National d'Oka, 2003). In the spring of 2018, an average peak in the water level in the LDDM occurred, whereas in 2019 extreme levels of flooding were observed. During the sample period, the water level in the lake reached levels of 24.75 m, which is near the maximum observed since 1986 (24.77) (Figure 4.2). During the study period, the level in LDDM fluctuated by more than 3 m, and in the piezometer, manual measurements fluctuated by roughly 2 m (manual measurements), over 3 m during the period with automatic measurements, and over 3.5 m with combined manual and automatic measurements.

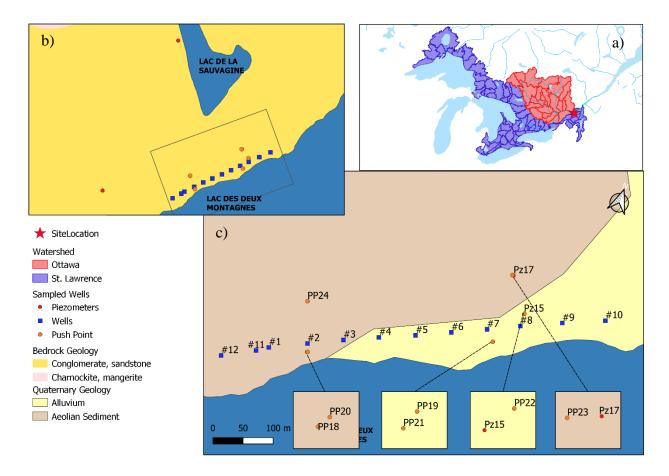


Figure 4.1: Study Site a) Location of the study site and drainage basins that feed the LDDM; b) all sample locations with bedrock geology; c) quaternary units surrounding the study site and setup of wells and observation wells.

4.3.2 Hydrogeology

The area is underlain by the Potsdam group, however there are no outcrops within the area of the park (Parc National d'Oka, 2003). Overlying these sedimentary rocks, the Champlain Sea clays are present. Above the clay deposits, there is a sand deposit of varied thickness (Parc National d'Oka, 2003; Richard & Benhouhou, 2001). The pumping wells are located within the sandy aquifer in close proximity to a sandy beach. These sandy deposits were exploited during the 1950s. To the north, the Lac de la Sauvagine was exploited during various phases and the area directly in front of the well field in the Lac des Deux Montagnes has also been dredged for sand (Parc National d'Oka, 2003). In the area of the well field, the sand deposit reaches a thickness of up to 12 meters. The sand unit in the pumping wells is described as fine to medium sand, and the hydraulic conductivity estimated at between 1.0 and 1.4 x 10⁻² cm/s (Richard et al., 2009). Regional quaternary maps indicate that the origin of the sandy aquifer is alluvial or aeolian. The regional scale quaternary mapping (MERN, 2018) indicate a potential geological contact in proximity to the well field (Figure 4.1) however there is no indication of a distinct change in geology or grain size in the well logs. The aquifer is therefore considered to be homogenous.

Studies have also been conducted by consulting firms prior to the construction of the site and in order to expand the overall capacity of the site. Studies were conducted by (Richard & Benhouhou, 2001, 2012; Richard et al., 2004, 2009) and Qualilab (Gauthier, 2003). The initial report by AGÉOS in 2001 (Richard & Benhouhou, 2001) was conducted on a regional scale in order to compile data and evaluate the entire region based on historical work, and four exploratory drill hole locations were proposed near the Lac de la Sauvagine and Lac des Deux Montagnes. These exploratory holes were drilled and a part two of the report (Richard & Benhouhou, 2001) was produced which contained information on pumping tests conducted on these wells and also much more detail on the uppermost granular aquifer, and its continuity. Through their exploratory drilling and the drilling of the production wells the continuity of the surface sand aquifer was confirmed, and the variable thickness was also determined, with a narrowing of the aquifer resulting from a change in elevation of the clay unit toward the west which subdivides the unconfined aquifer into a western and eastern basin. The narrowing was observed in Pz25 the westernmost piezometer in Figure 4.1b (Piezometer near the old treatment plant). Additionally, their regional work revealed a greater thickness of the aquifer in the region surrounding the Lac de la Sauvagine. Chemically, during initial characterisation, all the test wells displayed elevated Fe and Mn concentrations, and the concentration decreases from the center of the park to the north, toward the LDDM. There were also higher concentrations measured in LDDM which is interpreted as to be the influence of the deeper rock aquifer, but this is could be related to discharge of groundwater into the lake under normal conditions. During the 24-hour pumping test at well StJo-2002-09 was the only well with low Fe Mn concentrations, the concentrations were high at the time of construction but decreased after the 24h pumping test. This, was interpreted as potential dilution of GW with SW, however the high concentrations in SW do not agree with this interpretation, and rather must be related to changing redox conditions caused by pumping.

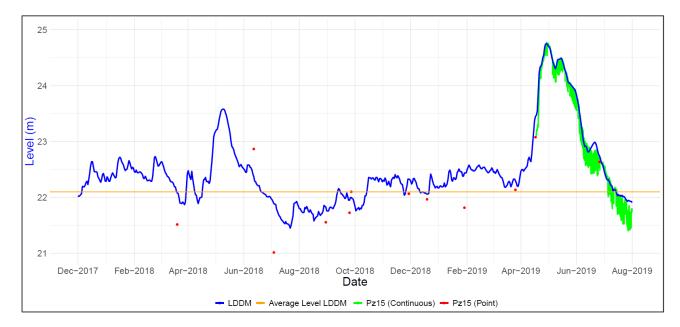


Figure 4.2: Water levels in LDDM (Blue), Pz15 level logger (Green), Pz15 point measurements (red), calculated average value for LDDM since 1986 (orange). LDDM data from (Centre d'expertise hydrique du Québec, 2019)

4.3.3 Pumping wells

Pumping rates vary greatly from season to season, with most wells experiencing an increase in hours pumped during the summer months and a decrease in the winter (Table 4.1). The frequently pumping wells are an exception to this rule with the hours pumped rarely dropping below 500 hours per month.

The wells were not equipped with flow meters, and only the pumping time is recorded at each well. Of the 10 wells, 2 are equipped with variable pumps, and the remaining 8 wells have fixed flow

rate pumps. The two wells that can pump at variable rate are the highest priority wells. For the vast majority of the sample program, the two highest priority well were #2 and #7, however they each had brief periods where technical problems required shutdown of the pumps. Since these wells are high priority but equipped with variable pumps, the drawdown at these wells does not stabilize. This can be seen in time series of water level in the wells, which are not presented in this manuscript since these data were not able to be converted to a useable format. Although these wells have high pumping hours, the rate of flow can change and the flow velocities and directions can change even during times when the pump is on. The remaining wells are activated on demand, and generally pump less than 150 hours per month which works out to an average of roughly 5 hours per day or less (Table 4.1). Pumping rates are highest in the summer months from June to July and July 2018 saw the highest number of combined hours pumped.

Table 4.1: Hours pumped per month for each well.

| Well | P#1 | P#2 | P#3 | P#4 | P#5 | P#6 | P#7 | #8 | P#9 | P#10 | P#11 | P#12 | Total |
|----------|-----|-----|-----|-----|-----|-----|-----|-----|-----|------|------|------|-------|
| Nov2017 | 68 | 685 | 61 | 29 | 62 | 71 | 686 | 49 | 33 | 30 | 0 | 0 | 1774 |
| Dec 2017 | 85 | 704 | 57 | 60 | 89 | 66 | 704 | 33 | 29 | 46 | 0 | 0 | 1873 |
| Jan2018 | 85 | 711 | 63 | 41 | 85 | 56 | 710 | 34 | 50 | 51 | 0 | 0 | 1886 |
| Feb2018 | 72 | 623 | 48 | 26 | 45 | 54 | 646 | 44 | 33 | 42 | 0 | 0 | 1633 |
| Mar2018 | 0 | 727 | 71 | 63 | 41 | 43 | 708 | 112 | 43 | 39 | 0 | 0 | 1847 |
| Apr2018 | 0 | 680 | 86 | 55 | 0 | 114 | 680 | 95 | 63 | 48 | 12 | 45 | 1878 |
| May2018 | 0 | 702 | 109 | 128 | 0 | 120 | 692 | 132 | 114 | 90 | 94 | 143 | 2324 |
| Jn2018 | 0 | 691 | 142 | 131 | 0 | 117 | 691 | 142 | 156 | 158 | 150 | 110 | 2488 |
| J12018 | 0 | 708 | 179 | 222 | 0 | 253 | 710 | 250 | 273 | 265 | 178 | 259 | 3297 |
| Aug2018 | 0 | 678 | 153 | 112 | 0 | 169 | 707 | 158 | 159 | 218 | 227 | 170 | 2751 |
| Sep2018 | 0 | 682 | 91 | 42 | 0 | 145 | 686 | 124 | 85 | 97 | 64 | 117 | 2133 |
| Oct2018 | 0 | 552 | 39 | 38 | 0 | 15 | 553 | 22 | 18 | 9 | 21 | 27 | 1294 |
| Nov2018 | 0 | 714 | 22 | 25 | 0 | 155 | 671 | 96 | 60 | 34 | 46 | 38 | 1861 |
| Dec2018 | 0 | 730 | 41 | 31 | 0 | 76 | 567 | 46 | 28 | 58 | 32 | 22 | 1631 |
| Jan2019 | 0 | 735 | 47 | 20 | 0 | 51 | 551 | 50 | 22 | 38 | 35 | 40 | 1589 |
| Feb2019 | 0 | 647 | 39 | 49 | 0 | 31 | 503 | 24 | 32 | 45 | 46 | 32 | 1448 |
| Mar2019 | 0 | 685 | 53 | 45 | 0 | 80 | 676 | 74 | 48 | 48 | 45 | 49 | 1803 |
| Apr2019 | 0 | 711 | 92 | 60 | 0 | 61 | 677 | 11 | 32 | 47 | 84 | 52 | 1827 |
| May2019 | 0 | 719 | 94 | 15 | 0 | 120 | 719 | 106 | 65 | 85 | 24 | 65 | 2012 |
| Jn2019 | 0 | 705 | 192 | 171 | 0 | 276 | 219 | 201 | 203 | 247 | 0 | 219 | 2433 |
| J12019 | 0 | 723 | 171 | 193 | 0 | 262 | 717 | 174 | 3 | 171 | 108 | 179 | 2701 |
| Aug2019 | 0 | 723 | 152 | 204 | 0 | 227 | 631 | 148 | 0 | 174 | 146 | 155 | 2560 |
| Average | 93 | 676 | 92 | 80 | 84 | 114 | 624 | 103 | 81 | 96 | 77 | 155 | 2221 |

4.3.4 Transient flow regime

The water level in Pz15, an observation well located inland of the site, fluctuates from slightly greater than the level of LDDM and frequently drops below the level of LDDM when pumping occurs (Figure 4.2). Since the levels are often quite close, and are strongly affected by pumping, it appears that the flow direction and velocity change frequently. The transient pumping and flow conditions lead to extremely complex interactions between surface water and groundwater in this area. It is therefore hard to predict the flow direction at any given moment with traditional hydrogeological tools. This was one of the motivating factors in developing a sampling program using environmental tracers, specifically Rn and EC, to better understand the flow conditions and residence times of infiltrated water.

4.4 Methodology

4.4.1 Groundwater and Surface Water sampling

Samples were collected from December 2017 to August 2019 on a monthly basis, whenever possible. These samples were collected during periods with various hydraulic conditions including before and after a major flooding period in 2019, however no samples of ²²²Rn could be taken during flooding as the site was completely submerged. During each sampling campaign, all active wells were sampled as well as the adjacent lake and the inland piezometers Pz15 and 17. A series of other observation wells were also installed throughout the study area; however they were not sampled as rigorously (Figure 4.1). These observation wells, PP18, 19 20, 21, 22, 23, 24 were installed by hand at various depths and various locations and were sampled a few times. PP18 and PP20 were installed at the same location at depths of 10 and 5 m respectively. These piezometers are located between Lac des Deux Montagnes and well #2 and were designed to intercept infiltrating lake water. PP19 and 21 serve a similar purpose and are located between well #7 and the LDDM. PP22 and 23 were installed next to Pz15 and Pz17 at depths of 5 m and were used to evaluate stratification of the aquifer. Finally, PP24 was installed inland of well #2 to determine if there is some spatial variability of the groundwater chemistry.

