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CARACTÉRISATION INTÉGRÉE D’UN AQUIFÈRE GRANULAIRE POUR L’ÉVALUATION DES PROCESSUS GÉOCHIMIQUES INFLUENÇANT L’ATTÉNUATION NATURELLE D’UN PANACHE DE LIXIVIAT

Par

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Résumé

Cette thèse aborde trois problèmes d'intérêt général en hydrogéologie : 1) la difficulté d'évaluer, avec certitude, l'efficacité de l'atténuation naturelle en tant que mode de gestion d'un site contaminé; 2) le besoin de développer des approches de caractérisation hydrogéologique efficaces qui permettent de définir l'hétérogénéité des aquifères en 3D afin de soutenir le développement de modèles conceptuels et numériques représentatifs de l'écoulement de l'eau souterraine; et 3) l'intérêt de disposer de données de terrain variées et indépendantes pour vérifier les résultats obtenus des modèles numériques sur le devenir des contaminants dans l'eau souterraine. Les travaux réalisés devraient permettre d'améliorer les approches utilisées pour atteindre une bonne compréhension des systèmes aquifères, particulièrement dans le but de développer des modèles numériques qui permettent de prédire avec justesse la migration des contaminants et leurs impacts sur la qualité des ressources en eau souterraine et les milieux récepteurs.

Les étapes recommandées par les agences gouvernementales pour définir l'atténuation naturelle en tant que mode de gestion sont laborieuses et les travaux de terrain et de laboratoire requis sont nombreux, spécialisés et couteux. Le principal défi de ce type d'évaluation repose sur le fait que les processus d'atténuation naturelle devant être définis sont spécifiques à chaque site, car ils dépendent des conditions hydrogéologiques et hydrogéochimiques de la nature de la source de contamination. En fait, l'hétérogénéité d'un aquifère contrôle en grande partie la migration des contaminants et sa méconnaissance est souvent à l'origine des échecs des mesures de réhabilitation. De plus, la prédiction du comportement à long terme des contaminants requiert l'utilisation de modèles numériques représentatifs. Par contre, malgré la capacité actuelle des simulateurs numériques à intégrer un grand nombre d'informations pertinentes, les méthodes de caractérisation hydrogéologique conventionnelles ne permettent que l'acquisition partielle des données requises pour développer des modèles détaillés représentatifs de la réalité.

Les travaux de recherche réalisés dans le cadre de la présente thèse contribuent à combler ces lacunes par : 1) le développement d'une approche de caractérisation intégrée qui définit adéquatement l'hétérogénéité des aquifères ainsi que les conditions hydrogéologiques spécifiques à un site contaminé, 2) la démonstration de l'intérêt d'intégrer des données de

sources variées pour développer des modèles conceptuel et numérique de l'écoulement et du transport de masse contraint avec des données géochimiques et, 3) la démonstration de cette approche par son application à un ancien site d'enfouissement sanitaire afin de statuer, avec peu d'incertitude, sur l'efficacité des processus d'atténuation naturelle prêtées à ce site. L'approche de caractérisation intègre des méthodes indirectes et continues, telles que la géophysique de surface et les méthodes par enfoncement, avec des méthodes directes et ponctuelles de haute résolution comme les mesures hydrauliques et géochimiques dans les puits d'observation. Le choix des méthodes à combiner et la séquence logique des travaux utilisés permettent d'acquérir, de façon efficace, un grand nombre d'informations détaillées et spatialement bien distribuées.

Les travaux de terrain ont été réalisés dans le sous-bassin versant de 12 km^2 entourant l'ancien site d'enfouissement de Saint-Lambert-de-Lauzon. Les résultats obtenus par la caractérisation démontrent une grande variabilité spatiale des conditions physiques et géochimiques du système aquifère étudié. La combinaison des levés géophysiques de surface avec les sondages par enfoncement a permis, notamment, de définir de façon détaillée : 1) la structure de l'aquifère (limites, stratigraphie et hydrofaciès), 2) l'étendue du panache de lixiviat, 3) la distribution spatiale des contextes hydrogéologiques ainsi que les voies d'écoulement et de migration du lixiviat, et 4) les conditions géochimiques dans l'aquifère. Les données géochimiques ont été primordiales pour raffiner le modèle conceptuel de l'écoulement de l'eau souterraine en donnant des indications sur les zones de recharge et d'émergence, la magnitude de la recharge déduite à partir des profils d'âge $^{3}H/^{3}He$ ainsi que des évidences sur les points d'échange entre l'aquifère granulaire peu profond et l'aquifère rocheux sous-jacent. L'ensemble de ces conditions contrôlent l'écoulement de l'eau souterraine ainsi que la migration et l'atténuation du panache de lixiviat. La caractérisation détaillée, et la compréhension globale de l'aquifère et des processus d'atténuation en découlant, n'auraient pas été possibles au niveau du temps et des ressources impartis avec les méthodes conventionnelles. De plus, l'approche de caractérisation intégrée peut être adaptée à d'autres régions d'étude présentant tout type d'environnement hydrogéologique et d'échelle géographique.

Chacun des quatre articles scientifiques réunis dans la présente thèse aborde des questions scientifiques d'intérêt et fait des contributions originales. Le premier article décrit le développement et l'application d'une approche de caractérisation de l'hétérogénéité des

aquifères. Ces travaux se distinguent parce qu’ils portent sur un site de terrain réel (et non pas un cas synthétique), sont réalisés à une échelle régionale (plutôt que très locale) et utilisent de multiples méthodes dont le rôle et la séquence d’application sont bien définis. Le deuxième article porte sur l’utilisation d’indicateurs superficiels pour identifier les zones (et taux) de recharge et d’émergence et l’interaction entre les eaux souterraines et de surface. L’étude est originale par l’usage de traceurs et de méthodes multiples qui permettent l’étude des taux et mécanismes de recharge sur une petite région, par rapport aux études régionales, notamment le rôle des conditions du sol et leurs effets sur la géochimie de l’eau souterraine. Le troisième article documente une étude des facteurs à l’origine des conditions géochimiques hétérogènes d’un aquifère et leur incidence sur l’atténuation naturelle de contaminants. Cette étude se distingue par l’utilisation de méthodes originales de caractérisation chimique des sédiments de l’aquifère et des sols et par l’établissement de relations entre ces caractéristiques et la géochimie de l’eau souterraine qui contrôle la dégradation du lixiviat. Enfin, le dernier article présente l’évaluation des effets de l’hétérogénéité hydrogéologique et géochimique sur l’atténuation naturelle du lixiviat émis par un site d’enfouissement. Ces travaux sont originaux par l’utilisation complémentaire de la modélisation numérique et des données géochimiques; méthodes qui permettent d’évaluer les effets des conditions géochimiques variables sur deux voies distinctes d’atténuation naturelle du lixiviat.

Globalement, les travaux de recherche doctoraux réalisés ont fait l’usage de méthodes variées, ont permis l’acquisition d’un ensemble de données exceptionnel, et ont considéré des échelles originales d’étude. Ces travaux ont ainsi permis d’avancer les méthodes et les connaissances sur des questions de fond en hydrogéologie, notamment le développement des modèles conceptuels, la définition de l’hétérogénéité physique et chimique des aquifères, l’incertitude des modèles numériques, et les processus géochimiques influençant les conditions naturelles des eaux souterraines et les processus d’atténuation des contaminants.

Mots-clés : Caractérisation hydrogéologique, modélisation numérique, géochimie de l’eau souterraine, atténuation naturelle, site d’enfouissement sanitaire.

Abstract

This thesis tackles three questions of general interest in hydrogeology: 1) the difficulty to assess, with certainty, the efficiency of monitored natural attenuation as a viable approach to contaminated site management; 2) the need to develop efficient characterization methods allowing the definition of aquifer heterogeneity in 3D to allow the development of representative numerical models; and 3) the relevance of independent field data to verify answers obtained from numerical models that are used to predict the fate of contaminants in groundwater. This work should lead to improve the approaches used to gain a complete understanding of aquifer systems; especially with the goal to develop numerical models for predicting contaminant migration and its impacts on groundwater resources and other receptors.

The recommended steps by regulating agencies to assess monitored natural attenuation as an environmental management approach are laborious and include several field and laboratory analyses that are specialized and costly. The main challenge in evaluating monitored natural attenuation lays in the fact that attenuation processes are site-specific since they depend on the hydrogeological conditions and the source of contamination. Aquifer heterogeneity largely controls the migration of contaminants and poor understanding of it is often responsible for the failure of remediation systems. Moreover, the prediction of the long-term fate of contaminants requires the use of realistic numerical simulations. However, despite the present capabilities of numerical simulators to integrate a large amount of relevant data, conventional characterization methods do not provide the data required to develop representative models of aquifer heterogeneity.

The work carried out as part of this doctoral research addresses these issues by: 1) proposing a characterization approach to efficiently define aquifer heterogeneity and specific hydrogeological conditions at contaminated sites; 2) demonstrating the interest of integrating multi-source data to develop a representative conceptual and numerical model of groundwater flow and mass transport constrained by geochemical data; 3) the application of the developed approaches to a decommissioned landfill with the goal of assessing the efficiency of natural attenuation with certainty. The proposed methodology suggests the integration of indirect continuous data (laterally and vertically), such as surface geophysics and direct push soundings,

with high resolution direct point data such as hydraulic tests and geochemical sampling that were used to constrain and correlate the indirect data. The choice of the methods to be combined and the logical field work sequence allow the acquisition of a wide range of information that is spatially well distributed, while minimizing the number of measuring sites.