Larger diameter piezometers were sampled with a 12V-Typhoon, Proactive Environmental Products, Bradenton, FL, USA, and smaller piezometers were sampled with a Masterflex E/S Portable Sampler peristaltic pump. EC was measured with YSI Pro Plus 6051030 and Pro Series

pH/ORP/ISE and Conductivity Field Cable 6051030-1, YSI Incorporated, Yellow Springs, OH, USA and is accurate to 1% of the measurement. Samples were taken after stability of physicochemical parameters (pH, electrical conductivity, temperature, redox potential) was achieved for pumping wells and after three times the volume of the piezometer was purged for piezometers. Samples for ²²²Rn were collected in 2L Coca-Cola bottles. Samples for EC were collected in 50 mL low-density polyethylene (LDPE) containers and filtered at 0.45 µm, as described in Masse-Dufresne et al. (2020b). Lake water was sampled by wading into the lake to a depth of roughly 30-50 cm filling sample bottles by submerging them in the lake. During winter, when the lake was frozen to a safe thickness, an ice auger was used to drill a hole through the ice and bottles were filled by submerging them in the hole. Radon activity is often less stable than EC (Baskaran, 2016) meaning that stability of EC may not mean stability of EC as an indicator that a sufficient purge volume had been achieved.

4.4.2 Analytical techniques

Radon activities were analysed using a Durridge RAD7 Radon Detector using the Big Bottle System. Groundwater samples were analysed for a total of 100 minutes (20 cycles of 5 minutes) with counting cycles of 5 minutes using the Normal counting mode. Lake water was sampled using the Sniff protocol for a total of 80 5-minute cycles (6h40m analysis time). The error on the radon measurements for pumping wells is generally between 3 and 5% (average error of all samples: 237 Bq/m³). it increases to greater than 20% in surface water samples where activities are much lower. Duplicate samples were taken throughout the sampling campaign. These duplicates had similar values (within analytical error) but on one occasion, there was a 1250 Bq/m³ difference (November 29, 2018). The sample with the lower activity was analyzed roughly 24 hours later and some radon gas may have escaped.

4.4.3 Quality control

A quality control and quality assurance (QA/QC) was not systematically incorporated into the sample program. The QA/QC was implemented only as duplicate samples (7 total samples), and no blanks or standards were used. Most of these duplicates had similar values but on two occasions, there was a 1250 Bq/m³ (November 29, 2018) and 1500 Bq/m³ (January 29, 2019) difference

between duplicate samples. On one occasion, it was the sample that was analysed first, and on the other it was the sample that was analysed second. On both occasions, the actual measured values were similar, and only the corrected values showed a significant difference. The reason for this difference in values could not be explained.

4.4.4 Residence time of infiltrating groundwater

4.4.4.1 Residence time calculation

The radon method described initially by Hoehn and von Gunten (1989) forms the basis of this study with the incorporated use of mixing ratios proposed by Bertin and Bourg (1994). This methodology is relatively simple in its application and involves establishing a few basic underlying assumptions for the site initially. This method was applied to all of the endmember options of both mixing ratios (Scenarios A, B, and C) and of radon equilibrium activity (Scenarios 1, 2, 3, 4) for a total of 12 different residence time calculations. The assumption that forms the basis of this equation is that regional groundwater is at equilibrium with the substrate, meaning groundwater flowing from the Lac de la Sauvagine toward Lac des Deux Montagnes takes longer than 5 times the half-life of 222 Rn (Bertin & Bourg, 1994).

The activity of radon at any given time is described by the radioactive decay equation, where λ is the decay constant equal to $\ln(2)/T_{1/2}$:

$$Rn(t) = Rn_e(1 - e^{-\lambda t})$$

Residence times can be given by solving for t.

$$Rn(t) = Rn_e - Rn_e e^{-\lambda t}$$
 $Rn_e - Rn(t) = Rn_e e^{-\lambda t}$

$$\frac{Rn_e - Rn(t)}{Rn_e} = e^{-\lambda t}$$
 $ln\left(\frac{Rn_e - Rn(t)}{Rn_e}\right) = -\lambda t$
 $ln(Rn_e - Rn(t)) - ln(Rn_e) = -\lambda t$

$$\frac{1}{-\lambda} \left(ln \left(Rn_e - Rn(t) \right) - ln \left(Rn_e \right) \right) = t$$

$$\frac{1}{\lambda} \left(-ln \left(Rn_e - Rn(t) \right) + ln \left(Rn_e \right) \right) = t$$

$$t = \frac{1}{\lambda} ln \left(\frac{Rn_e}{Rn_e - Rn(t)} \right)$$

Which can be re-written, with Rn_{GW} accounting for the Rn_e value at equilibrium with the matrix in the regional groundwater, Rn_{inf} accounting for the Rn(t) value of the infiltrating surface water and by adding the Rn_{sw} term to the numerator to account for the non-zero activity of infiltrating surface water, as illustrated by Figure 4.3:

$$t = \frac{1}{\lambda} * \ln \left(\frac{Rn_{GW} - Rn_{SW}}{Rn_{GW} - Rn_{inf}} \right)$$

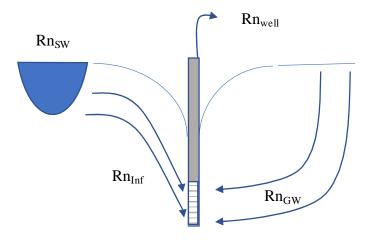


Figure 4.3: Schematic of the radon values used in the calculation of residence times.

In order to determine the Rn value that corresponds to Rn_{inf} out of the measured Rn_{well} value from the pumping wells, a mixing ratio calculation must be added to separate the activity that is related to groundwater from the activity of radon that is associated with infiltrating surface water. The proportion of infiltrating surface water X_{inf} can be determined using a conservative tracer, in the case of this study, EC was considered conservative as it correlated well with Cl (Appendix B), and applying a simple binary linear mixing model that follows the following formula:

$$X_{inf} = \frac{(EC_{Well} - EC_{GW})}{(EC_{SW} - EC_{GW})}$$

Once the proportion of surface water and groundwater has been determined, this value can be used to correct the measured radon and attribute a radon activity to the infiltrating surface water. The correction is applied using the following formula:

$$Rn_{well} = Rn_{inf} \bullet X_{inf} + Rn_{GW} \bullet (1 - X_{inf})$$

Which can be rearranged to isolate for Rn_{inf} which gives the activity that is attributed to infiltrating surface water.

$$Rn_{inf} \bullet X_{inf} = Rn_{well} - Rn_{GW} \bullet (1 - X_{inf})$$

$$Rn_{inf} = \frac{Rn_{well} - Rn_{GW} \bullet (1 - X_{inf})}{X_{inf}}$$

Making the substitutions into the residence time calculation, completed equation becomes:

$$t = \frac{1}{\lambda} * \ln \left(\frac{Rn_{GW} - Rn_{SW}}{Rn_{GW} - \frac{Rn_{well} - Rn_{GW} \bullet (1 - X_{inf})}{X_{inf}}} \right)$$

4.4.4.1 End-members assessment

As illustrated by Table 4.2, geochemical end-members used to used to define the inland regional groundwater were taken from:

- P15: Observation well on the inland side of the well field, with screen location representative for the deep flow line (>10m, unknown depth, unknown presence of a filtering sand). For EC, its monthly, average and max values for EC (named A, B and C, respectively). For ²²²Rn, its average and maximum values (named 1 and 2, respectively) were used. P17 was not used due to a limited number of samples and since its values were close to PZ15, as detailed in the results section.
- Pumping wells: For each one, calculations were made based on the maximum ²²²Rn value measured (named 3).

- PZ22: Observation well on the inland side of the well field, with screen location representative for the shallow flow line (5 m). Its maximum ²²²Rn value (named 4) was used.

The value for surface water end-member was based on measurements in the adjacent Lac des Deux Montagnes. The ²²²Rn end member for surface water, although often considered zero (0), as activity rapidly dissipates in surface water, was considered to be higher in this study as there are portions of the surface water body where groundwater is expected to discharge to surface water. The infiltrating water might then already have started to equilibrate with the soil prior to infiltration. The average and minimum ²²²Rn values measured in the lake were used for the calculations, as for EC. A temporally variable value for the infiltrating surface water was considered but was ultimately not used for any of the final calculations since it was deemed to introduce considerable errors to the values calculated during the periods with elevated ²²²Rn in surface water.

4.4.4.2 Selecting plausible scenarios

The reader is referred to Table 4.2 to visualize the different scenarios tested in this study. Mainly, three EC scenarios were considered for the calculation of mixing ratios. The first mixing ratio (used in scenarios A1 to A4) was calculated using the average EC value at PZ15 (495 µS/cm Table 4.2) and the average value measured in the LDDM. The second mixing ratio (scenarios B1 to B4) used a temporally variable value measured each month at PZ15 (394-567 µS/cm) (when not available, the average value was used) and the minimum value measured in LDDM. The third mixing ratio (scenarios C1 to C4) was calculated using the maximum value measured at PZ 15 and the minimum value measured in LDDM. Regarding the surface water end member, the monthly ²²²Rn value for the LDDM was not considered an appropriate end member choice as, on occasion, surface water values were quite high and using them could lead to an underestimate of residence times.

Table 4.2: Summary of all mixing ratio and radon scenarios tested.

| | Groundwater | | Surface water | |
|----------|-------------|-------------------------------------|---------------|-------------------------------------|
| Scenario | EC (μS/cm) | ²²² Rn Bq/m ³ | EC (μS/cm) | ²²² Rn Bq/m ³ |
| A1 | 495 | 5536 | 76 | 376 |
| A2 | 495 | 6680 | 76 | 19 |
| A3 | 495 | Variable (6231-7721) | 76 | 19 |
| A4 | 495 | 8263 | 76 | 19 |

| B1 | Variable (394-567) | 5536 | Variable (42-121) | 376 |
|----|--------------------|----------------------|-------------------|-----|
| B2 | Variable (394-567) | 6680 | Variable (42-121) | 19 |
| В3 | Variable (394-567) | Variable (6231-7721) | Variable (42-121) | 19 |
| B4 | Variable (394-567) | 8263 | Variable (42-121) | 19 |
| C1 | 567 | 5536 | 42 | 376 |
| C2 | 567 | 6680 | 42 | 19 |
| C3 | 567 | Variable (6231-7721) | 42 | 19 |
| C4 | 567 | 8263 | 42 | 19 |

4.5 Results

4.5.1 Electric conductivity

Several trends appear in the spatial distribution of electrical conductivities of groundwater throughout the pumping wells (Figure 4.4). Overall, with the exception of the wells on either extremity of the well field which have high EC, the wells on the eastern side of the well field (wells P7 through P10) have higher EC (~230 μ S/cm to 410 μ S/cm) than the western end (P11 to P6;110 μ S/cm to 300 μ S/cm). The Maximum EC values at the wells were measured at different moments throughout the sample campaign. The majority of the wells on the eastern side experienced their lowest EC values in the fall 2018 period (Appendix A). The minimum values are spread over a few months at wells #3, #4, #6. #11 where lower values occurred during fall-winter of 2018-2019.

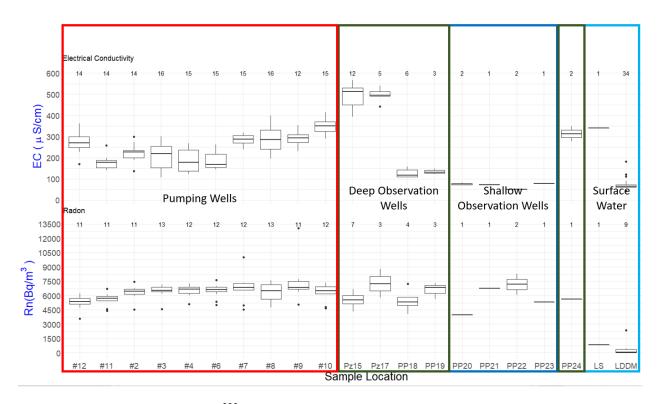


Figure 4.4: Boxplot of EC and ²²²Rn data for all sampling points based on measurements made throughout the entire sample program. The number of data points used to generate the boxplots are indicated above each boxplot and range from 1 point up to 16 data points

The EC of water sampled at all shallower (5m) push-point piezometers (PP20 to PP23) on both the inland and lake side of the well field was similar to lake water (average of all samples = $66 \mu \text{S/cm}$)

and was lower than the deeper piezometers and the pumping wells. The deeper push points located between the well field and the lake, PP18 and PP19, had lower EC (average 128 μ S/cm) than most of the wells, with the exception of some of the lowest values measured in the wells on the eastern side of the well field.

Throughout the entirety of the sample program, EC remained relatively stable in LDDM and Pz15 which were selected as the surface water and groundwater end-member locations respectively. As they were able to encompass all measurements at the wells, Pz15 and LDDM were selected as the two end-members for mixing ratio calculations. LDDM had EC ranging from 41 to 182 μ S/cm and an average of 71 μ S/cm. Pz17 and Pz15 were expected to be intercepting regional groundwater that is flowing towards the LDDM, since they are the deepest groundwater observation wells and located inland from the pumping wells. Pz15 (394-567 μ S/cm) and Pz17 (442-539 μ S/cm) had a similar range of measured EC values, although Pz17 was sampled less frequently and appeared to be more variable from month to month. Groundwater from Pz15 was consistently more mineralized than at the pumping wells throughout most of the sample campaign, with the exception of a drop in EC values during and following the flooding event of 2019. Another potential groundwater end-member location, Pz24, is located inland of the wells on the western side of the well field however its two samples followed the flooding event of 2019 and had significantly lower EC (275-350 μ S/cm) than the average values at Pz15 and Pz17.