Field work was carried out in the subwatershed encompassing the decommissioned landfill of Saint-Lambert-de-Lauzon. The integration of the multiple data sources revealed in great detail: 1) the aquifer structure (aquifer boundaries, stratigraphy and hydrofacies); 2) the leachate plume extent; 3) the spatial distribution of hydrogeological contexts as well as groundwater flow patterns and leachate migration paths; and 4) groundwater geochemical conditions found throughout the aquifer. The geochemical data were key in refining the conceptual model of groundwater flow by identifying locations of recharge and discharge areas, the magnitude of recharge inferred by $^{3}H/^{3}He$ age profiles, and evidence of points of exchange between the aquitard separating the shallow granular aquifer and the underlying bedrock aquifer. These conditions control groundwater flow, contaminant migration and natural attenuation of the plume. The detailed characterization allowed the understanding of a complex groundwater flow system, including site-specific natural attenuation processes, could not have been achieved with conventional approaches using the available time and resources. This characterization approach was found to be time and cost efficient and could be adapted to other types of hydrogeological environments and scales of study area.

Each of the four scientific papers of the thesis deals with scientific questions of interest and makes original contributions. The first paper describes the development and application of a workflow for the characterization of aquifer heterogeneity. This thesis departs from previous studies by dealing with a real field site (rather than a synthetic case), and is carried out at a regional scale (rather than a small site) and uses multiple methods whose role and sequence of application are well-defined. The second paper describes the use of multiple shallow indicators to identified zones (and rates) of recharge and discharge as well as the interaction between groundwater and surface water. This study makes an original use of tracers as well as multiple methods to assess the rates and mechanisms of recharge over a relatively small area, compared to regional studies, especially the role of soils and surface conditions and their impact on groundwater geochemistry. The third paper documents a study of the factors controlling

heterogeneous geochemical conditions in an aquifer and their impact on natural attenuation of contaminants. This study makes original use of aquifer sediments and soil chemical characterization and by defining their relations with groundwater geochemistry, which controls leachate degradation. Finally, the fourth paper presents an assessment of the effects of hydrogeological and geochemical heterogeneity on landfill leachate natural attenuation. This work makes an original complementary use of numerical modeling and geochemical data, which allow the evaluation of the effects of different geochemical conditions on two distinct paths of leachate natural attenuation.

Globally, this doctoral research made use of varied methods, allowed the acquisition of an exceptional dataset, and considered original scales of study. This work advanced methods of study and knowledge about a few fundamental hydrogeological issues, notably the development of conceptual models, the definition of the physical and chemical heterogeneity of aquifers, the uncertainty of numerical models, and the geochemical processes controlling natural groundwater geochemical conditions as well as the natural attenuation of contaminants.

Keywords: Hydrogeological characterization, numerical modelling, groundwater geochemistry, natural attenuation, sanitary landfill.

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Partie 1

Mise en contexte

Chapitre 1

INTRODUCTION

1.1 Contexte, problématique et objectifs du projet doctoral

L'INRS-ETE a établi un partenariat avec la Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière (RIGDCC) pour vérifier l'efficacité de l'atténuation naturelle comme mode de gestion de l'ancien lieu d'enfouissement sanitaire (LES) de Saint-Lambert-de-Lauzon. Cette entente visait principalement l'investigation de l'aquifère granulaire entourant l'ancien site. Plusieurs étudiants de 2^{e} et 3^{e} cycles et stagiaires ont participé aux activités scientifiques plus larges découlant de ces travaux et ont bénéficié des données collectées dans le cadre du projet. Les travaux réalisés par l'INRS-ETE pour la RIGDCC ont débuté en 2006 et se sont poursuivis jusqu'à la fin 2012. L'ancien LES, localisé sur une propriété d'environ 1 km^{2}, est situé à 35 km au sud de Québec et a été exploité de 1974 à 1997. En 24 ans, 900 000 tonnes de déchets d'origines municipale, agricole et industrielle, provenant de 15 municipalités différentes, ont été déposées sur un aquifère granulaire composé de silt, de sable et de gravier. Les anciens LES peuvent causer la contamination de l'eau de surface et de l'eau souterraine par l'émission et la migration des eaux de lixiviation qu'ils produisent. Avec l'enfouissement, la décomposition des déchets entraîne la consommation rapide de l'oxygène et produit un lixiviat dans un environnement extrêmement réducteur. Le lixiviat produit, dépourvu d'oxygène, est aussi distinct par ses concentrations importantes en composés organiques et inorganiques, et contraste avec les conditions physico-chimiques naturelles normalement rencontrées dans les aquifères. Conséquemment, lors de sa migration avec l'écoulement de l'eau souterraine, le lixiviat peut être soumis à des processus d'atténuation naturelle d'ordres chimique, biologique et physique qui, sans intervention humaine, réduisent la masse, la toxicité, la mobilité, le volume ou la concentration des contaminants au-delà de ce qui pourrait être obtenu par de simples effets de dilution. L'évaluation de l'atténuation naturelle et son acceptabilité en tant que mode de gestion est complexe et requiert une bonne compréhension du système d'écoulement de l'eau souterraine et des processus d'atténuation naturelle spécifiques au site impliqué. L'importance des ces processus sont dépendents de plusieurs facteurs, dont la géologie du milieu en place et la géochimie de la source de contamination.

Saisir la complexité des systèmes d'écoulement et définir les différentes caractéristiques d'un site contaminé représentent de grands défis techniques et scientifiques. Les échecs des mesures de gestion ou de réhabilitation des sites contaminés sont souvent dus à une compréhension insuffisante des aquifères et en particulier des effets liés à leur hétérogénéité. Au cours des dernières décennies, le développement des capacités des simulateurs numériques a permis la réalisation de modèles donnant une représentation de plus en plus détaillée de systèmes d'écoulement complexes. Par contre, les méthodes conventionnelles de caractérisation hydrogéologique ne permettent pas d'acquérir toutes les données requises pour développer des modèles sufisamment détaillés pour permettre de réaliser des simulations plus représentatives. De plus, les résultats produits par ces modèles hydrogéologiques sophistiqués ne sont généralement vérifiés qu'au moyen de mesures de charges hydrauliques dans les puits, ne tenant pas compte, ainsi, d'autres paramètres indépendants importants tels que des données hydrogéochimiques. Les données géochimiques peuvent fournir des informations indépendantes qui, combinées aux mesures hydrauliques, permettraient de mieux contraindre un modèle numérique et d'obtenir des réponses plus représentatives.

Ce projet doctoral vise à identifier l'efficacité des processus d'atténuation naturelle d'un ancien site d'enfouissement sanitaire tout en approfondissant des aspects scientifiques et techniques d'intérêt en hydrogéologie. L'objectif général était de développer et d'appliquer une approche de caractérisation hydrogéologique intégrée et efficace. Cette caractérisation doit fournir les informations nécessaires à l'élaboration d'un modèle conceptuel détaillé du fonctionnement du système aquifère. Enfin, cette compréhension du système aquifère doit permettre l'évaluation de l'atténuation naturelle comme mode de gestion du panache de lixiviat émis par le site d'enfouissement sanitaire. Une telle approche de caractérisation devrait être applicable à d'autres sites contaminés localisés dans des environnements géologiques complexes. La Figure 1.1 montre les étapes de réalisation des travaux en relation avec les objectifs secondaires du projet. Découlant de l'objectif général, un objectif secondaire visait à caractériser l'hétérogénéité du système aquifère de la région d'étude en 3D afin de développer des modèles numériques plus représentatifs de l'écoulement de l'eau souterraine et du transport de masse afin d'améliorer la prédiction du devenir des panaches de contamination. L'autre objectif secondaire était d'évaluer la recharge et l'interaction entre l'eau de surface et souterraine à l'aide d'indicateurs mesurés dans la partie superficielle de l'aquifère. Le dernier objectif secondaire était de caractériser et comprendre les processus géochimiques qui contrôlent les conditions géochimiques naturelles de modèles conceptuels ainsi qu'un modèle numérique en 2D du système aquifère ont été développés et utilisés pour représenter l'écoulement et le transport de masse et d'âge.