4.5.2 Radon activities

 222 Rn activities were measured at all of the pumping wells. Overall, the median radon activity tends to increase from west to east similarly to EC. With the exception of wells #12 and #11 having lower median activities, similar median values were found, in the range of 6400 to 6800 Bq/m³, as illustrated by Figure 4.4. In other words, with the exception of a few outliers, roughly half of all measurements at each well lie within a range of <1000 Bq/m³. All but one to two values at each well fall within roughly 1000 Bq/m³ of the median value.

The maximum values at each well occurred at different moments of the year with no more than two wells having maximum measured values at the same moment in time (Appendix A). Wells # 6 (7633 Bq/m³) and #7 (9996 Bq/m³) both had a spike during the same sampling campaign in the month of July 2018. A spike in radon was also observed at well #9 (13069 Bq/m³) in December 2018. The minimum values unlike the maximum values were focused at a single point in time

(Appendix A). All wells experienced either their lowest or second lowest value in the month of August 2018, from 3596 Bq/m³ at well #12 to 5303 Bq/m³ at well #8. Wells #2 (4550 Bq/m³), #7 (4528 Bq/m³), and #8 (4732 Bq/m³) had their overall minimum value in the first sample campaign in December 2017. Two other wells, #6 (5014 Bq/m³) and #11 (4419 Bq/m³) had their minimum values in November 2018.

The push-point observation wells had variable radon activities. The deeper push points in between the lake and the wells PP18 and PP19 had different tendencies. PP18 had a mean activity of 5472 Bq/m³ and varied by roughly 3000 Bq/m³. PP19 had a higher mean activity at around 6800 Bq/m³ and varied by about 1600 Bq/m³ These deeper push points had similar activities to those seen in the pumping wells. The shallower push points between the lake and the wells PP20 and PP21 were only sampled on one occasion each and had either similar the median values of most pumping wells during regular times (PP21= 6700 Bq/m³) or slightly lower than the minimum value seen in the wells (PP20 = 3986 Bq/m³).

The activity at the deep inland groundwater were somewhat inconsistent and ranged from 4308-6680 Bq/m³ at Pz15, to 5779-8788 Bq/m³ at Pz17. At shallower depth (5m) PP22 featured activities (8250 and 6000 Bq/m³) equal to or higher than those measured at depth in Pz15; while PP23 had higher measured activity than Pz17. At the one location on the western end of the wellfield (PP24), the activity was only measured once and was 5643 Bq/m³.

The LDDM has values that typically are less than 100 Bq/m³. These low values are expected for surface waters, however there were some outliers at 435 and 2000 Bq/m³

4.5.3 Mixing ratios

The evolution of mixing ratios at the pumping wells using the three mixing ratio scenarios are shown in Figure 4.5. Calculated mixing ratios at each well are quite variable over time and the range of values is different. The various EC endmembers used for calculating the mixing ratio do not have large impact on the results and do not typically vary more than 15% from one endmember choice to another. The variation for each well is a reflection of the measured EC values.

Wells on the western end of the well field can sometimes be groundwater-dominated but generally are dominated by surface water. The eastern extremity of the well field has a much greater contribution from groundwater than the western end. Wells #7, #8, #9, #10 mostly hover around

50% SW-GW with the exception of well #10 that has an average contribution of 37% SW. Wells #8 and 12 have the highest range of fluctuation (from below 25% to greater than 75%). Well #7 does not vary more than +/- 12.5% from the 50% mark.

The lowest range of variability is found at wells #2, #7 and #11, each having different average values. Well #2 generally hovers around 60 -70% SW (average 65% SW) with a single month closer to 85% and a period during the winter of 2019 where proportions dropped to near 50% surface water. Well #11 generally hovers around 75% (average 76% SW) surface water and also does not vary by more than +/- 12.5% surface water with the exception of the first month of sampling.

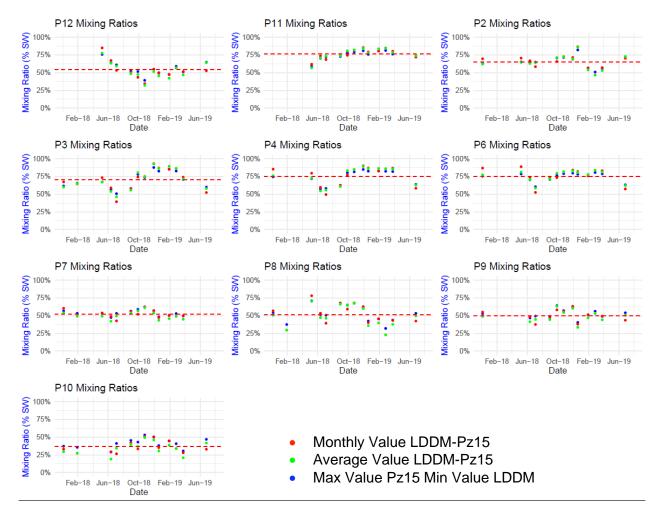


Figure 4.5: Mixing Ratios at all wells using each mixing ratio scenario. The dashed line represents the average value throughout the sample campaign.

4.5.4 Residence times of the bank filtrate

Overall, the majority of methods estimate median residence times inferior to 10 days throughout most of the sampling period (Figure 4.6). The median residence times for all scenarios increase toward the center of the well field reaching a maximum at well #3 (roughly 8 days). The lowest median residence times are found at wells #12 and well #10 at the extremities of the well field (median of roughly 5.5 and 5.25 days respectively).

The greatest variability is found when using the average ²²²Rn value from Pz15 as the groundwater end-member (Figure 4.7; scenario 1). Such reference value for groundwater is equalized or exceeded by the majority of samples (113 out of 147 measurements), leading to the occurrence of a large number of results above 21 days of residence time. Using subsequently higher radon end-members lead to a lower number of measured values corresponding to a residence time greater than 21 days. Since the maximum value used in scenario 2 is higher than the one used in scenario 1, fewer measurements (52 out of 147) exceed this value. As a consequence, it had far fewer calculated residence times that exceeded 21 days. Scenario 3 (Maximum value at each well) uses the local maximum value which means that there are infrequently values that exceed equilibrium value (aside from positive outliers) and residence times are nearly always inferior to 21 days.

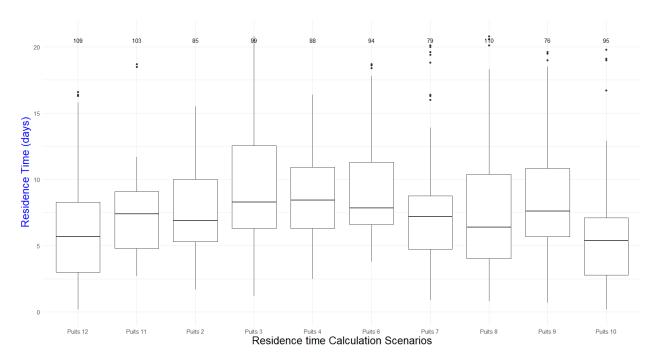


Figure 4.6: Boxplot of residence times for all scenarios at each well.

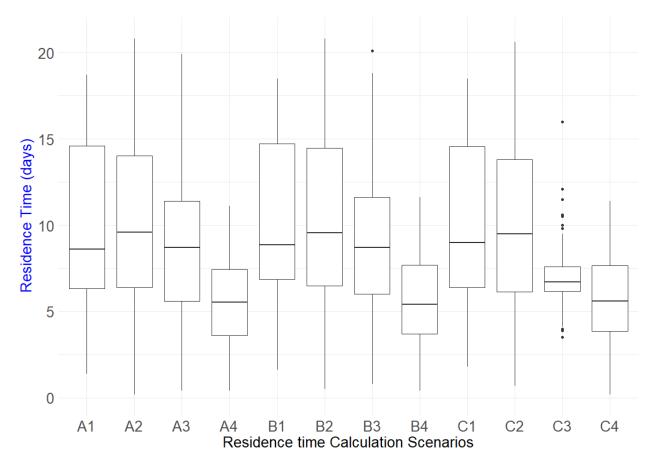


Figure 4.7: Calculated residence times for each of the residence time scenarios with residence times exceeding 21 days removed. Note that scenarios A1, B1, and C1 have lower medians than A2, B2, and C2, however they contain the most residence times exceeding 21 days.

4.6 Discussion

4.6.1 Impact of mixing rates on calculations of residence times

Overall, the three scenarios of EC end-members for calculating mixing ratios tend to produce comparable residence times. The variation was not as significant from one end-member choice to the other and therefore EC end-members did not have a major impact on the residence time calculations.

The impact of the mixing ratio itself on the results is higher insofar as it dictates the value of the corrected Rn activity. The end-members that result in the lowest percentage of groundwater will have a higher calculated residence times since the correction applied to the measured value is smaller. In other words, the Rn end-member choices that result in the smallest proportion of surface water end up having a larger correction applied and become systematically lower. This is a significant limitation of the method, and necessitates further investigation. For instance, well #10 has consistently shorter calculated residence times than well #11 even though well #11 has lower measured activity than well #10. This is the result of a much greater proportion of groundwater (higher EC) at well #10 than at well #11, and not necessarily a reflection of the actual residence times being significantly shorter at well #10.

4.6.2 Impact of Rn end-member on residence time calculations

In scenarios 1 and 2, many measured values exceed the regional equilibrium value for the 222 Rn end-members well (Rn_{GW}) beyond analytical error. In scenarios 3 and 4, very few values fall within error of end-member choice. Therefore, depending on which Rn_{GW} are chosen, the interpretations they provide are very different, varying from frequently being at equilibrium (residence time of >21 days) or else rarely exceeding equilibrium (only anomalously high values and values near equilibrium). Even scenario 4 which used the highest Rn value of all scenarios, still had values that exceeded this choice by greater than analytical error. It is therefore difficult to evaluate which end-member values are the most representative since all of the possible choices have values that are not theoretically possible.

Overall, the Rn_{GW} scenarios considered in this study had a significant impact on the resulting calculated residence times. They varied to a greater degree than for EC and the resulting differences

in residence times ranged from being less than 5 days to being considered at equilibrium (i.e. >21 days). Scenarios for residence time calculation found in the literature can thus be discussed.

In the manner of Bertin and Bourg (1994), Stellato et al. (2013) or Macheleidt et al. (2006), one would expect a piezometer located on the inland side of a well field where regional groundwater discharges toward surface water to provide a representative value, since equilibrium would be constantly reached. Surprisingly, neither of the two scenarios using PZ15 (i.e. average and maximum ²²²Rn) provided consistent results, meaning that there were frequent values that exceed these end-members. Even though a significant portion of the measured values in scenario 2, which used the maximum value at Pz15 (6680 Bq/m³) exceed Rn_{GW}, greater than 50% of calculated residence times are inferior to 10 days. Meanwhile, the scenarios tested at PP22 provided residence times that generally do not exceed 5.5 days, well below the other scenarios. Since certain end-member choices are often exceeded by measured radon values (Scenario 1 and 2) and others are rarely exceeded (Scenario 3 and 4) determining which of these end-member choices is the most representative is not simple.

Hoehn and von Gunten (1989), Dawe and MacQuarrie (2005), Hoehn and Cirpka (2006) and (Kobierska et al., 2015) used the maximum ²²²Rn value measured in the well field as the Rn_{GW}. This method of end-member selection was also tested in the present study, although not presented in detail. Indeed, it resulted in short residence times that did not exceed 4.5 days which would only be in agreement with hydrodynamic calculations when pumping is continuous and at maximum rates (Richard et al., 2009). In addition, many calculated residence times are negative which would imply that transit times of infiltrating surface water are instantaneous.

Snow and Spalding (1997) and Bourke et al. (2014) used the maximum 222 Rn measured in a well, whose groundwater was aimed to be dated, as Rn_{GW}. In the present study, with anomalous maximums excluded, this scenario produced consistent results. Consistent results were expected since this scenario maximizes the difference between samples at each well and minimizes the instances of > 21-day residence times. By design, it can only result in positive radon outliers and radon values equal to equilibrium being interpreted as at equilibrium. Although this method produces appealing results, its representativity of the true residence times remains doubtful, since certain wells experienced anomalously high values and others did not.

In this study, complementary insights into flow dynamics are provided by the four push-points that intercept the bank filtrate between the continuously pumping wells #2 and #7 and the lake. Since on one occasion radon activities in PP19 and PP21 were of the same magnitude as in the wells, the pumped mixture was probably at equilibrium with the substrate prior to reaching the pumping wells. If this is the case, using higher radon values could be returning false interpretations. Although scenarios 1 and 2 frequently had measured values that exceed the equilibrium constant, it is not clear whether they should be considered appropriate or not. If infiltrating lake water is already at equilibrium at the push point locations, then measured values should exceed or be equal to the equilibrium constant. However, since the equilibrium values in these two scenarios are frequently exceeded, this scenario also indicates that this method of end-member selection could be too low.