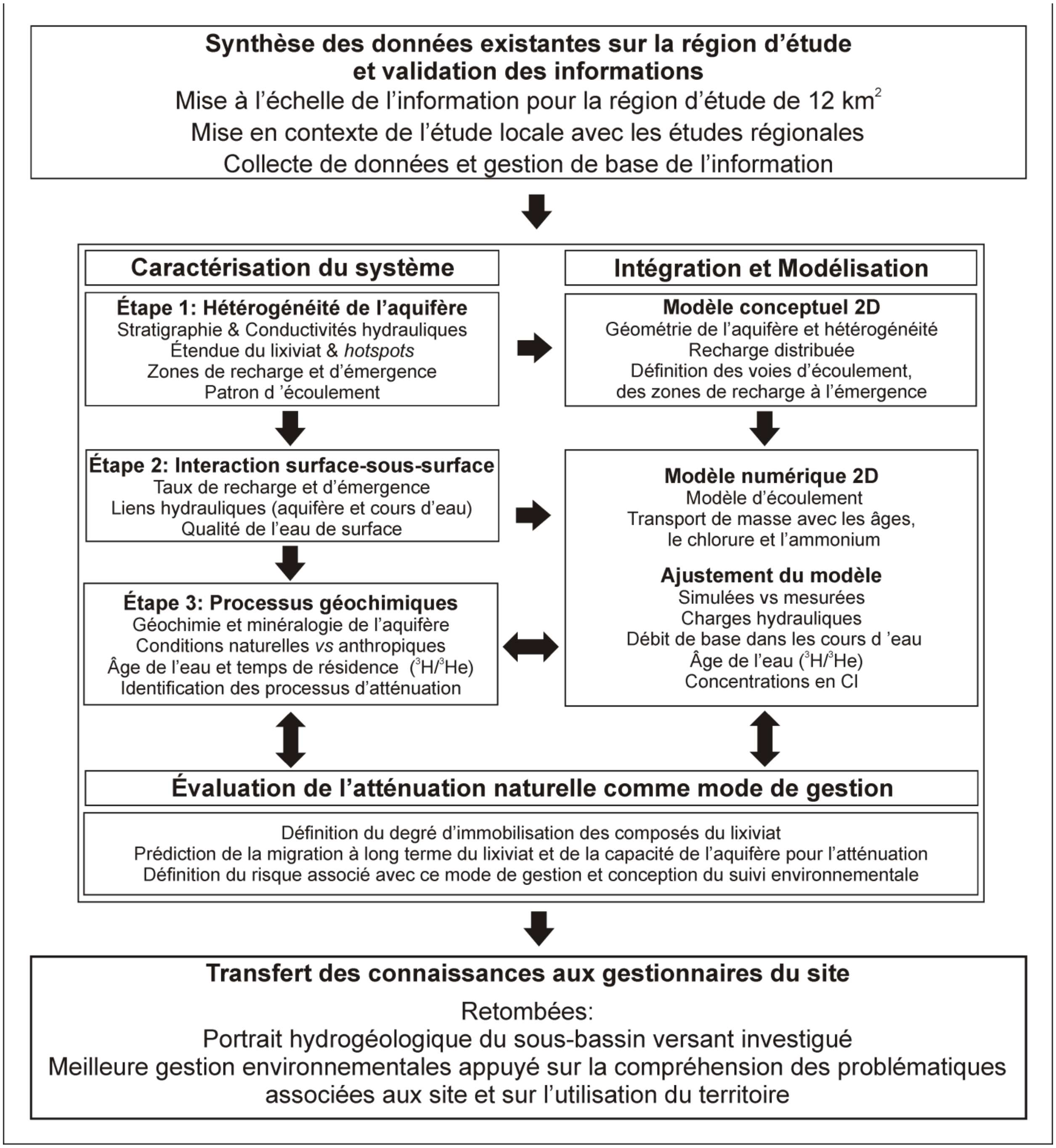
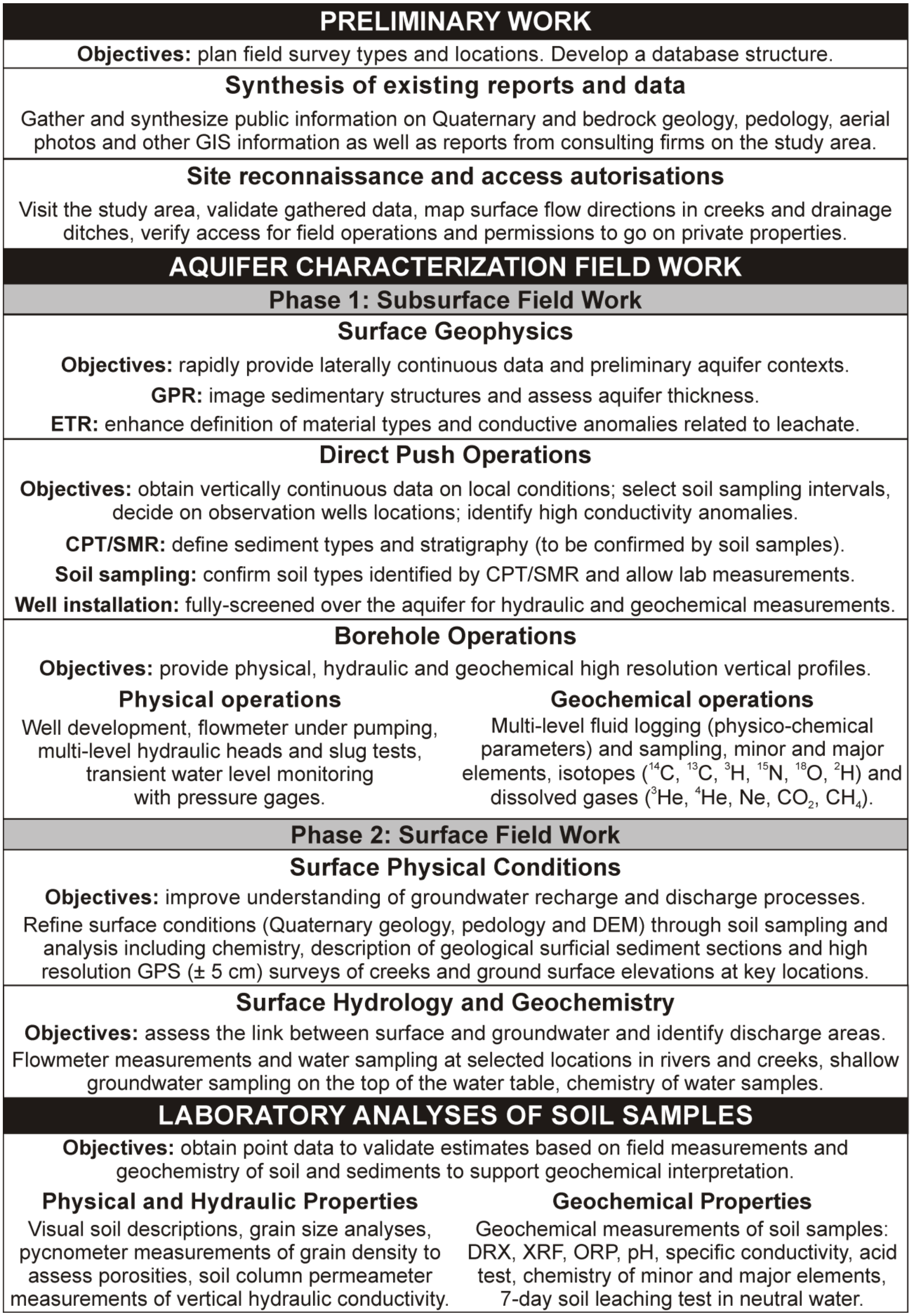


Figure 1.1 : Approche de caractérisation intégrée

Ainsi, dans le cadre des travaux réalisés pour cette thèse, des avancées techniques significatives reliées au développement d'approches novatrices de caractérisation des aquifères ont été réalisées. De plus, d'autres avancées au niveau des connaissances scientifiques ont permis de diagnostiquer les conditions spécifiques entourant l'ancien site d'enfouissement sanitaire de Saint-Lambert-de-Lauzon, d'évaluer les processus d'atténuation naturelle et d'émettre des recommandations pour le suivi et la gestion environnementale dans le futur.

1.2 Approche novatrice générale, méthodologie et retombées scientifiques

L'approche de caractérisation proposée combine des méthodes de terrain connues et éprouvées. L'idée générale est de corréler et de contraindre plusieurs types de mesures afin d'obtenir l'information la plus précise et la plus détaillée possible. En ce sens, la séquence des travaux est primordiale (Figures 1.2 et 1.3) et elle suit une approche similaire à celle utilisée pour la caractérisation des réservoirs pétroliers, ce qui n'est pas commun pour la caractérisation hydrogéologique (Bradford et Babcock 2013). Par exemple, la géophysique de surface réalisée en premier lieu donne une image quasi-continue de la sous-surface et expose sa variabilité. Sur la base de ces résultats, des sondages d'autres types tels que l'échantillonnage de sols et les sondages par enfoncement peuvent être effectués aux endroits les plus pertinents en fonction du contexte défini par la géophysique. Des mesures de terrain indirectes et continues (verticalement et latéralement), telle que la géophysique de surface et les sondages par enfoncement, peuvent être contraintes et corrélées avec des mesures directes et ponctuelles de haute résolution comme des valeurs précises mesurées sur des échantillons de sol ou des mesures de haute résolution effectuées dans les puits d'observation. Ceci permet de suivre une approche séquentielle allant du grossier au détaillé afin de minimiser le nombre de sites et de levés, tout en maximisant la qualité et la pertinence des informations acquises. Les travaux visant à définir l'hétérogénéité de l'aquifère incorporent aussi plusieurs méthodes de caractérisation: des méthodes de géophysique de surface, des méthodes par enfoncement avec une foreuse géotechnique, et des mesures hydrauliques et géochimiques de haute résolution dans les puits d'observations. Plusieurs autres levés de terrain supplémentaires ont été réalisés dans le but d'évaluer les patrons de recharge, d'écoulement et d'émergence de l'eau souterraine qui peuvent influencer la migration et l'atténuation du panache de lixiviat.



PRELIMINARY WORK

Objectives: plan field survey types and locations. Develop a database structure.

Synthesis of existing reports and data

Gather and synthesize public information on Quaternary and bedrock geology, pedology, aerial photos and other GIS information as well as reports from consulting firms on the study area.

Site reconnaissance and access autorisations

Visit the study area, validate gathered data, map surface flow directions in creeks and drainage ditches, verify access for field operations and permissions to go on private properties.

AQUIFER CHARACTERIZATION FIELD WORK

Phase 1: Subsurface Field Work

Surface Geophysics

Objectives: rapidly provide laterally continuous data and preliminary aquifer contexts.

GPR: image sedimentary structures and assess aquifer thickness.

ETR: enhance definition of material types and conductive anomalies related to leachate.

Direct Push Operations

Objectives: obtain vertically continuous data on local conditions; select soil sampling intervals, decide on observation wells locations; identify high conductivity anomalies.