Each scenario that used subsequently higher Rn_{GW} values ended up with lower residence times and less values that exceeded the Rn_{GW} value, as expected. Depending on which value is chosen, over and under-estimates of residence times are extremely likely. Using Rn_{GW} that is closer to the measured values, small changes in corrected and measured radon have a larger impact on calculated residence times since single day ingrowth is progressively lower as you approach equilibrium.

4.6.3 Evidence of spatial, vertical and temporal heterogeneity of the endmembers and the impact on calculated residence times

Residence time calculations were done using a simple binary mixing model. Since measurements might be affected by spatial heterogeneity, vertical stratification and temporal variability of both EC and ²²²Rn used to define regional groundwater at equilibrium, this section aims at discussing these factors and their impact on residence time calculations.

Evidence of spatial variation in the average Rn and EC is shown by Figure 4.4. On the landward side of well #2, on the western side of the well field, PP24 was used to evaluate the potential for a second groundwater endmember. The low EC at PP24, similar to PZ15 after the 2019 flooding event, may have still been under the influence of the recharge induced by the flooding event. It is therefore not known if the EC values would return to the same levels as those observed in PZ15, or if there is a different EC value for either side of the well field. Not only did the parameters used in the residence time calculations indicate some degree of spatial heterogeneity, the map of surficial

deposits shows a geological contact in the aquifer. Despite the maps showing some change in geology, no distinct differences were noted in the well logs and the grain size distribution curves were mostly similar. These different geological units, if they are indeed present may have different chemistries and therefore lead to differences in Rn and EC which would necessitate the use of at least a spatially variable end-member.

Evidence of vertical stratifications of EC was observed within the aquifer with the upper portion of groundwater having lower EC on both the inland and lake side of the well field. As a result, calculations may not correctly attribute the lower radon activity to infiltrating water from the lake. In fact, a certain proportion of the radon would be incorrectly attributed to surface water but would in fact originate from the upper portion of the aquifer that has similar EC and much higher Rn, and may in fact be at equilibrium with the substrate. This would lead to an over-estimation of the radon activity of infiltrating surface water and an overestimate of the proportion of surface water contributing to the pumped mixture.

Vertical stratification for ²²²Rn is not as clear. Generally speaking, when water infiltrates vertically, radon activity should increase with depth until it reaches equilibrium. This relationship was not consistent on the study site. At certain moments the activity in the surface and deep piezometers were similar, at others the deeper sample had lower activity and some had the expected stratification with lower activity at the surface and higher at depth. This may indicate that there is vertical stratification of radon production, i.e. heterogeneous Rn_{GW} value, throughout the well field. Consistently lower activity measured upgradient at observation wells (Pz15) than at the pumping wells, would be due to lower pumping rates at the first providing less of the higher activity near surface water. In such case where a lower Rn_{GW} value would be responsible for the lower observed radon values on the inland side of the wells, a more robust mathematical or statistical model would be required to produce reliable residence time calculations.

Within close proximity of the study site, there is a well-known carbonatite deposit that contains elevated concentrations of uranium. Depending on the origin of the sediments within the aquifer, it is possible that the erosion of this deposit could have contributed to some spatial heterogeneity of the radon progenitors and in turn spatial variability of radon production within the aquifer. If the aquifer is composed of deltaic deposits originating near these uranium rich rocks, this could create some spatial variability of radon production within the aquifer. However, the carbonatite deposits

themselves are located several kilometers away and therefore, groundwater that flows from those deposits towards the well field would not have an influence on activities within the aquifer since they would have time to decay (greater than 5 half lives or 19 days) and re-equilibrate with the aquifer before reaching the pumping wells.

Temporal variability of the Rn_{GW} end-member is also possible. A dip in activity in August 2018 was observed in all pumping wells and in observation well Pz15. It is uncertain if this decrease is related to an overall decrease in the radon production in the aquifer or some other factor.

4.6.4 Explaining the spikes in radon observed

Based on the foundational theories that were used in this study, the activities measured should not exceed the activity at secular equilibrium. Therefore, the largest spikes in activity should be representative of the equilibrium value, or else an indication that the equilibrium value is in fact even greater than the maximum value measured. However, resulting residence times using these values make the representativity of this value as the equilibrium constant unlikely. These spikes could not be explained which is therefore a significant source of uncertainty on the radon endmembers that merits further studies.

4.6.5 Influence of pumping schemes

Regarding the temporal variability of pumping schemes, it appears that more stable proportion of GW at the frequently pumping wells does not have a significant impact on calculated residence times. In fact, all of the wells have similar variability when excluding values greater than 21 days with the exception of wells #3, #8, and #9.

It was expected that calculated residence times of the wells that pump frequently (#2 and #7), would be shortest and that the wells that pump less frequently would have the longest calculated residence times, if considering all other aspects of the site identical. This however is not the case. The wells that are located at the extremities of the well field and have higher EC values (#10 and #12) are those that have the lowest residence times. This goes against the hydrogeological calculations and is driven largely by their greater proportion of GW. Therefore, we can conclude that monthly pumped hours do not correlate inversely with radon measurements. Looking at the frequency of on/off pumping cycles, average pumping rates, and pumping time before sampling may show a better correlation with the measured radon and calculated residence times, but this could not be

demonstrated as this data was not available on this site. The most apparent factor related to pumping that affected residence times were changes in EC, that seem to be related the total number of hours pumped and the position within the well field (Figure 4.8).

Residence times of 2-7 days for most wells, that were calculated using Scenario 4, are in close agreement with those determined by Richard et al. (2009). However, Richard et al. (2009) considered steady pumping conditions at maximum rates while most samples from the present study came from intermittently pumping wells that do not pump for multiple days in a row, but rather a few hours a day, and the wells that do pump constantly pump at variable rates. Water would thus not transit directly from the lake to the well. Unless gradients remain inversed for prolonged periods in between pumping due to surrounding pumping at wells #2 and #7, the residence times calculated would not represent actual residence times from the lake to the well.

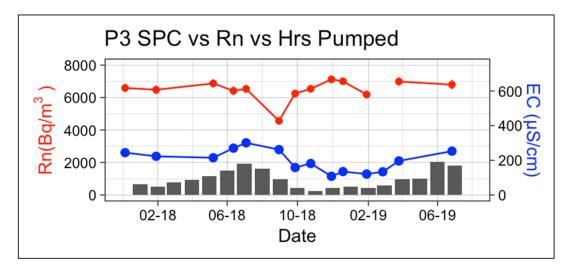


Figure 4.8: Time series of pumping hours, measured ²²²Rn, and measured EC for well 3. The EC seems to have a direct correlation with pumping hours at this well

4.6.6 Implications of higher values measured in surface water

At certain moments throughout the sampling campaign, elevated ²²²Rn activities were observed in surface water, indicating a likely discharge of groundwater into the lake. This may mean that infiltrating water has higher radon activities than what was used for the calculations, and reach the equilibrium value more rapidly. This would lead to an overestimate of residence times. It would also provide strong evidence of flow reversals between groundwater discharge toward the lake and surface water infiltration towards the well. These higher surface water values might be restricted

to a particular place since there is likely discharge of groundwater at various locations throughout the LDDM depending on which wells are pumping.

4.6.7 Potential evidence for a link between residence times and viscosity?

In the month of August 2018, a dip in ²²²Rn activity was observed throughout the well field. It did not correspond to an evident shortening of flow paths, but did occur during a month with an increase in hours pumped overall (3rd highest pumping hours) at all wells and warmer surface water. Since the decrease in activity was observed at nearly all of the observation points, including Pz15, and all measurements fall within roughly 1000 Bq/m³ (average analytical error is typically around 243 Bq/m³) an overall flow regime change as the sole cause of this decrease in activity is unlikely, and some other factor is probably at play here. It is possible that higher demand during this period did change overall flow regimes in the aquifer, however other months with greater amounts of pumping did not experience drops in activity (July 2018 and June 2019). This observed decrease in activity could be related to a shortening of residence times, related to the viscosity contrast between GW which has stable temperature and SW which would be near its hottest temperature during this period and would have had a lower viscosity. There was however no significant drop in EC at most of the wells at this time so this decrease in residence times may not be related to viscosity as that should affect mixing ratios as well as residence times.

Note that an investigation into the possibility that the decrease in activities was related to an overall decrease of residence times in the entire aquifer due to a heavy recharge event prior to sampling was also considered. However, there was no significant precipitation event prior to the sample campaign and this seems to be an unlikely scenario (Meteoblue, 2020).

4.6.8 Recommendations

Although there have been a number of studies that have used Radon activity to estimate residence times in aquifers, there is still some considerable work that should be done to outline the sampling procedures and the hydrogeological conditions that are required for the method to provide representative results.

4.6.8.1 Establishing when data should be excluded from calculations

Certain papers such as Schilling et al. (2017) excluded values (considered them at equilibrium) once they fell within error of the equilibrium constant. This seems appropriate for situations where there is 100% infiltrating surface water, but may not be appropriate when taking into consideration mixing ratios. As Gilfedder et al. (2019) mentions, there is an underestimate of calculated residence times when there is mixing with ambient groundwater. This was also observed in this study, most notably at wells #10 and #12. Although the proportion of groundwater required for the underestimates to be significant is unknown, it is clear that at a certain proportion of groundwater, the resulting residence times are no longer representative. Therefore, it is proposed that rather than excluding values within error of equilibrium, a more robust method of excluding data prior to calculations that combines mixing ratios and error on the measurements is used. Alternatively, this problem can be rectified more simply by establishing a threshold based on mixing ratios for excluding values prior to calculations.

4.6.8.2 Pumping time required for representative sampling

In order to evaluate the pumping time required for stable representative radon measurements, and since EC stability does not necessarily indicate stability of the ²²²Rn measurements, it is recommended that early on in the sample program, a test should be conducted in a similar manner as Macheleidt et al. (2006). The test can be done using a RAD Aqua that takes continuous measurements. This will help reduce uncertainty in the causes of residence time fluctuations and rule out purge time as a cause of error. These measurements will also establish if a stable ²²²Rn value is ever achieved.

A test taking continuous measurements, or multiple samples should also be performed on observation wells situated both up gradient and between the pumping wells and the surface water source. This would help to ensure that a stable representative value was taken and will increase confidence in the ²²²Rn_{GW} value for the aquifer.

4.6.8.3 Identifying and quantifying sources of error

There is still a high degree of uncertainty as to the principal causes of fluctuations in radon activity at the well field. It does not appear to be directly related to pumping, nor does it appear to be related to hydraulic conditions in the lake. In future studies, a number of aspects should be tested in order

to better evaluate the representativity of data, and isolate certain potential causes of error, and determine the Rn_{GW} value.

The error of the measurements is not known precisely and we can therefore not determine the exact limits of the residence time calculations. For instance, if error is determined to be in the range of 1000 Bq/m³, all but one to two value at most wells would fall within error of the median value. The duplicate values taken indicate that there is a potentially significant source of error that exists within the sampling procedure, or analytical methods and that this range of error is possible.

The measurements may be affected by the flow-rate, or various other factors related to sampling. Considering sampling was done using different types of pumps with different flow rates for each type of piezometer, and the wells were sampled directly from a tap in the well chamber, some variability may be related to these variations.

Once the time between the start of pumping and stable measurements is determined, a series of experiments should be conducted on a single well with the sole objective of determining sources of error and their impact on measured results. Multiple bottles should be taken on the same well in order to determine the impact of the time elapsed between sampling and laboratory analysis. A second series of bottles should be taken to test the physical state of the bottle on results. Certain bottles had been dented over the course of the sampling campaign, and it is uncertain whether that impacts the measurements.

These tests should also be repeated seasonally in order to see if water temperature impacts repeatability of measurements. The analysis during winter and summer will also help determine if storage of bottles in a warm or cold location prior to sampling impacts measurements. Thermal expansion and contractions may affect the seal of the bottles and cause differences in different conditions.

Performing this battery of tests would provide a better idea of what the absolute error on measurements. Only analytical error has been quantified at this time and absolute error and the precision of the measurements using the RAD7 Big Bottle System to the best of the authors knowledge have not been precisely quantified.

4.6.8.4 Implementation of a rigorous QA/QC program

The duplicates that returned different values raise doubt in the representativity of the measurements. It will be important to implement systematic duplicates of both a single well during each campaign, but also a random duplicate sample. When these duplicates are taken, the physical state of the bottle should be noted as well. It is suspected that the age and quality of the bottle or the seal of the cap may have had a role in the difference in duplicate values. In addition to duplicate values, standards should be integrated into the sampling program systematically in order to isolate error associated with instrumental drift.