CPT/SMR: define sediment types and stratigraphy (to be confirmed by soil samples).

Soil sampling: confirm soil types identified by CPT/SMR and allow lab measurements.

Well installation: fully-screened over the aquifer for hydraulic and geochemical measurements.

Borehole Operations

Objectives: provide physical, hydraulic and geochemical high resolution vertical profiles.

Physical operations

Well development, flowmeter under pumping, multi-level hydraulic heads and slug tests, transient water level monitoring with pressure gages.

Geochemical operations

Multi-level fluid logging (physico-chemical parameters) and sampling, minor and major elements, isotopes (^{14}C, ^{13}C, ^{3}H, ^{15}N, ^{18}O, ^{2}H) and dissolved gases (^{3}He, ^{4}He, Ne, CO_2, CH_4).

Phase 2: Surface Field Work

Surface Physical Conditions

Objectives: improve understanding of groundwater recharge and discharge processes.

Refine surface conditions (Quaternary geology, pedology and DEM) through soil sampling and analysis including chemistry, description of geological surficial sediment sections and high resolution GPS (± 5 cm) surveys of creeks and ground surface elevations at key locations.

Surface Hydrology and Geochemistry

Objectives: assess the link between surface and groundwater and identify discharge areas.

Flowmeter measurements and water sampling at selected locations in rivers and creeks, shallow groundwater sampling on the top of the water table, chemistry of water samples.

LABORATORY ANALYSES OF SOIL SAMPLES

Objectives: obtain point data to validate estimates based on field measurements and geochemistry of soil and sediments to support geochemical interpretation.

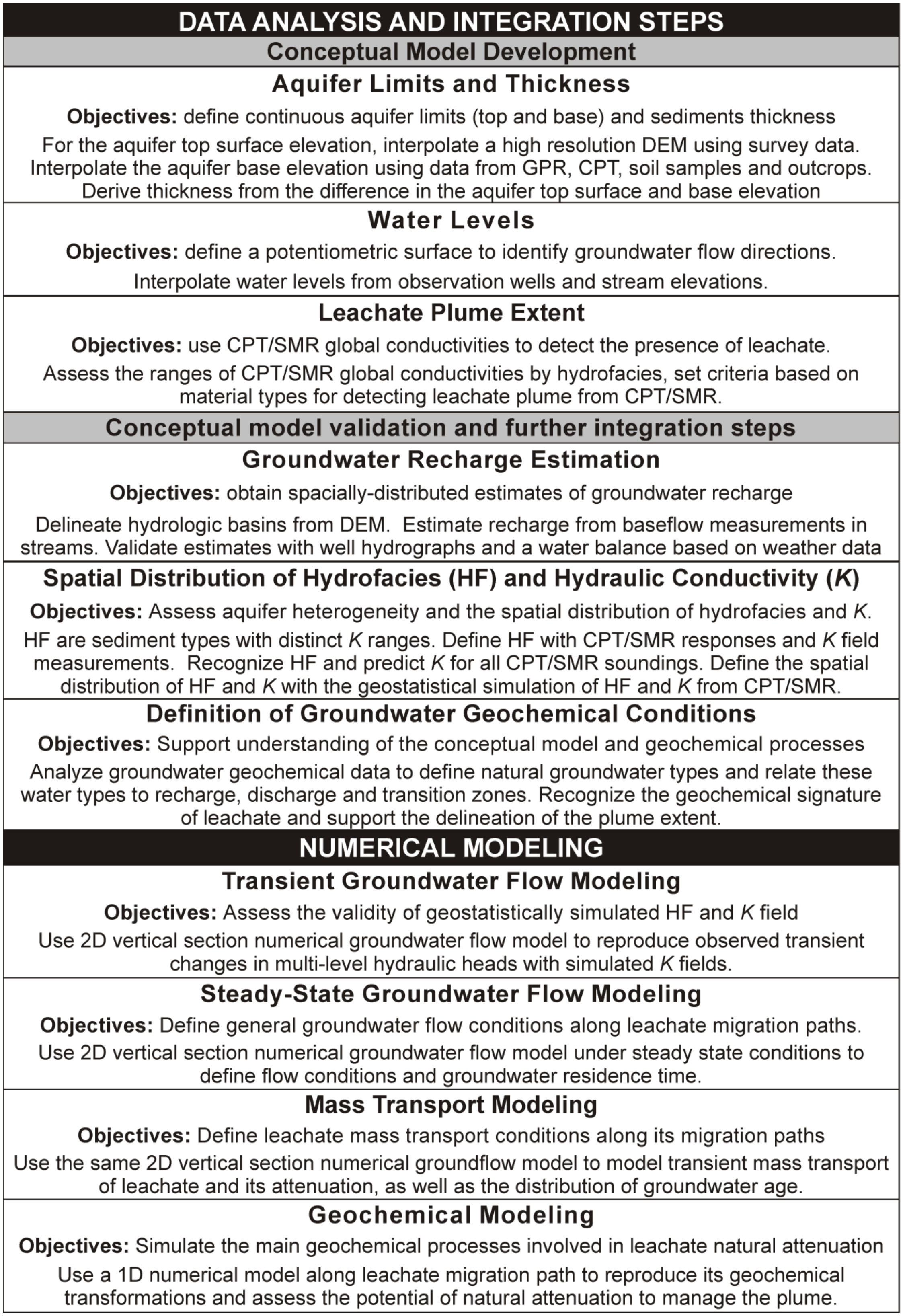
Physical and Hydraulic Properties

Visual soil descriptions, grain size analyses, pycnometer measurements of grain density to assess porosities, soil column permeameter measurements of vertical hydraulic conductivity.

Geochemical Properties

Geochemical measurements of soil samples: DRX, XRF, ORP, pH, specific conductivity, acid test, chemistry of minor and major elements, 7-day soil leaching test in neutral water.

Figure 1.2 : Séquence des travaux de caractérisation



DATA ANALYSIS AND INTEGRATION STEPS

Conceptual Model Development

Aquifer Limits and Thickness

Objectives: define continuous aquifer limits (top and base) and sediments thickness

For the aquifer top surface elevation, interpolate a high resolution DEM using survey data. Interpolate the aquifer base elevation using data from GPR, CPT, soil samples and outcrops. Derive thickness from the difference in the aquifer top surface and base elevation

Water Levels

Objectives: define a potentiometric surface to identify groundwater flow directions.

Interpolate water levels from observation wells and stream elevations.

Leachate Plume Extent

Objectives: use CPT/SMR global conductivities to detect the presence of leachate.

Assess the ranges of CPT/SMR global conductivities by hydrofacies, set criteria based on material types for detecting leachate plume from CPT/SMR.

Conceptual model validation and further integration steps

Groundwater Recharge Estimation

Objectives: obtain spacially-distributed estimates of groundwater recharge

Delineate hydrologic basins from DEM. Estimate recharge from baseflow measurements in streams. Validate estimates with well hydrographs and a water balance based on weather data

Spatial Distribution of Hydrofacies (HF) and Hydraulic Conductivity (*K*)

Objectives: Assess aquifer heterogeneity and the spatial distribution of hydrofacies and *K*.

HF are sediment types with distinct *K* ranges. Define HF with CPT/SMR responses and *K* field measurements. Recognize HF and predict *K* for all CPT/SMR soundings. Define the spatial distribution of HF and *K* with the geostatistical simulation of HF and *K* from CPT/SMR.

Definition of Groundwater Geochemical Conditions

Objectives: Support understanding of the conceptual model and geochemical processes

Analyze groundwater geochemical data to define natural groundwater types and relate these water types to recharge, discharge and transition zones. Recognize the geochemical signature of leachate and support the delineation of the plume extent.

NUMERICAL MODELING

Transient Groundwater Flow Modeling

Objectives: Assess the validity of geostatistically simulated HF and *K* field

Use 2D vertical section numerical groundwater flow model to reproduce observed transient changes in multi-level hydraulic heads with simulated *K* fields.

Steady-State Groundwater Flow Modeling

Objectives: Define general groundwater flow conditions along leachate migration paths.

Use 2D vertical section numerical groundwater flow model under steady state conditions to define flow conditions and groundwater residence time.

Mass Transport Modeling

Objectives: Define leachate mass transport conditions along its migration paths

Use the same 2D vertical section numerical groundflow model to model transient mass transport of leachate and its attenuation, as well as the distribution of groundwater age.

Geochemical Modeling

Objectives: Simulate the main geochemical processes involved in leachate natural attenuation

Use a 1D numerical model along leachate migration path to reproduce its geochemical transformations and assess the potential of natural attenuation to manage the plume.

Figure 1.3 : Séquence des travaux d'intégration des données et de modélisation

Les travaux de caractérisation intégrés visant à définir les conditions spécifiques au site ont été réalisés en trois phases distinctes (Figure 1.1): 1) caractérisation hydrogéologique de l'hétérogénéité de la sous-surface; 2) caractérisation de la qualité des eaux de surface et des processus gouvernant l'interaction entre les eaux de surface et souterraines; et 3) caractérisation géochimique de la matrice de l'aquifère et de l'eau souterraine. Chacune de ces trois étapes, successives et complémentaires, comporte des aspects originaux dans les méthodes employées et dans l'apport scientifique des résultats obtenus, constituant un article de la présente thèse. Il est à noter que le volet de caractérisation hydraulique a été réalisé par Daniel Paradis dans le cadre de son projet de doctorat en combinant les méthodes hydrauliques et les sondages au piézocône avec des mesures électriques dans le but de mieux représenter la distribution spatiale des hydrofaciès et de leurs propriétés hydrauliques. Ces propriétés ont été intégrées dans le modèle numérique développé dans le cadre du projet doctoral de cette thèse. De plus, Christine Bélanger, a réalisé les levés de résistivité et leur inversion dans le cadre de sa maîtrise à l'INRS-ETE, et les résultats de ces levés ont été utilisés par l'auteure pour confirmer le cadre géologique de l'aquifère granulaire investigué.