4.6.8.5 Determining optimal groundwater end-member and observation well placement

It is important to outline what factors should be considered when selecting the end-members. These factors could not all be respected within this study which is a significant source of uncertainty that leads to a low degree of confidence in the results. The factors to consider when selecting an end member are as follows

- The location of the observation point: the observation point should be on the landward side at a sufficient distance and depth that it is not influenced by infiltrating surface water that would be reaching the observation well in less than 21 days. In other words, the groundwater sample location should be located outside of the area of influence of the wells. The location chosen should have a stable activity that only fluctuates within error of the measurement and no measurements should exceed the equilibrium value.
- Geology of the aquifer: The geology and aquifer thickness at the end-member location should be the same as sample location where residence times have to be assessed and the sample depth should be similar. Hoehn and von Gunten (1989) observed higher activity in a well near the surface that was not in agreement with the ingrowth in the rest of the aquifer. They attributed this higher value to the fact that the well screen was completely submerged at this sample location. Stellato et al. (2013) also observed higher activities in the period of high-water levels during their study. They attributed this to longer residence times, however it may be an indication of a spatially or temporally variable Rn_{GW} value.

Mixing ratio and radon tracer end-member location: Both end-members should be
from the same location. If radon and groundwater end-members cannot be chosen
at the same location, significant bias would be introduced in the selection of each.

These criteria could not all be met during this study and therefore the end-member choice could not be made in the most logical of manners and thus the presented results must be considered to contain a significant bias despite the attempts to minimize it using various end-member choices. To avoid some complications, end-member locations should be subjected to a continuous test similar to those done at the pumping wells in order to ensure that a stable representative value is taken and to reduce uncertainty. For these results to be most meaningful, a comparison should also be done between the RAD Aqua and Big Bottle systems to confirm that the same relationship exists using both methods of analysis.

4.7 Conclusions

The objective was to evaluate the impact of end-member choice on calculated residence times using ²²²Rn at a bank filtration site. Several scenarios for both the EC and ²²²Rn end-members choices were selected, and their impact was discussed. End-members for calculating mixing ratios were fairly clear on the site throughout most of the sample program, with the values at all wells falling within the range of values observed in surface water (LDDM) and regional groundwater (Pz15). The EC end-member choice that led to the greater proportion of groundwater led to shorter residence times as the applied correction was greater. This was particularly evident when the proportion of surface water was low (<50%) and the resulting residence times was typically short (<5 days). The radon end-members were less evident and presented an opportunity to determine the impact of the end-member choice on calculated residence times. Scenarios using various groundwater end-members and the maximum value at each well were tested. Using progressively higher radon values also led to shorter calculated residence times, and using the maximum measured radon values caused all samples to have short residence times. Through this investigation, it was confirmed that mixing with ambient groundwater leads to an underestimate of residence times since the correction that is applied to the radon measurements increases. As a consequence of this observation, the uncertainty on the Rn end-member, and the precision of measurements, raised a series of questions about the use of the radon method for calculating residence times. Furthermore, presence of spatial (stratification and throughout the site) and temporal variability of the end-members complicated interpretations. Despite this method being considered well developed by some authors, additional work should be conducted to establish the exact hydrogeological conditions that are required for this approach to provide representative results. A clear stable radon end-member, high confidence in the representativity of samples and simple binary mixing are all essential for the approach presented herein to be valid. Several steps are recommended in order to improve this method and determine when and under what conditions it is appropriate. Also, some general guidelines to consider when selecting an end-member sampling location were outlined.

CHAPTER 5 GENERAL DISCUSSION

The work completed during this research project was able to demonstrate that there is likely to be a large number of people within the province of Quebec that are supplied by bank filtration sites. Since there are no specific regulations that address these sites, they are most often characterized as traditional groundwater sites and are monitored and managed in the same manner. Additional criteria should be incorporated into the existing framework to improve management of these sites namely by determining early on whether there is mixing between surface and groundwater. This can be done by sampling groundwater and surface water in addition to pumping wells, and EC is a promising tracer for this purpose, but it does require infrastructure in appropriate locations to evaluate mixing at pumping wells. The second tracer that was investigated as a tool for assessing interactions between groundwater and surface water, ²²²Rn using the low cost RAD7 apparatus, proved to be less promising. The use of ²²²Rn requires very specific flow and mixing conditions, and a high degree of precision in measurements to produce representative residence. The precision of the measurements is an extremely important factor in determining whether the calculated residence times are meaningful. In fact, with decreased precision of measurements, increased uncertainty on the end-members, and mixing with groundwater, there is a very narrow range of values that do not fall within error of either equilibrium or the minimum theoretical value. This approach does not generally provide reliable quantification of residence times and has more potential as a tool to assess broadly whether residence times are short (<21 days) than as a high precise tool. Even as a tool to broadly assess whether residence times are short, a high degree of confidence in the end-members is required.

CHAPTER 6 GENERAL CONCLUSION

The objectives of this study were to evaluate the occurrence of bank filtration sites in the province of Quebec and to evaluate the potential of ²²²Rn as a tracer for calculating residence times on a multi-well bank filtration site.

Through the GIS approach, it was demonstrated that there are likely to be a large number of municipalities that are taking advantage of the benefits of bank filtration throughout the province of Quebec. From this provincial scale approach, we show that nearly one million people are supplied by groundwater from municipal wells located < 500 m from a surface water body and half a million would have a significant probability to be supplied by IBF wells. A more focused look at watershed scale distribution of wells allows us to improve our interpretations by considering the aquifer type and other regional factors. This approach reveals strong spatial variability in the distribution of wells in proximity to surface water. Of the three selected regions, one has high potential for IBF (Laurentides), one requires additional information do draw precise conclusions (Nicolet), and the third region (Vaudreuil-Soulanges) is unlikely to have widespread use of IBF. With this study, we demonstrate that extensive use of IBF is likely and that there is a need for improved understanding and management of these sites in order to properly protect the drinking water supply. This study of the widespread use of bank filtration throughout the province of Quebec has enabled regional scale studies of the use of environmental tracers in more precisely evaluating the occurrence of bank filtration with some easily accessible environmental tracers.

Although ²²²Rn was identified as a promising tracer for evaluating residence times for infiltrating surface water, there are many conditions present on this site that led to a low degree of confidence in the resulting residence times. The influence of mixing ratios on calculated residence times was evident, and led to clear underestimates of residence times in wells with a higher proportion of groundwater. The relationship between pumping and calculated residence times could not be clearly identified and as such it remains unclear whether the calculated residence times at any well were representative of the actual residence times. For wells with uncertain ²²²Rn end-members, using a maximum measured value as an end-member leads to interpretations of short (<21 day) residence time and using lower values leads to a large number of values exceeding equilibrium. The use of the RAD7 apparatus for precise calculations does not appear to be promising on the study site as all measured values fall within a small range and the precision of the apparatus is not

certain. Further work is therefore required to evaluate the precise hydrogeological conditions required on a site for radon to be effective and reliable tracer, and to evaluate the precision of the measurements required, and the proportion of groundwater where values should be excluded from interpretations. Guidelines for choosing end-member locations were also discussed and should be considered in further study to avoid any ambiguity that this can cause in the interpretations of the site.

Now that there is a higher degree of awareness of the use of BF techniques for supplying many municipalities within the province of Quebec, some more targeted studies can be done on the regions with high and moderate potential to further refine the results. The use of EC is appropriate for estimating mixing ratios with ambient groundwater on the study site. Although this parameter may not be a perfect tracer for precise mixing ratio calculations on the study site, it can still be used with a high degree of confidence to determine more broadly if mixing with groundwater is occurring at a pumping well at the municipalities that were identified in the first section of this dissertation.

With the abundance of potential BF sites in the province of Quebec, the importance of developing techniques to further refine the level of confidence in the actual occurrence of these sites using chemical tracer techniques has become even more important. Although radon did not provide detailed insights into the residence times of infiltrating groundwater on the study site investigated, it remains a promising tool for evaluating transit times, although additional work to refine and improve this technique is required. Electrical conductivity remains a promising tracer for distinguishing between surface water and groundwater on sites similar to the one studied herein. It can therefore be concluded that both of these tools are useful for a more detailed study of BF sites, although these tracers do require prior knowledge of groundwater flow and a rough idea of residence times prior to being implemented. Radon especially requires a thorough understanding of flow dynamics and the nature of pumping and the special heterogeneity of the aquifer to be a useful tracer. We recommend a large-scale geochemical study in order to further refine the results from the GIS study and to confirm that the cut-off of 500 m that was used in order to considering BF likely is appropriate.

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APPENDIX A RESULTS

| Well | Sample | SPC (µS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | B4 | C1 | C2 | С3 | C4 |
|--------|---------------|----------------|------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| Well 1 | JOS-171206-01 | 199,2 | 6454 | 0,71 | 0,79 | 0,70 | 16,6 | 21,0 | 6,4 | 21,0 | 17,3 | 21,0 | 7,0 | 21,0 | 16,6 | 21,0 | 6,4 |
| Well 1 | JOS-180129-01 | 187,5 | 5994 | 0,73 | 0,73 | 0,72 | 10,8 | 12,8 | 5,4 | 21,0 | 10,8 | 12,8 | 5,4 | 21,0 | 10,7 | 7,5 | 5,3 |
| Well 2 | JOS-181024-02 | 192,4 | 7430 | 0,72 | 0,72 | 0,71 | 21,0 | 21,0 | 10,8 | 21,0 | 21,0 | 21,0 | 10,8 | 21,0 | 21,0 | 8,7 | 10,7 |
| Well 2 | JOS-181219-02 | 136,7 | 6762 | 0,85 | 0,86 | 0,82 | 21,0 | 12,3 | 8,5 | 21,0 | 21,0 | 12,4 | 8,5 | 21,0 | 21,0 | 8,2 | 8,2 |
| Well 2 | JOS-190626-02 | 190 | 6715 | 0,73 | 0,67 | 0,72 | 21,0 | 11,1 | 7,4 | 21,0 | 21,0 | 10,6 | 6,9 | 21,0 | 21,0 | 7,1 | 7,3 |
| Well 2 | JOS-180508-02 | 225 | 6603 | 0,64 | 0,70 | 0,65 | 22,0 | 9,6 | 6,4 | 21,0 | 21,0 | 10,1 | 6,9 | 21,0 | 21,0 | 6,6 | 6,4 |
| Well 2 | JOS-181129-02 | 205,8 | 6586 | 0,69 | 0,71 | 0,69 | 21,3 | 9,9 | 6,7 | 21,0 | 21,5 | 10,1 | 6,9 | 21,0 | 21,0 | 6,7 | 6,7 |
| Well 2 | JOS-190326-02 | 273,8 | 6457 | 0,53 | 0,57 | 0,56 | 15,1 | 7,6 | 4,8 | 21,0 | 15,5 | 8,0 | 5,2 | 21,0 | 15,4 | 6,2 | 5,1 |
| Well 2 | JOS-180704-02 | 227 | 6268 | 0,64 | 0,58 | 0,65 | 12,8 | 7,7 | 5,3 | 21,0 | 12,3 | 7,2 | 4,8 | 21,0 | 12,9 | 6,1 | 5,4 |
| Well 2 | JOS-180612-02 | 231,9 | 6233 | 0,63 | 0,67 | 0,64 | 12,3 | 7,4 | 5,1 | 21,0 | 12,6 | 7,8 | 5,5 | 21,0 | 12,3 | 6,0 | 5,2 |
| Well 2 | JOS-190129-02 | 270 | 6077 | 0,54 | 0,57 | 0,56 | 9,8 | 5,9 | 3,9 | 21,0 | 10,0 | 6,2 | 4,2 | 21,0 | 10,0 | 5,8 | 4,1 |
| Well 2 | JOS-180927-02 | 196,4 | 5963 | 0,71 | 0,66 | 0,71 | 10,4 | 7,0 | 5,1 | 21,0 | 9,9 | 6,6 | 4,7 | 21,0 | 10,3 | 5,8 | 5,1 |
| Well 2 | JOS-171206-02 | 235,5 | 4550 | 0,62 | 0,69 | 0,63 | 3,6 | 2,5 | 1,7 | 7,1 | 4,3 | 3,2 | 2,4 | 6,5 | 3,7 | 4,0 | 1,8 |
| Well 3 | JOS-181129-03 | 107,6 | 7122 | 0,92 | 0,93 | 0,87 | 21,0 | 21,0 | 10,4 | 21,0 | 21,0 | 21,0 | 10,4 | 21,0 | 21,0 | 12,1 | 10,1 |
| Well 3 | JOS-181219-03 | 134,8 | 7003 | 0,86 | 0,87 | 0,82 | 21,0 | 21,0 | 9,5 | 21,0 | 21,0 | 21,0 | 9,5 | 21,0 | 21,0 | 10,0 | 9,2 |
| Well 3 | JOS-190326-03 | 196,3 | 6989 | 0,71 | 0,74 | 0,71 | 21,0 | 19,9 | 8,4 | 21,0 | 21,0 | 20,1 | 8,6 | 21,0 | 21,0 | 8,3 | 8,3 |
| Well 3 | JOS-180508-03 | 214,7 | 6871 | 0,67 | 0,73 | 0,67 | 21,0 | 16,1 | 7,5 | 21,0 | 21,0 | 16,6 | 8,0 | 21,0 | 21,0 | 7,7 | 7,6 |
| Well 3 | JOS-181024-03 | 181,6 | 6541 | 0,75 | 0,75 | 0,73 | 19,6 | 12,1 | 7,0 | 21,0 | 19,6 | 12,1 | 7,0 | 21,0 | 19,5 | 7,5 | 6,9 |
| Well 3 | JOS-190129-03 | 120,7 | 6187 | 0,89 | 0,85 | 0,85 | 13,6 | 10,5 | 6,9 | 21,0 | 13,4 | 10,2 | 6,7 | 21,0 | 13,4 | 7,8 | 6,7 |
| Well 3 | JOS-190626-03 | 252,9 | 6796 | 0,58 | 0,49 | 0,60 | 21,0 | 13,9 | 6,4 | 21,0 | 21,0 | 13,0 | 5,5 | 21,0 | 21,0 | 7,2 | 6,6 |
| Well 3 | JOS-180927-03 | 158,1 | 6250 | 0,80 | 0,74 | 0,78 | 13,8 | 10,3 | 6,5 | 21,0 | 13,4 | 9,9 | 6,1 | 21,0 | 13,6 | 7,2 | 6,4 |
| Well 3 | JOS-171206-03 | 244,3 | 6591 | 0,60 | 0,67 | 0,61 | 20,8 | 11,4 | 5,9 | 21,0 | 21,5 | 12,0 | 6,6 | 21,0 | 21,0 | 6,9 | 6,1 |
| Well 3 | JOS-180129-03 | 223 | 6480 | 0,65 | 0,65 | 0,65 | 16,8 | 10,8 | 6,0 | 21,0 | 16,8 | 10,8 | 6,0 | 21,0 | 16,9 | 6,9 | 6,1 |
| Well 3 | JOS-180612-03 | 270,3 | 6416 | 0,54 | 0,59 | 0,56 | 14,3 | 9,2 | 4,8 | 21,0 | 14,8 | 9,7 | 5,3 | 21,0 | 14,6 | 6,6 | 5,1 |
| Well 3 | JOS-180704-03 | 301 | 6536 | 0,46 | 0,39 | 0,51 | 16,8 | 9,4 | 4,3 | 21,0 | 15,9 | 8,6 | 3,4 | 21,0 | 17,3 | 6,7 | 4,8 |
| Well 3 | JOS-180830-03 | 262 | 4563 | 0,56 | 0,58 | 0,58 | 3,1 | 2,4 | 1,2 | 6,1 | 3,3 | 2,6 | 1,4 | 6,2 | 3,3 | 4,3 | 1,4 |
| Well 4 | JOS-181219-04 | 135 | 7286 | 0,86 | 0,87 | 0,82 | 21,0 | 21,0 | 10,9 | 21,0 | 21,0 | 21,0 | 10,9 | 21,0 | 21,0 | 10,5 | 10,6 |
| Well 4 | JOS-181129-04 | 121,9 | 6956 | 0,89 | 0,90 | 0,85 | 21,0 | 16,3 | 9,5 | 21,0 | 21,0 | 16,4 | 9,5 | 21,0 | 21,0 | 9,8 | 9,2 |
| Well 4 | JOS-180927-04 | 146,5 | 6806 | 0,83 | 0,77 | 0,80 | 21,0 | 13,9 | 8,5 | 21,0 | 21,0 | 13,4 | 8,0 | 21,0 | 21,0 | 8,5 | 8,3 |