Les travaux de Daniel Paradis et de Christine Bélanger ont été réalisés en relation avec des objectifs de recherche plus large de l'équipe de René Lefebvre. L'ensemble de ces travaux a fourni un important jeu de données multi-sources distribuées sur l'ensemble de la région d'étude. L'intégration de ces données par l'auteure, grâce à la géostatistique, a permis de mieux définir la géométrie de l'aquifère en 3D, l'hétérogénéité des matériaux granulaires de l'aquifère, les voies et les dynamiques d'écoulement de l'eau souterraine, l'étendue du panache de lixiviat et ses voies de migration ainsi que les conditions géochimiques et les processus d'atténuation naturelle répartis dans la zone d'étude. Ce niveau de détails découlant de la caractérisation n'aurait pas pu être atteint avec des méthodes conventionnelles dans le temps requis avec les ressources disponibles. Ces données de caractérisation ont servi au développement d'un modèle numérique d'écoulement et de transport de masse, validé avec des données géochimiques indépendantes pour l'évaluation de l'atténuation naturelle du panache de lixiviat. Tous les paramètres du modèle numérique de l'écoulement ont été déterminés selon les résultats de la caractérisation intégrée. L'assignation des différents paramètres du modèle, notamment les conductivités hydrauliques des différentes couches, les conditions aux limites (incluant les valeurs d'infiltration distribuée) et les patrons d'écoulement, a été basée en bonne partie sur le modèle conceptuel d'écoulement développé suite aux travaux de caractérisation. Ces valeurs ont ensuite été ajustées selon le degré de complexité choisi pour le modèle.

Cette méthodologie a été efficace en termes de ressources et de temps et elle peut être adaptée à tout type d'environnement hydrogéologique et d'échelle géographique. Le développement d'un modèle conceptuel détaillé de l'eau souterraine a été orienté davantage par l'interprétation des données géochimiques, ce qui démontre qu'un modèle conceptuel développé à partir de levés de terrain détaillés et représentatifs est essentiel à la conception rigoureuse d'un modèle numérique et à ses capacités de prédiction. Plus précisément, les données géochimiques sur les constituants inorganiques ont été primordiales pour raffiner le modèle conceptuel de l'écoulement de l'eau souterraine en donnant des indications sur les zones de recharge et d'émergence, et des évidences sur la discontinuité entre l'aquifère granulaire peu profond et l'aquifère rocheux. Les données isotopiques ^{3}H/^{3}He ont permis de quantifier la magnitude de la recharge et ont été utilisées pour contraindre le modèle numérique en vérifiant les voies de migration simulées du panache tout en contraignant les propriétés hydrauliques nécessaires pour reproduire les temps de résidence et l'étendue du panache de chlorure. Les données géochimiques de certains composés organiques (carbone dissous, phosphate) et inorganiques couplées avec l'isotope ^{15}N contenu dans l'ammonium ont permis d'identifier la présence de lixiviat dans les eaux souterraines et de reconnaitre les processus de dégradation et d'immobilisation des contaminants. En retour, les données de la caractérisation hydrogéophysique ont offert le niveau de détail requis pour réévaluer le modèle conceptuel et numérique contraint par les données géochimiques. La modélisation numérique, quant à elle, apporte une meilleure compréhension quantitative du système d'écoulement et permet de vérifier la plausibilité du modèle conceptuel et des processus d'atténuation invoqués. Les données géochimiques couplées avec la modélisation numérique ont permis une meilleure compréhension de l'écoulement de l'eau souterraine et des mécanismes d'atténuation du panache de lixiviat émis par l'ancien site d'enfouissement sanitaire. En plus, de confirmer les conditions hydrogéochimiques actuelles du panache, ces outils ont permis de prédire avec confiance le devenir du panache. Finalement, la méthodologie intégrée et les outils utilisés pour caractériser l'aquifère peuvent être utilisés pour évaluer efficacement l'atténuation naturelle d'autres sites contaminés localisés dans des environnements géologiques complexes et permettre d'identifier les principaux mécanismes d'atténuation naturelle propres à chaque site.

1.3 Travaux réalisés par l'étudiante

L'étudiante a été impliquée dans le projet depuis le tout début des travaux de l'INRS-ETE, initialement en tant que stagiaire, puis en tant qu'étudiante à la maîtrise, avant un passage accéléré au doctorat. Le projet sur l'atténuation naturelle des eaux de lixiviation de l'ancien site d'enfouissement sanitaire de Saint-Lambert-de-Lauzon a débuté en mai 2006. Initialement, l'auteure a débuté en tant que stagiaire et a travaillé sur la revue de la documentation disponible sur l'ancien site d'enfouissement sanitaire de Saint-Lambert et a réalisé des travaux préliminaires permettant la vérification de l'efficacité et la capacité de la foreuse de l'INRS-ETE et du géoradar dans les conditions du site.

Suite au succès des essais de caractérisation initiaux réalisés par l'équipe, l'INRS-ETE a présenté au comité de la RIGDCC une synthèse initiale des conditions hydrogéologiques et hydrogéochimiques du site, tout en proposant des travaux de terrain supplémentaires afin de déterminer l'étendue du panache des eaux de lixiviation et de produire une étude de caractérisation détaillée de la zone d'étude. Du printemps à l'été 2007, une première phase des travaux de terrain a été réalisée. Suite à cette phase de travaux, des levés de terrain et des analyses en laboratoire supplémentaires et complémentaires ont été réalisés dans le sous-bassin versant de 12 km^2 de la région d'étude entre 2007 et 2012.

Tout au long de la réalisation des travaux de terrain et de son projet de doctorat, l'auteure a orienté plusieurs stagiaires et étudiants dans la réalisation de travaux spécifiques et de traitements de certaines des données acquises. De plus, l'étudiante est première auteure de quatre articles scientifiques qui seront soumis à des revues internationales avec comité de lecture et d'un article déjà publié. L'étudiante a aussi présenté les travaux réalisés lors de plusieurs conférences locales et internationales. Finalement, tous les travaux, les interprétations et analyses de données, la modélisation et la préparation des figures de cette thèse ont été réalisés par l'auteure, sauf spécifications différentes.

Les différentes activités auxquelles l'auteure a participé dans le cadre de la recherche doctorale sont énumérées ci-dessous :

- Numérisation et recherche de documents pertinents à l'étude.

- Numérisation et digitalisation en SIG de photos aériennes et de cartes pertinentes à l'étude.
- Familiarisation avec la zone d'étude, validation des données, et recherche d'accès pour les travaux de terrain (sources de données validées: rapports de consultants, données de puits privés obtenues du système d'information hydrogéologique (SIH), carte du Quaternaire et carte pédologique au 1/50 000, révision de cartes de la Base de données topographiques du Québec (BDTQ), révision de la localisation des rivières et du sens d'écoulement et mise à jour de l'utilisation du territoire).
- Responsable des communications avec le gestionnaire du site (Régie intermunicipale de gestion des déchets des Chutes de la Chaudière, RIGDCC) et les propriétaires de terrains dans la région d'étude.
- Mise à jour et préparation des documents d'entente de servitude avec les propriétaires pour la réalisation des travaux géophysiques, les forages, l'installation des puits d'observations et les mesures dans les puits.
- Assistance à la réalisation des levés de terrain au géoradar (avec Yves Michaud de Ressources Naturelles Canada) et des travaux avec la foreuse de l'INRS-ETE (avec Jean-Marc Ballard).
- Planification et réalisation de la phase 1 des travaux de terrain (2007), incluant la localisation des levés, la vérification des accès et les demandes de servitude (Géoradar, tomographie électrique, sondage par enfoncement, installation des puits d'observation, développement des puits d'observation).
- Présentation annuelle de l'avancement des travaux lors d'assemblées de la RIGDCC
- Rédaction de rapports techniques et supervision de campagnes de forages et de puits d'observation réalisées par des firmes d'experts.
- Réalisation des levés durant la phase 2 (recharge) et la phase 3 (géochimie) des travaux : suivi des sondes à niveaux d'eau installées dans certains des puits et suivi des niveaux d'eau, profils de température et mesures physicochimiques dans les puits d'observation, mesures de débitmètre dans les cours d'eau, échantillonnage de sol de surface, campagne d'échantillonnage géochimique et analyses en laboratoire réalisées à l'INRS-ETE.

- Interpolation d'un modèle d'élévation de terrain à l'échelle de la région d'étude et conception de nombreux fonds de carte en SIG (rivière, utilisation du territoire) pour représenter le contexte de la région d'étude à l'échelle du sous-bassin versant.
- Mise à l'échelle (de 1/50 000 à 1/5 000) de la carte du Quaternaire pour le sous-bassin versant de 12 km^2 avec la collaboration de Lise Lamarche, étudiante au doctorat en géologie du Quaternaire à l'INRS-ETE avec la collaboration de Michel Parent de Ressources Naturelles Canada (interprétation et vérification de terrain, participation à réalisation de la carte finale). Les nouvelles connaissances acquises sur la géologie du Quaternaire lors d'observation de terrain ou lors de la réalisation de forage commercial pour la RIGDCC (identification d'affleurement rocheux ou de la présence de diamicton à la surface du sol, définition de la séquence de déposition géologique par l'identification de matériaux géologiques et détermination de l'origine des matériaux s'inscrivent aussi indirectement dans les contributions de l'étudiante).
- Réalisation des coupes géologiques et hydrogéologiques.