| Well | Sample | SPC (µS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | В4 | C1 | C2 | С3 | C4 |
|--------|---------------|----------------|------------|------|------|------|------|------|------|------|------|------|------|------|------|-----|------|
| Well 4 | JOS-190326-04 | 137,8 | 6725 | 0,85 | 0,86 | 0,82 | 21,0 | 13,2 | 8,3 | 21,0 | 21,0 | 13,2 | 8,4 | 21,0 | 21,0 | 8,5 | 8,1 |
| Well 4 | JOS-190626-04 | 232 | 7050 | 0,63 | 0,55 | 0,64 | 21,0 | 16,2 | 7,9 | 21,0 | 21,0 | 15,5 | 7,2 | 21,0 | 21,0 | 7,5 | 8,0 |
| Well 4 | JOS-180508-04 | 190,5 | 6886 | 0,73 | 0,79 | 0,72 | 21,0 | 14,1 | 8,1 | 21,0 | 21,0 | 14,6 | 8,5 | 21,0 | 21,0 | 7,8 | 8,0 |
| Well 4 | JOS-190129-04 | 133,5 | 6310 | 0,86 | 0,83 | 0,82 | 15,0 | 10,2 | 7,1 | 21,0 | 14,8 | 10,0 | 6,8 | 21,0 | 14,8 | 7,5 | 6,8 |
| Well 4 | JOS-181024-04 | 139,7 | 6041 | 0,85 | 0,84 | 0,81 | 11,9 | 8,8 | 6,3 | 21,0 | 11,9 | 8,7 | 6,3 | 21,0 | 11,7 | 6,8 | 6,1 |
| Well 4 | JOS-180704-04 | 262 | 6639 | 0,56 | 0,49 | 0,58 | 24,7 | 10,0 | 5,7 | 21,0 | 21,0 | 9,4 | 5,0 | 21,0 | 21,0 | 6,7 | 5,9 |
| Well 4 | JOS-171206-04 | 177,1 | 6012 | 0,76 | 0,85 | 0,74 | 11,1 | 8,0 | 5,6 | 21,0 | 11,7 | 8,7 | 6,2 | 21,0 | 11,0 | 6,2 | 5,5 |
| Well 4 | JOS-180612-04 | 266,9 | 6287 | 0,54 | 0,59 | 0,57 | 12,2 | 7,5 | 4,5 | 21,0 | 12,7 | 8,0 | 5,0 | 21,0 | 12,4 | 6,2 | 4,8 |
| Well 4 | JOS-180830-04 | 240,7 | 5083 | 0,61 | 0,62 | 0,62 | 5,1 | 3,8 | 2,5 | 10,8 | 5,2 | 4,0 | 2,6 | 10,7 | 5,2 | 4,7 | 2,6 |
| Well 5 | JOS-171206-05 | 160,5 | 6889 | 0,80 | 0,90 | 0,77 | 21,0 | 21,0 | 8,6 | 21,0 | 21,0 | 21,0 | 9,2 | 21,0 | 21,0 | 9,5 | 8,4 |
| Well 5 | JOS-180129-05 | 146,9 | 6483 | 0,83 | 0,83 | 0,80 | 18,3 | 14,5 | 7,4 | 21,0 | 18,3 | 14,5 | 7,4 | 21,0 | 18,1 | 8,6 | 7,2 |
| Well 6 | JOS-180704-06 | 250 | 7633 | 0,58 | 0,52 | 0,60 | 21,0 | 21,0 | 11,1 | 21,0 | 21,0 | 21,0 | 10,6 | 21,0 | 21,0 | 7,8 | 11,3 |
| Well 6 | JOS-190326-06 | 154,7 | 6967 | 0,81 | 0,83 | 0,78 | 21,0 | 12,2 | 9,0 | 21,0 | 21,0 | 12,3 | 9,1 | 21,0 | 21,0 | 7,9 | 8,8 |
| Well 6 | JOS-181024-06 | 153,1 | 6922 | 0,82 | 0,81 | 0,79 | 21,0 | 11,9 | 8,8 | 21,0 | 21,0 | 11,9 | 8,8 | 21,0 | 21,0 | 7,8 | 8,6 |
| Well 6 | JOS-180508-06 | 155,3 | 6852 | 0,81 | 0,89 | 0,78 | 21,0 | 11,3 | 8,5 | 21,0 | 21,0 | 11,8 | 9,0 | 21,0 | 21,0 | 7,6 | 8,3 |
| Well 6 | JOS-180612-06 | 198,7 | 6818 | 0,71 | 0,73 | 0,70 | 21,0 | 10,3 | 7,6 | 21,0 | 21,0 | 10,6 | 7,8 | 21,0 | 21,0 | 6,9 | 7,6 |
| Well 6 | JOS-190129-06 | 168,8 | 6510 | 0,78 | 0,76 | 0,76 | 18,7 | 9,1 | 7,1 | 21,0 | 18,6 | 9,0 | 7,0 | 21,0 | 18,6 | 6,7 | 7,0 |
| Well 6 | JOS-180927-06 | 162,5 | 6470 | 0,79 | 0,73 | 0,77 | 17,7 | 9,0 | 7,1 | 21,0 | 17,2 | 8,6 | 6,6 | 21,0 | 17,5 | 6,7 | 6,9 |
| Well 6 | JOS-171206-06 | 171,4 | 6479 | 0,77 | 0,87 | 0,75 | 17,8 | 8,9 | 7,0 | 21,0 | 18,4 | 9,6 | 7,6 | 21,0 | 17,6 | 6,6 | 6,8 |
| Well 6 | JOS-190626-06 | 235,6 | 6692 | 0,62 | 0,54 | 0,63 | 21,0 | 8,8 | 6,5 | 21,0 | 21,0 | 8,0 | 5,7 | 21,0 | 21,0 | 6,4 | 6,6 |
| Well 6 | JOS-181219-06 | 157,8 | 6140 | 0,80 | 0,82 | 0,78 | 12,6 | 7,7 | 6,2 | 21,0 | 12,7 | 7,8 | 6,3 | 21,0 | 12,4 | 6,2 | 6,1 |
| Well 6 | JOS-181129-06 | 149,1 | 5014 | 0,82 | 0,84 | 0,80 | 6,5 | 4,8 | 4,0 | 11,6 | 6,6 | 4,9 | 4,1 | 11,3 | 6,3 | 4,5 | 3,8 |
| Well 6 | JOS-180830-06 | 195,5 | 5355 | 0,71 | 0,73 | 0,71 | 7,0 | 4,8 | 3,9 | 16,6 | 7,1 | 4,9 | 4,0 | 16,5 | 7,0 | 4,8 | 3,8 |
| Well 7 | JOS-171206-07 | 269,9 | 4528 | 0,54 | 0,60 | 0,57 | 2,8 | 1,9 | 0,9 | 6,2 | 3,4 | 2,5 | 1,6 | 5,8 | 3,1 | 4,2 | 1,2 |
| Well 7 | JOS-180830-07 | 271 | 4970 | 0,53 | 0,55 | 0,56 | 4,0 | 2,8 | 1,6 | 8,9 | 4,2 | 3,0 | 1,8 | 9,0 | 4,3 | 4,7 | 1,9 |
| Well 7 | JOS-180508-07 | 289 | 6589 | 0,49 | 0,54 | 0,53 | 19,6 | 8,8 | 4,8 | 21,0 | 20,1 | 9,3 | 5,3 | 21,0 | 20,0 | 6,6 | 5,2 |
| Well 7 | JOS-180612-07 | 318,5 | 6589 | 0,42 | 0,49 | 0,47 | 18,8 | 8,0 | 4,0 | 21,0 | 19,6 | 8,8 | 4,8 | 21,0 | 19,4 | 6,7 | 4,6 |
| Well 7 | JOS-180129-07 | 287 | 6633 | 0,50 | 0,50 | 0,53 | 23,3 | 9,2 | 5,0 | 21,0 | 21,0 | 9,2 | 5,0 | 21,0 | 21,0 | 6,6 | 5,4 |
| Well 7 | JOS-190129-07 | 303 | 6700 | 0,46 | 0,50 | 0,50 | 21,0 | 9,3 | 4,8 | 21,0 | 21,0 | 9,9 | 5,3 | 21,0 | 21,0 | 6,7 | 5,3 |

| Well | Sample | SPC (μS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | B4 | C1 | C2 | С3 | C4 |
|--------|---------------|----------------|------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| Well 7 | JOS-180927-07 | 258 | 6978 | 0,56 | 0,52 | 0,59 | 21,0 | 13,9 | 7,1 | 21,0 | 21,0 | 13,4 | 6,6 | 21,0 | 21,0 | 7,2 | 7,3 |
| Well 7 | JOS-181024-07 | 239 | 7080 | 0,61 | 0,62 | 0,62 | 21,0 | 16,3 | 7,9 | 21,0 | 21,0 | 16,4 | 8,0 | 21,0 | 21,0 | 7,5 | 8,1 |
| Well 7 | JOS-181129-07 | 270 | 7282 | 0,54 | 0,57 | 0,56 | 21,0 | 21,0 | 8,3 | 21,0 | 21,0 | 21,0 | 8,6 | 21,0 | 21,0 | 7,6 | 8,5 |
| Well 7 | JOS-190326-07 | 307 | 7291 | 0,45 | 0,50 | 0,49 | 21,0 | 21,0 | 7,3 | 21,0 | 21,0 | 21,0 | 7,9 | 21,0 | 21,0 | 7,6 | 7,9 |
| Well 7 | JOS-181219-07 | 313 | 7307 | 0,43 | 0,47 | 0,48 | 21,0 | 21,0 | 7,2 | 21,0 | 21,0 | 21,0 | 7,7 | 21,0 | 21,0 | 7,6 | 7,8 |
| Well 7 | JOS-180704-07 | 288 | 9996 | 0,49 | 0,43 | 0,53 | 21,0 | 21,0 | 21,0 | 21,0 | 21,0 | 21,0 | 21,0 | 21,0 | 21,0 | 16,0 | 21,0 |
| Well 8 | JOS-180927-08 | 225,7 | 7566 | 0,64 | 0,59 | 0,65 | 21,0 | 21,0 | 11,1 | 21,0 | 21,0 | 21,0 | 10,7 | 21,0 | 21,0 | 8,1 | 11,2 |
| Well 8 | JOS-181024-08 | 213 | 7135 | 0,67 | 0,68 | 0,67 | 21,0 | 13,5 | 8,7 | 21,0 | 21,0 | 13,6 | 8,8 | 21,0 | 21,0 | 7,4 | 8,7 |
| Well 8 | JOS-190626-08 | 287,6 | 7208 | 0,49 | 0,39 | 0,53 | 21,0 | 12,8 | 7,4 | 21,0 | 21,0 | 11,5 | 6,1 | 21,0 | 21,0 | 7,1 | 7,8 |
| Well 8 | JOS-190326-08 | 337 | 7300 | 0,38 | 0,43 | 0,44 | 21,0 | 13,0 | 6,4 | 21,0 | 21,0 | 13,7 | 7,2 | 21,0 | 21,0 | 7,3 | 7,2 |
| Well 8 | JOS-180508-08 | 196,1 | 6465 | 0,71 | 0,78 | 0,71 | 17,0 | 8,7 | 6,5 | 21,0 | 17,5 | 9,2 | 7,0 | 21,0 | 16,9 | 6,4 | 6,4 |
| Well 8 | JOS-180612-08 | 297,4 | 6600 | 0,47 | 0,53 | 0,51 | 20,1 | 7,1 | 4,6 | 21,0 | 20,8 | 7,8 | 5,3 | 21,0 | 20,6 | 6,3 | 5,1 |
| Well 8 | JOS-190129-08 | 328,5 | 6477 | 0,40 | 0,45 | 0,45 | 14,1 | 5,5 | 3,3 | 21,0 | 14,8 | 6,3 | 4,1 | 21,0 | 14,8 | 6,4 | 4,0 |
| Well 8 | JOS-181219-08 | 344,5 | 6580 | 0,36 | 0,40 | 0,42 | 17,4 | 5,5 | 3,1 | 21,0 | 18,0 | 6,2 | 3,7 | 21,0 | 18,3 | 6,6 | 4,0 |
| Well 8 | JOS-180704-08 | 301 | 6186 | 0,46 | 0,39 | 0,51 | 10,0 | 5,1 | 3,3 | 21,0 | 9,1 | 4,2 | 2,4 | 21,0 | 10,5 | 5,9 | 3,8 |
| Well 8 | JOS-180830-08 | 218 | 5303 | 0,66 | 0,68 | 0,66 | 6,4 | 4,3 | 3,3 | 14,8 | 6,5 | 4,5 | 3,5 | 14,7 | 6,4 | 4,7 | 3,4 |
| Well 8 | JOS-181129-08 | 244,7 | 5308 | 0,60 | 0,63 | 0,61 | 5,8 | 3,8 | 2,8 | 14,5 | 6,1 | 4,1 | 3,1 | 14,4 | 6,0 | 4,7 | 2,9 |
| Well 8 | JOS-171206-08 | 283 | 4732 | 0,51 | 0,57 | 0,54 | 3,0 | 1,6 | 0,9 | 7,1 | 3,6 | 2,3 | 1,5 | 6,8 | 3,4 | 4,4 | 1,3 |
| Well 8 | JOS-180129-08 | 370 | 5625 | 0,30 | 0,30 | 0,37 | 3,5 | 0,8 | | 21,0 | 3,5 | 0,8 | | 21,0 | 4,7 | 6,1 | 0,9 |
| Well 9 | JOS-181219-09 | 354,8 | 13069 | 0,33 | 0,38 | 0,40 | 21,0 | 18,2 | 10,7 | 21,0 | 21,0 | 18,4 | 10,9 | 21,0 | 21,0 | 7,6 | 10,8 |
| Well 9 | JOS-171206-09 | 290 | 7721 | 0,49 | 0,55 | 0,53 | 21,0 | 21,0 | 11,0 | 21,0 | 21,0 | 21,0 | 11,6 | 21,0 | 21,0 | 7,6 | 11,4 |
| Well 9 | JOS-181129-09 | 242,7 | 7553 | 0,60 | 0,63 | 0,62 | 21,0 | 18,2 | 10,7 | 21,0 | 21,0 | 18,5 | 10,9 | 21,0 | 21,0 | 7,6 | 10,8 |
| Well 9 | JOS-181024-09 | 267,9 | 7340 | 0,54 | 0,55 | 0,57 | 21,0 | 13,1 | 8,6 | 21,0 | 21,0 | 13,2 | 8,8 | 21,0 | 21,0 | 7,1 | 8,9 |
| Well 9 | JOS-190326-09 | 310 | 7438 | 0,44 | 0,49 | 0,49 | 21,0 | 13,6 | 8,1 | 21,0 | 21,0 | 14,2 | 8,7 | 21,0 | 21,0 | 7,2 | 8,7 |
| Well 9 | JOS-190626-09 | 283,7 | 6849 | 0,50 | 0,40 | 0,54 | 21,0 | 8,2 | 5,9 | 21,0 | 21,0 | 6,9 | 4,6 | 21,0 | 21,0 | 6,4 | 6,3 |
| Well 9 | JOS-180927-09 | 230 | 6560 | 0,63 | 0,58 | 0,64 | 19,5 | 7,9 | 6,1 | 21,0 | 19,0 | 7,4 | 5,7 | 21,0 | 19,6 | 6,1 | 6,2 |
| Well 9 | JOS-190129-09 | 298,3 | 6778 | 0,47 | 0,51 | 0,51 | 21,0 | 7,4 | 5,2 | 21,0 | 21,0 | 7,8 | 5,7 | 21,0 | 21,0 | 6,4 | 5,7 |
| Well 9 | JOS-180612-09 | 321,8 | 6830 | 0,41 | 0,48 | 0,47 | 21,0 | 7,0 | 4,7 | 21,0 | 21,0 | 7,8 | 5,6 | 21,0 | 21,0 | 6,5 | 5,4 |
| Well 9 | JOS-180704-09 | 308 | 6423 | 0,45 | 0,38 | 0,49 | 13,4 | 5,3 | 3,8 | 21,0 | 12,5 | 4,4 | 2,8 | 21,0 | 14,0 | 6,0 | 4,3 |

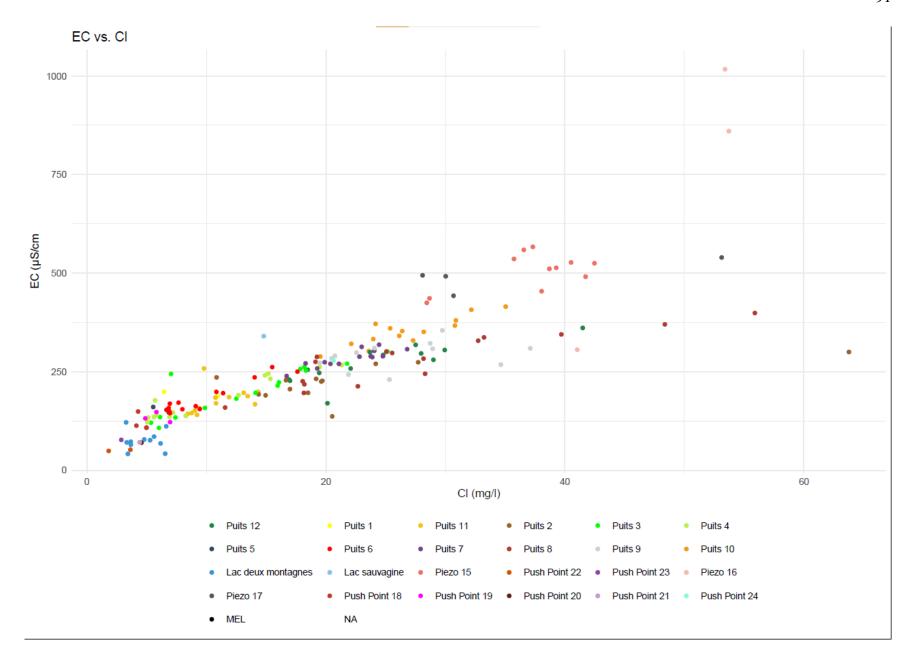
| Well | Sample | SPC (µS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | B4 | C1 | C2 | С3 | C4 |
|---------|---------------|----------------|------------|------|------|------|------|------|-----|------|------|------|-----|------|------|-----|-----|
| Well 9 | JOS-180830-09 | 309,3 | 5050 | 0,44 | 0,47 | 0,49 | 3,2 | 1,3 | 0,7 | 8,8 | 3,5 | 1,6 | 1,0 | 9,0 | 3,8 | 4,8 | 1,3 |
| Well 10 | JOS-181024-10 | 288,5 | 7010 | 0,49 | 0,51 | 0,53 | 21,0 | 12,8 | 6,4 | 21,0 | 21,0 | 12,9 | 6,6 | 21,0 | 21,0 | 7,1 | 6,8 |
| Well 10 | JOS-190626-10 | 320,7 | 7007 | 0,42 | 0,29 | 0,47 | 21,0 | 11,8 | 5,5 | 21,0 | 21,0 | 9,9 | 3,6 | 21,0 | 21,0 | 7,1 | 6,2 |
| Well 10 | JOS-190326-10 | 407 | 7363 | 0,21 | 0,28 | 0,30 | 21,0 | 21,0 | 3,6 | 21,0 | 21,0 | 21,0 | 5,2 | 21,0 | 21,0 | 8,5 | 5,6 |
| Well 10 | JOS-171206-10 | 371,3 | 6888 | 0,29 | 0,33 | 0,37 | 21,0 | 8,3 | 3,1 | 21,0 | 21,0 | 8,9 | 3,8 | 21,0 | 21,0 | 7,4 | 4,4 |
| Well 10 | JOS-180704-10 | 351 | 6632 | 0,34 | 0,26 | 0,41 | 21,2 | 6,8 | 3,0 | 21,0 | 19,8 | 5,4 | 1,6 | 21,0 | 21,0 | 6,9 | 4,0 |
| Well 10 | JOS-190129-10 | 332,7 | 6337 | 0,39 | 0,45 | 0,45 | 11,1 | 5,6 | 2,8 | 21,0 | 11,8 | 6,4 | 3,5 | 21,0 | 11,8 | 6,4 | 3,5 |
| Well 10 | JOS-180927-10 | 341 | 6393 | 0,37 | 0,33 | 0,43 | 11,7 | 5,6 | 2,6 | 21,0 | 11,2 | 5,1 | 2,1 | 21,0 | 12,6 | 6,6 | 3,5 |
| Well 10 | JOS-181219-10 | 367 | 6207 | 0,30 | 0,35 | 0,38 | 8,0 | 3,6 | 1,1 | 21,0 | 8,8 | 4,4 | 1,9 | 21,0 | 9,2 | 6,7 | 2,3 |
| Well 10 | JOS-180612-10 | 415 | 6620 | 0,19 | 0,29 | 0,29 | 16,7 | 3,5 | | 21,0 | 19,1 | 5,8 | 2,1 | 21,0 | 19,0 | 7,9 | 2,0 |
| Well 10 | JOS-180129-10 | 380 | 6150 | 0,27 | 0,27 | 0,36 | 6,8 | 2,8 | 0,4 | 21,0 | 6,8 | 2,8 | 0,4 | 21,0 | 8,2 | 6,8 | 1,8 |
| Well 10 | JOS-181129-10 | 302,2 | 4786 | 0,46 | 0,50 | 0,50 | 2,6 | 1,5 | 0,5 | 6,8 | 3,1 | 1,9 | 0,9 | 6,8 | 3,1 | 4,7 | 1,0 |
| Well 10 | JOS-180830-10 | 329 | 4678 | 0,40 | 0,42 | 0,45 | 1,5 | 0,4 | | 5,1 | 1,8 | 0,8 | | 5,5 | 2,2 | 4,8 | 0,2 |
| Well 11 | JOS-190326-11 | 167,5 | 6719 | 0,78 | 0,80 | 0,76 | 21,0 | 21,0 | 7,8 | 21,0 | 21,0 | 21,0 | 8,0 | 21,0 | 21,0 | 9,3 | 7,7 |
| Well 11 | JOS-190626-11 | 184,4 | 6096 | 0,74 | 0,68 | 0,73 | 11,7 | 11,4 | 5,7 | 21,0 | 11,2 | 10,9 | 5,2 | 21,0 | 11,6 | 7,3 | 5,6 |
| Well 11 | JOS-180927-11 | 156,8 | 5811 | 0,81 | 0,74 | 0,78 | 10,0 | 9,8 | 5,5 | 21,0 | 9,5 | 9,3 | 5,0 | 21,0 | 9,8 | 7,0 | 5,3 |
| Well 11 | JOS-181024-11 | 151,5 | 5701 | 0,82 | 0,82 | 0,79 | 9,4 | 9,2 | 5,3 | 21,0 | 9,4 | 9,3 | 5,3 | 21,0 | 9,2 | 6,9 | 5,1 |
| Well 11 | JOS-180704-11 | 188 | 5917 | 0,73 | 0,68 | 0,72 | 10,2 | 9,9 | 5,2 | 21,0 | 9,8 | 9,5 | 4,8 | 21,0 | 10,1 | 6,8 | 5,1 |
| Well 11 | JOS-190129-11 | 145 | 5395 | 0,83 | 0,80 | 0,80 | 8,0 | 7,9 | 4,8 | 18,5 | 7,8 | 7,7 | 4,6 | 18,5 | 7,8 | 6,3 | 4,6 |
| Well 11 | JOS-181219-11 | 169,9 | 5497 | 0,78 | 0,79 | 0,76 | 8,1 | 7,9 | 4,6 | 21,0 | 8,2 | 8,0 | 4,7 | 21,0 | 7,9 | 6,2 | 4,4 |
| Well 11 | JOS-180612-11 | 198,9 | 5697 | 0,71 | 0,73 | 0,70 | 8,6 | 8,4 | 4,5 | 21,0 | 8,8 | 8,6 | 4,7 | 21,0 | 8,5 | 6,3 | 4,4 |
| Well 11 | JOS-180508-11 | 258 | 5929 | 0,56 | 0,62 | 0,59 | 8,8 | 8,6 | 3,8 | 21,0 | 9,3 | 9,1 | 4,3 | 21,0 | 9,1 | 6,4 | 4,0 |
| Well 11 | JOS-181129-11 | 142,9 | 4419 | 0,84 | 0,85 | 0,81 | 5,0 | 4,9 | 3,2 | 7,5 | 5,0 | 5,0 | 3,3 | 7,2 | 4,7 | 4,6 | 3,0 |
| Well 11 | JOS-180830-11 | 185,4 | 4575 | 0,74 | 0,75 | 0,73 | 4,7 | 4,6 | 2,7 | 7,7 | 4,8 | 4,7 | 2,8 | 7,5 | 4,6 | 4,6 | 2,7 |
| Well 12 | JOS-180508-12 | 170 | 5592 | 0,77 | 0,85 | 0,76 | 8,5 | 11,1 | 4,8 | 21,0 | 9,0 | 11,6 | 5,3 | 21,0 | 8,4 | 7,3 | 4,6 |
| Well 12 | JOS-181129-12 | 280,1 | 6041 | 0,51 | 0,55 | 0,55 | 9,2 | 15,5 | 3,5 | 21,0 | 9,6 | 15,8 | 3,9 | 21,0 | 9,5 | 7,3 | 3,9 |
| Well 12 | JOS-190626-12 | 227 | 5542 | 0,64 | 0,56 | 0,65 | 7,2 | 9,6 | 3,6 | 21,0 | 6,5 | 8,9 | 2,9 | 21,0 | 7,3 | 6,6 | 3,7 |
| Well 12 | JOS-190129-12 | 318 | 6231 | 0,42 | 0,47 | 0,47 | 10,1 | 21,0 | 2,9 | 21,0 | 10,7 | 21,0 | 3,6 | 21,0 | 10,7 | 7,6 | 3,6 |
| Well 12 | JOS-180612-12 | 230,1 | 5370 | 0,63 | 0,67 | 0,64 | 6,4 | 8,3 | 3,2 | 16,6 | 6,7 | 8,6 | 3,5 | 16,4 | 6,5 | 6,3 | 3,3 |