Il est important de souligner que l'expertise de Jean-Marc Ballard, professionnel de recherche à l'INRS-ETE, lors de la réalisation des travaux avec la foreuse a été primordiale, et que l'auteure a profité des nombreux conseils de Daniel Paradis de Ressources Naturelles Canada qui a aussi fabriqué le système d'obturateurs qui a servi à l'échantillonnage géochimique. Roger Morin de l'USGS a réalisé les diagraphies et les mesures de débitmètre dans les puits. De plus, Erwan Gloaguen, professeur à l'INRS-ETE, et Christine Bélanger, étudiante à la maîtrise à l'INRS-ETE, ont réalisé les levés de résistivité électrique et leur inversion. Lise Lamarche, étudiante au doctorat à l'INRS-ETE lors de l'étude et Michel Parent de Ressources Naturelles Canada ont été impliqués pour l'interprétation de la géologie du Quaternaire. Denis Lavoie de Ressource Naturelle Canada a révisé et édité le texte et l'information sur la géologie du roc dans la région d'étude. De plus Jeanne Percival de Ressources Naturelles Canada a réalisé l'analyse semi-quantitative de la minéralogie de l'aquifère à partir des résultats des analyses par diffraction-X réalisé à l'Université Laval par Jean Frenette.

1.4 Structure de la thèse

Cette thèse utilise un format par article et elle comprend quatre parties distinctes : la 1$^{\text{ère}}$ partie décrit le contexte de l'étude; la 2$^{\text{e}}$ partie regroupe les articles scientifiques qui documentent les travaux de recherche doctoraux; la 3^{e} partie synthétise les résultats et les met en perspective par rapport au domaine de l'hydrogéologie; et, enfin, la 4^{e} partie comprend les annexes électroniques contenant les données et résultats produits dans le cadre de la recherche doctorale.

La 1ère partie comprend les chapitres 1 à 3 qui définissent les bases de la thèse et servent de mise en contexte. Le 1er chapitre sert d'introduction et défini la problématique abordée et les objectifs, de même que l'approche utilisée. Le 2^{e} chapitre est une revue de littérature couvrant les sujets scientifiques en relations avec les travaux doctoraux documentés dans la thèse. Le 3^{e} chapitre décrit la région d'étude.

La 2^{e} partie représente l'élément central de la thèse et elle comprend quatre articles scientifiques documentant les travaux réalisés. Chaque article représente une étape de la réalisation du projet de recherche définie par les trois phases des travaux de caractérisation réalisés (Figure 1.1). Chacune de ces étapes successives et complémentaires comporte des aspects originaux, autant dans les méthodes employées que dans l'apport scientifique des résultats obtenus :

- Le 1er article couvre la méthodologie générale de caractérisation hydrogéologique qui a été développée dans le cadre de ce projet pour définir l'hétérogénéité de l'aquifère, ainsi que l'approche utilisée pour faire l'intégration des données et produire un modèle hydrogéologique conceptuel initial. Cette approche de caractérisation de l'hétérogénéité pour un site réel se démarque par l'échelle de quelques km^2 qu'elle vise, alors que les approches hydrauliques ou géophysiques utilisées pour caractériser l'hétérogénéité vise généralement des zones de petite dimension (< 50x50 m). La combinaison et la séquence des méthodes utilisées est aussi originale, notamment l'usage de puits crépinés sur toute l'épaisseur de l'aquifère installés par enfoncement (*direct push*);
- Le 2^{e} article couvre les travaux reliés à l'usage d'indicateurs superficiels des processus de recharge de la nappe et des interactions entre l'aquifère et les eaux de surface, qui ont des incidences sur l'écoulement, la migration du lixiviat et son incidence sur la qualité de l'eau de surface. Ces travaux se démarquent aussi par l'échelle d'un sous-bassin pour l'estimation de la recharge avec une combinaison de méthodes originales. L'évaluation de la recharge se fait généralement à l'échelle plus régionale. Les indicateurs utilisés sont originaux et permettent de montrer la complexité des facteurs contrôlant la recharge et

l'influence des conditions superficielles sur la géochimie de l'eau souterraine ainsi que les échanges entre l'aquifère et l'eau de surface.

- Le 3^{e} article couvre la caractérisation des conditions hétérogènes rencontrées dans la région d'étude aux niveaux de la géochimie des sédiments composants l'aquifère ainsi que de l'eau souterraine, ce qui affecte l'évolution géochimique de l'eau souterraine et du lixiviat. Ces travaux se démarquent d'abord par les échelles variées qui ont été considérées, allant de l'échelle du grand bassin versant de la Chaudière, à celle du sous-bassin de la région d'étude à l'échelle ponctuelle des échantillons de sol. Les travaux sont aussi originaux parce qu'ils établissent, grâce à des indicateurs et à une analyse multi-variables, le lien entre les propriétés des sols ainsi que les conditions à la surface avec leurs effets sur la géochimie de l'eau souterraine et ses incidences sur les processus d'atténuation naturelle du lixiviat. Ces travaux illustrent l'importance de l'hétérogénéité géochimique sur les conditions géochimiques et sur les processus d'atténuation.
- Le 4^{e} article utilise la modélisation numérique et les évidences géochimiques pour identifier les processus d'atténuation naturelle et prédire le devenir du panache de lixiviat émis par le site d'enfouissement. Ces travaux se démarquent par l'intégration d'un vaste ensemble de données géochimiques et l'utilisation de diverses méthodes d'interprétation (géochimiques et modélisation) pour faire l'évaluation qualitative et quantitative des processus d'atténuation naturelle, tout en considérant le contexte particulier du système aquifère. Ces travaux ont aussi utilisé une variété originale d'approches pour la vérification du modèle numérique d'écoulement et de transport de masse, réduisant ainsi l'incertitude dans les résultats de la modélisation.

La 3^{e} partie de la thèse comprend un chapitre faisant la synthèse des travaux et donnant une perspective sur les contributions scientifiques et les questions à aborder par des travaux futurs. Les contributions scientifiques de l'étudiante y sont décrites en détail et cette section se termine avec les conclusions. Enfin, la 4^{e} et dernière partie de la thèse contient les annexes électroniques qui servent à documenter en détail les méthodes utilisées ainsi qu'à assurer la pérennité des données produites.

Chapitre 2

REVUE DE LITTÉRATURE

2.1 Problématiques reliées aux sites d'enfouissement sanitaire

La gestion des matières solides résiduelles est une problématique majeure à échelles locale, régionale et internationale, car le stockage des déchets peut induire la pollution de l'air, du sol, de l'eau souterraine et de l'eau de surface (INTOSAI 2004). Bien que l'incinération soit évaluée comme la meilleure technologie actuelle pour la gestion des matières solides résiduelles, l'enfouissement demeure l'option de disposition des déchets la plus utilisée dans la majorité des pays industrialisés (Ehrlich 2011; Dijkgraaf et Volebergh 2004). De plus, les problèmes de gestion des déchets sont amplifiés par le taux de croissance de la population et les habitudes de consommation (INTOSAI 2004).

Au Québec, des statistiques démontrent que le volume de déchets produits a augmenté de 47% en 10 ans, même si les Québécois recyclent davantage qu'ils ne jettent (Lacourse 2012). De nos jours, les lieux d'enfouissement technique (LET) sont munis de membranes imperméables ainsi que d'un système de captage des eaux de lixiviation conforme aux normes actuelles de gestion (MDDEP 2012a; Butt et al. 2008); ce qui réduit les impacts environnementaux sur les milieux récepteurs. Cependant, avant les années 80, il y avait très peu de considérations environnementales dans la conception et l'opération d'un site d'enfouissement sanitaire. À cette époque, on optait souvent pour les anciennes zones d'exploitation de granulats (sablière/gravière) comme lieu d'enfouissement sanitaire (LES) et aucune barrière ou membrane imperméable n'était installée sous ou au-dessus des déchets pour intercepter les lixiviat (Roy 1994). Dans de telles conditions, l'eau des précipitations peut s'infiltrer librement à travers les matières solides résiduelles en décomposition et se charger de contaminants. Ces eaux de lixiviation s'infiltrent ensuite dans le sol, atteignent la nappe phréatique et forment un panache qui migre grâce à l'écoulement de l'eau souterraine (Fetter 2001). Les eaux de lixiviation provenant d'un LES peuvent contenir une grande quantité de contaminants organiques et inorganiques, et leur composition dépend du type de déchets, de la localisation géographique ainsi que de la gestion du site au cours de l'exploitation (Roy 1994). Souvent, on ignore la provenance ou la nature des déchets enfouis dans les anciens LES. Ces sites peuvent potentiellement contenir divers contaminants provenant de sources municipale, agricole, et industrielle, de même que des contaminants radioactifs ou autres composés toxiques et dangereux (Bjerg et al. 2003; Christensen et al. 1994).

Les anciens sites d'enfouissement sanitaire ne sont plus en exploitation, car ils ne respectent pas les normes environnementales actuelles. Suite à leur fermeture, certains anciens LES ont été recouverts d'une membrane imperméable, empêchant ainsi l'eau de pluie de s'infiltrer dans les déchets. Bien que la production des eaux de lixiviation soit réduite considérablement lorsque le site est recouvert d'une membrane imperméable, les déchets continuent de se décomposer et constituent une source potentielle de pollution pour des décennies, voire des siècles (Bjerg et al. 2003). De plus, dans le cas d'un ancien site recouvert d'une membrane, l'intégrité de cette dernière doit être considérée, car au fil des années, si la membrane est perforée, la quantité de lixiviat émise dans les eaux souterraines pourrait augmenter (Christensen et al. 2001).

Au Québec, le règlement sur l'enfouissement et l'incinération de matières résiduelles (REIMR) assure la gestion environnementale des lieux d'enfouissement technique (MDDEP 2012a). Par contre, pour les LES, l'article 160 précise notamment que les lieux d'élimination qui ont été fermés définitivement avant l'entrée en vigueur du REIMR demeurent régis par les dispositions du règlement sur les déchets solides (RDS) et par les exigences spécifiées dans leur certificat d'autorisation ou de conformité. Cependant, dans le RDS, il n'existe aucun critère pour les concentrations de contaminants retrouvés dans les eaux souterraines ou pour celle de contaminants persistants qui pourraient faire résurgence dans les eaux de surface, tels que l'ammonium (MDDEP 2012b). Présentement, aucun suivi environnemental n'est exigé si l'ancien LES respectait, lors de sa fermeture, l'article 29 du RDS sur les conditions hydrogéologiques et l'article 30 pour les eaux de lixiviation qui font résurgence dans les eaux de surface. Dans certains cas spécifiques, un suivi des eaux souterraines et des eaux de surface pouvait être exigé par le MDDEP. Ce programme de suivi pouvait inclure l'échantillonnage de plusieurs piézomètres au cours de l'année et l'analyse de plusieurs paramètres, dont les composés phénoliques totaux et individuels. Ces paramètres peuvent être utilisés comme indicateur de la qualité de l'eau souterraine et, advenant une augmentation significative de l'un des paramètres, justifiant un échantillonnage complémentaire. Ceci dit, la plupart des LES fermés avant l'entrée en vigueur du REIMR ont reçu des certificats de conformité, impliquant qu'aucun suivi environnemental n'est exigé pour la grande majorité des sites. Pour cette raison, un grand nombre d'anciens sites d'enfouissement, au Québec mais aussi à travers le monde, représentent un risque sérieux de contamination pour les eaux souterraines et les eaux de surface (Cozzarelli et al. 2011; Butt et al. 2008; EA 2003; Kjeldsen et al. 2002; EC 2000, 1999).

Selon l'agence de protection environnementale des États-Unis (USEPA), aux États-Unis il y aurait plus de 10 000 anciens sites d'enfouissement sanitaire maintenant abandonnés et plus de 3000 lieux d'enfouissement technique présentement actifs sur le territoire des États-Unis. Les données disponibles sur les sites actifs munis de membranes imperméables et où il existe un suivi environnemental montrent que 82% de ces sites ont des fuites de lixiviat connues (Zero Waste America 2010). Selon Lee et Jones (1991), il est plus difficile de détecter des fuites de lixiviat dans l'eau souterraine dans le cas des LET que dans le cas des LES. En effet, puisque les LET possèdent des membranes imperméables, les panaches de lixiviat sont plus étroits comparativement à ceux issus des anciens LES qui émettent des panaches plus étendus.

2.2 Géochimie des sites d'enfouissement

2.2.1 Dégradation des déchets et production du lixiviat

Suite à l'enfouissement, les matières solides résiduelles subissent une série complexe de réactions chimiques, physiques et biologiques qui participent simultanément à la décomposition des déchets (Christensen et al. 1994). Les sites d'enfouissement connaissent quatre étapes de décomposition: 1) une phase aérobie initiale; 2) une phase anaérobie acide; 3) une phase méthanogène initiale; et 4) une phase méthanogène stable. Le Tableau 2.1 fait le sommaire des phases de dégradation des déchets définies sur la base d'essais en laboratoire (Kjeldsen et al. 2002; Andreottola et Cannas 1992).

La première phase de décomposition des substances organiques est d'une durée limitée, car la demande en oxygène sera élevée tandis que la quantité disponible à l'intérieur de la pile de déchets sera faible. Conséquemment, l'oxygène sera rapidement consommé et cette phase de décomposition ne sera possible que dans la partie supérieure de la pile de déchets, où l'oxygène est soit piégé entre les matières solides résiduelles fraîchement déposées ou transporté par de la pluie. Au cours de cette phase, les protéines se décomposent en acides aminés et la cellulose se transforme en glucose. Lors de ces transformations, la température augmente et certains

Tableau 2.1 : Phases de dégradation des déchets

(Kjeldsen et al. 2002; Andreottola et Cannas 1992)

Phase 1 **Aérobie initiale**	**Phase 2** **Anaérobie acide**	**Phase 3** **Méthanogène initiale**	**Phase 4** **Méthanogène stable**
• Présence d'oxygène, qui est toutefois rapidement consommé • **pH acide** • Dégradation des protéines en acides aminés • Transformation de la cellulose (majeure partie de la fraction organique des déchets) en glucose qui à son tour est converti en CO_2, eau, nitrate et sulfate • Augmentation de la température • Phase à durée limitée (seulement quelques jours) étant donné le manque d'oxygène suite au recouvrement des déchets • **Aucune production substantielle de lixiviat** • Production d'humidité	• Conditions anaérobies et réactions de fermentation • Bactéries biodégradant 45-60% des déchets • pH neutre à acide • Présence de bactéries anaérobies seulement • Réduction du potentiel d'oxydoréduction pour favoriser la production de bactéries méthanogènes (ORP < 300 mV) **Étape 1 :** • pH neutre • Biodégradation de la cellulose et de l'hémicellulose par les bactéries • Production de CO_2 et de CH_4 **Étape 2 :** • pH acide 5-6 • Dominance des bactéries hydrolytiques, fermentatives et acétogènes • Accumulation d'acide carboxylique • Mise en solution de plusieurs ions inorganiques (Cl^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+) • Fortes concentrations de COD et BOD • **Production d'ammoniac (500-1000 mg/L) dû à l'hydrolyse et à la fermentation des protéines** • **Production de lixiviat** • **Mise en solution de sels solubles**	• **Quantités mesurables de méthane produites** • **pH neutre (6-8)** • **Augmentation de l'alcalinité** et du pH et réduction des sulfates et précipitation subséquente en sulfure à cause de l'activité des bactéries • Conversion de l'acide accumulé durant la phase acide en CH_4 et en CO_2 par les bactéries méthanogènes toxiques • Les concentrations de méthane dans le gaz s'accroissent • **Décroissance de la solubilité du calcium, du fer et du manganèse** • **Faible concentration de COD et de BOD** • **Production et émission d'ammoniac se poursuivant de façon constante (cette phase dure plusieurs années, tant que la dégradation de la portion organique des déchets est active)** • **Production et émission de lixiviat**	• **La production de méthane atteint un maximum** et décroît selon la disponibilité des acides carboxyliques de la portion organique (cellulose et hémicellulose) • pH alcalin • Peu de données sur cette phase, car peu de sites ont été étudiés pendant plus de 30 ans • **Augmentation continue du pH** • Faible diminution de la production et de l'émission d'ammoniac (qui se poursuit de façon constante) • **Émission du lixiviat (peut se poursuivre pendant des décennies)**

composés sont produits, tels que l'eau, le dioxyde de carbone (CO_2), les sulfates (SO_4) et les nitrates (NO_3).

La deuxième phase est anaérobie et comprend deux étapes distinctes où des bactéries différentes sont impliquées. Il y a d'abord dégradation de la cellulose et de l'hémicellulose, représentant la portion organique qui compose généralement de 45 à 60% de la masse des déchets. Il y aura aussi formation et accumulation d'acide carboxylique qui aura comme impact une diminution de pH (5-6) et qui engendrera à son tour la mise en solution de plusieurs composés inorganiques. Suite à cette deuxième phase, des eaux de lixiviation seront produites.

La troisième phase est caractérisée par l'augmentation du pH et par la production considérable du méthane. Ce n'est toutefois qu'au cours de la quatrième et dernière phase que la production du méthane atteindra son maximum et que le pH augmentera davantage, réduisant ainsi la mise en solution des sels. Il y a toutefois peu d'information disponible sur cette dernière étape de stabilisation des déchets, car aucun de site n'a été investigué sur une période de plus de 30 années. L'investigation d'un ancien site d'enfouissement, 15 ans après sa fermeture, démontre que la décomposition biologique se poursuit et que la stabilisation est dépendante de plusieurs conditions, dont le type et la profondeur des déchets, l'effort de compactage, la densité initiale, la quantité de sols de recouvrement utilisée, la quantité de lixiviat, la température, le taux d'humidité et la production de gaz (Al-Yaqout et Hamoda 2007). Par contre, il est reconnu que, tout au long du processus de dégradation des déchets, des acides aminés seront produits, transformés et émis par les eaux de lixiviation sous la forme d'ammoniac et d'ammonium (Cozzarelli et al. 2011; Kjeldsen et al. 2002).

2.2.2 Composition du lixiviat et toxicité

De façon générale, la composition des eaux de lixiviation sera dépendante de la phase de décomposition des déchets (Christensen et al. 1994). Après plusieurs années d'enfouissement, il est possible que différentes sections du site soient dans une phase de décomposition différente. Des variations spatiales importantes dans les concentrations du lixiviat ont ainsi été observées à l'ancien site d'enfouissement sanitaire de Grindsted au Danemark, qui couvre une superficie de 10 hectares. Des concentrations très faibles de tous les paramètres géochimiques ont été observées sur 60-70% du site. Par contre, à un endroit occupant seulement 10% du site, des concentrations de contaminants de 20-1000 fois plus élevées ont été mesurées (Christensen et al. 2001). L'information sur la variabilité spatiale du lixiviat est donc importante, autant pour localiser les sources primaires de lixiviat que pour établir l'importance de la contamination causée par un ancien site d'enfouissement sanitaire.