| Well | Sample | SPC (μS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | В1 | В2 | В3 | В4 | C1 | C2 | С3 | C4 |
|-----------------------|----------------|----------------|-------------|------|------|------|------|------|-----|------|-----|------|-----|------|-----|-----|-----|
| Well 12 | JOS-190326-12 | 299,2 | 5791 | 0,47 | 0,51 | 0,51 | 6,9 | 10,3 | 2,4 | 21,0 | 7,4 | 10,9 | 3,0 | 21,0 | 7,3 | 6,8 | 2,9 |
| Well 12 | JOS-180704-12 | 247 | 5274 | 0,59 | 0,53 | 0,61 | 5,6 | 7,4 | 2,7 | 12,9 | 5,1 | 6,8 | 2,1 | 13,6 | 5,8 | 6,0 | 2,8 |
| Well 12 | JOS-180927-12 | 296,2 | 5336 | 0,47 | 0,43 | 0,51 | 4,7 | 6,5 | 1,6 | 13,2 | 4,2 | 6,0 | 1,1 | 14,2 | 5,1 | 6,2 | 2,0 |
| Well 12 | JOS-181219-12 | 305 | 4950 | 0,45 | 0,49 | 0,50 | 3,0 | 4,3 | 0,7 | 8,0 | 3,5 | 4,7 | 1,1 | 8,1 | 3,6 | 5,7 | 1,2 |
| Well 12 | JOS-181024-12 | 361 | 4700 | 0,32 | 0,34 | 0,39 | 0,4 | 1,4 | | 4,1 | 0,8 | 1,8 | | 4,8 | 1,5 | 6,0 | |
| Well 12 | JOS-180830-12 | 292,6 | 3596 | 0,48 | 0,50 | 0,52 | 0,2 | 0,7 | | 1,6 | 0,5 | 0,9 | | 1,8 | 0,7 | 4,1 | |
| Lac deux montagnes | JOS-180508-13 | 111,4 | 19 | 0,91 | 1,00 | 0,91 | | | | | | | | | | | |
| Lac deux montagnes | JOS-180612-13 | 68 | 453 | | | | | | | | | | | | | | |
| Lac deux montagnes | JOS-180704-13 | 64,7 | 21 | | | | | | | | | | | | | | |
| Lac deux montagnes | JOS-180830-13 | 78,1 | 64 | 0,99 | 1,00 | 0,99 | | | | | | | | | | | |
| Lac deux montagnes | JOS-180925-13 | 41,5 | 28 | | | | | | | | | | | | | | |
| Lac deux montagnes | JOS-181024-13 | 70,7 | 28 | | | | | | | | | | | | | | |
| Lac deux montagnes | JOS-181129-13 | 76,2 | 2355 | 1,00 | 1,00 | 1,00 | | | | | | | | | | | |
| Lac deux montagnes | JOS-190129-13 | 42 | 363,4249239 | | | | | | | | | | | | | | |
| Lac deux montagnes | JOS-190626-13 | 72,3 | 53 | | | | | | | | | | | | | | |
| Lac sauvagine | JOS-171206-14 | 340 | 835 | 0,31 | | 0,31 | 0,31 | | | | | | | | | | |
| Piezo 15 | JOS-180612-15 | 558,9 | 5331 | | | | | | | | | | | | | | |
| Piezo 15 | JOS-180704-15 | 454 | 5516 | | | | | | | | | | | | | | |
| Piezo 15 | JOS-180830-15A | 510,9 | 4308 | | | | | | | | | | | | | | |

| Well | Sample | SPC (μS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | В4 | C1 | C2 | C3 | C4 |
|------------------|----------------|----------------|------------|------|------|------|------|------|------|------|------|------|------|------|------|------------|------|
| Piezo 15 | JOS-180925-15 | 491 | 4931 | 0,01 | 0,00 | 0,01 | | | | | | | | | | | |
| Piezo 15 | JOS-181024-15 | 513,5 | 5917 | | | | | | | | | | | | | | |
| Piezo 15 | JOS-181129-15 | 527,1 | 6070 | | | | | | | | | | | | | | |
| Piezo 15 | JOS-181219-15 | 525 | 6680 | | | | | | | | | | | | | | |
| Piezo 16 | JOS-180612-16 | 305,9 | 127 | 0,45 | 0,52 | 0,45 | | | | | | | | | | | |
| Piezo 16 | JOS-180704-16 | 860 | 4654 | | | | | | | | | | | | | | |
| Piezo 16 | JOS-180830-16A | 1017 | 3593 | | | | | | | | | | | | | | |
| Piezo 17 | JOS-180925-17 | 492 | 7242 | 0,0 | 1 | 0,14 | 0,01 | | | | | | | | | | |
| Piezo 17 | JOS-181024-17 | 494,4 | 8788 | 0,00 | 0,04 | 0,00 | | | | | | | | | | | |
| Piezo 17 | JOS-181129-17 | 442,6 | 5779 | 0,12 | 0,19 | 0,12 | | | | | | | | | | | |
| Push Point | JOS-181024-18 | 113,1 | 7212 | 0,91 | 0,90 | 0,86 | 21,0 | 18,8 | 10,8 | 21,0 | 21,0 | 18,8 | 10,7 | 21,0 | 21,0 | 10,6 | 10,5 |
| 18 | | | | | | | | | | | | | | | | | |
| Push Point | JOS-181115-18A | 108 | 5359 | 0,92 | 0,93 | 0,87 | 8,4 | 6,5 | 5,3 | 18,1 | 8,5 | 6,6 | 5,3 | 17,7 | 8,1 | 5,7 | 5,0 |
| 18 Push Point | | | | | | | | | | | | | | | | | |
| 18 | JOS-181115-18B | 108 | 5262 | 0,92 | 0,93 | 0,87 | 8,0 | 6,3 | 5,1 | 15,7 | 8,1 | 6,3 | 5,1 | 15,4 | 7,7 | 5,5 | |
| Push Point | | | | | | | | | | | | | | | | | |
| 18 | JOS-190704-18 | 159 | 4057 | 0,80 | 0,88 | 0,78 | 3,9 | 3,1 | 2,5 | 6,1 | 4,4 | 3,6 | 3,0 | 5,5 | 3,7 | 3,5 | 2,3 |
| Push Point | | | | | | | | | | | | | | | | | |
| 19 | JOS-181024-19 | 147,6 | 7298 | 0,83 | 0,83 | 0,80 | 21,0 | 35,7 | 10,7 | 21,0 | 21,0 | 35,7 | 10,7 | 21,0 | 21,0 | 10,0 | |
| Push Point | JOS-181115-19 | 131,5 | 6851 | 0,87 | 0,88 | 0,83 | 21,0 | 111 | 0.0 | 21.0 | 21,0 | 14,5 | 0.0 | 21,0 | 21.0 | 9,0 | 8,6 |
| 19 | 102-181112-19 | 131,5 | 0831 | 0,87 | 0,88 | 0,83 | 21,0 | 14,4 | 8,9 | 21,0 | 21,0 | 14,5 | 9,0 | 21,0 | 21,0 | 9,0 | 8,0 |
| Push Point | 105 100704 10 | 122.2 | F.CO.2 | 0,89 | 1.00 | 0.05 | 0.2 | 7,3 | 5,6 | 21.0 | 10,0 | 8,0 | 6,2 | 21,0 | 9,1 | <i>c</i> 1 | |
| 19 | JOS-190704-19 | 122,2 | 5603 | 0,89 | 1,00 | 0,85 | 9,3 | 7,3 | ٥,٥ | 21,0 | 10,0 | ٥,0 | 0,2 | 21,0 | 9,1 | 6,1 | |
| Push Point | JOS-181115-20 | 70 | 3986 | | | | | | | | | | 6,3 | 4,7 | 3,9 | 3,3 | |
| 20 | 103-101113-20 | 70 | 3300 | | | | | | | | | | 0,3 | 4,/ | 3,3 | ی,ی | |
| Push Point 21 | JOS-181115-21 | 71 | 6770 | | | | | | | | | | 21,0 | 21,0 | 11,5 | 9,1 | |

| Well | Sample | SPC (μS/cm) | Rn (Bq/m³) | MR1 | MR2 | MR3 | A2 | А3 | A4 | B1 | В2 | В3 | В4 | C1 | C2 | C3 | C4 |
|------------------|---------------|----------------|------------|------|------|------|-----|------|------|------|-----|------|------|------|------|------|------|
| Push Point 22 | JOS-181129-22 | 49 | 8263 | | | | | | | | | | 21,0 | 21,0 | 21,0 | 21,0 | |
| Push Point 22 | JOS-190326-22 | 52 | 6076 | | | | | | | | | | 21,0 | 13,0 | 9,2 | 7,2 | |
| Push Point 23 | JOS-181129-23 | 77,1 | 5318 | 1,00 | 1,00 | 0,93 | 8,7 | 21,0 | 21,0 | 17,7 | 8,7 | 21,0 | 21,0 | 17,3 | 8,3 | 21,0 | 21,0 |
| Push Point 24 | JOS-190704-24 | 278 | 5643 | 0,52 | 0,55 | 0,55 | 6,6 | 2,0 | 2,7 | 21,0 | 6,9 | 2,4 | 3,0 | 21,0 | 6,9 | 4,4 | 3,0 |

APPENDIX B CHLORIDE VS. EC



APPENDIX C IBF SITES GLOBALMAR DATABASE

Distribution of IBF sites based on their distance