La variabilité des directions d'écoulement liée à la présence d'un site d'enfouissement est également un aspect important à considérer. En fait, des hauts piézométriques causés par la topographie de la pile de déchet et par la rétention d'eau due à une porosité variable dans les déchets peuvent engendrer des changements locaux de gradients hydrauliques et de directions de l'écoulement pour la nappe libre. De plus, ces gradients peuvent aussi changer en fonction des variations saisonnières dans les patrons d'écoulement (Christensen et al. 2001; Breukelen van 2003). Finalement, la densité et la viscosité des eaux de lixiviation (qui sont plus chargées en solides totaux dissous que l'eau souterraine naturelle), affecteront, respectivement, la position verticale du panache et les vitesses d'écoulement (Christensen et al. 2001).

Une grande quantité de contaminants organiques et inorganiques se retrouvent ainsi dans les eaux de lixiviation au cours de la décomposition des déchets. Les eaux de lixiviation sont définies comme étant une solution à base d'eau contenant quatre groupes principaux de contaminants: 1) matière organique dissoute; 2) macrocomposés inorganiques; 3) métaux lourds et; 4) composés organiques xénobiotiques (Christensen et al. 1994). Le Tableau 2.2, tiré de Christensen et al. (2001), résume les principaux composés et les concentrations typiques retrouvées dans les eaux de lixiviation pour les paramètres physicochimiques et les composés inorganiques. Le bore, le sulfure, le baryum, et le lithium sont des éléments qui peuvent se retrouver dans les eaux de lixiviation. La plupart du temps, ces composés ne sont pas analysés et, lorsqu'ils le sont, ils se retrouvent en faibles concentrations (Bjerg et al. 2003).

La composition chimique des eaux de lixiviation dépend de plusieurs facteurs incluant la masse totale et le type de déchets, la localisation géographique du site ainsi que la conception, la gestion et l'âge du site. Le degré de stabilisation des déchets et la quantité d'eau qui s'infiltre dans les déchets auront également une influence significative sur les eaux de lixiviation émises par le site (Andreottota et Cannas 1992). Pour les sites où une large quantité de produits chimiques a été déversée, il est aussi possible qu'une phase organique distincte puisse être présente (Christensen et al. 2001).

Tableau 2.2 : Composition typique d’eau de lixiviation

(compilation de Christensen et al. 2001)

Paramètres et composés inorganiques	**Plage (mg/l)**	**Composés organiques**	**Plage (µg/l)**
pH	4.5-9	*Hydrocarbure aromatique*	1-12 300
Conductivité spécifique ($\mu S\ cm^{-1}$)	2500-35 000	Benzène	1-1630
Solides totaux	2000-60 000	Toluène	4-3500
		Xylène	1-1280
Matière organique		Éthylbenzène	4-250
Carbone organique total	30-29 000	Naphthalène	0.1-260
Demande biologique en oxygène (DBO5)	20-57 000		
Demande chimique en oxygène (DCO)	140-152 000	*Hydrocarbure halogéné*	
DBO5/COD (ratio)	0.02-0.80	Chlorobenzène	0.1-110
Azote organique	14-2500	1,2-Dichlorobenzène	0.1-32
		1,4-Dichlorobenzène	0.1-16
Éléments inorganiques		1,1,1-Trichloroéthane	0.1-3810
Phosphore total	0.1-23	Trichloroéthylène	0.7-750
Chlorure	150-4500	Tétrachloroéthylène	0.1-250
Sulfate	8-7750	Chlorure de méthylène	1.0-64
Hydrogène carbonate	610-7320	Chloroforme	1.0-70
Sodium	70-7700		
Potassium	50-3700	*Phénols*	
Ammonium – N	50-2200	Phénol	1-1200
Calcium	10-7200	Crésol	1-2100
Magnésium	30-15 000		
Fer	3-5500	*Pesticides*	
Manganèse	0.03-1400	Mecoprop	2.0-90
Silicium	4-70		
		Divers	
Métaux lourds		Acétone	6-4400
Arsenic	0.01-1	Diethylphalate	10-660
Cadmium	0.0001-0.4	Di-n-buthylphthalate	5.0-15
Chrome	0.02-1.5	Tetrahydrofurane	9-430
Cobalt	0.005-1.5	Tri-n-buthylphosphate	1.2-360
Cuivre	0.005-10	Camphre	1
Plomb	0.001-5		
Mercure	0.0-0.16		
Nickel	0.15-13		
Zinc	0.03-1000		

Un grand nombre de composés dangereux et nocifs pour les humains et pour l’environnement peuvent être retrouvés dans les sites d’enfouissement sanitaire. Une étude effectuée en Suède a porté sur la caractérisation de 400 composés organiques et inorganiques dans 12 sites d’enfouissement (Öman et Junestedt 2008). Les auteurs notent que les composés organométalliques devraient être davantage caractérisés dans les études sur les sites d’enfouissement et que le chlore et l’ammoniac sont souvent retrouvés en concentration toxique pour la vie aquatique. Parmi toutes les analyses réalisées, 55 composés ont été documentés pour la première fois et certains d’entre eux présentaient des concentrations significatives. De plus, plusieurs composés ont seulement été retrouvés dans les sédiments, démontrant l’existence de

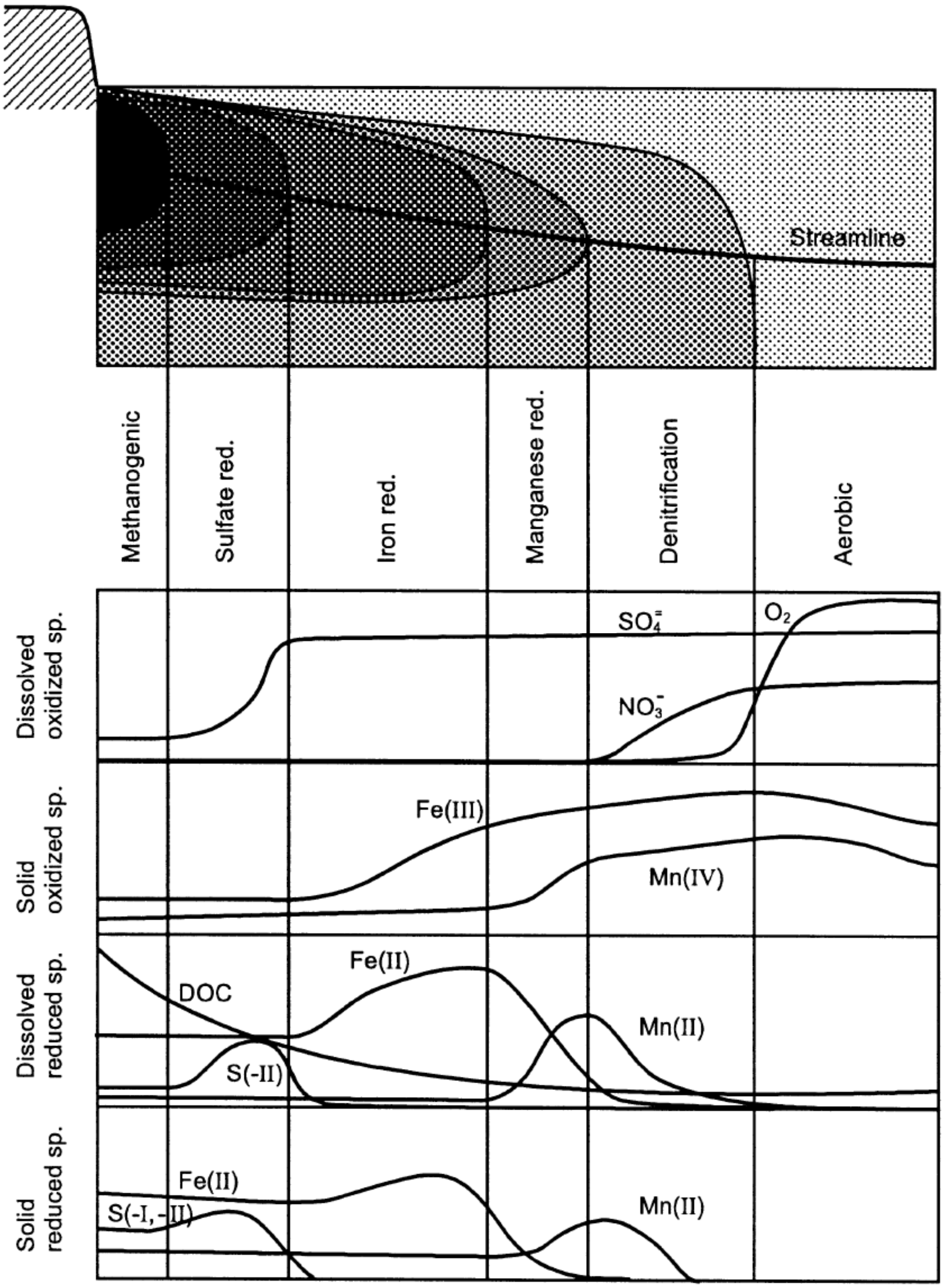


Figure 2.1 : Schéma des zones d’oxydoréduction.

Les zones d’oxydoréduction qui vont se former dans un aquifère originellement oxydant lorsqu’un panache de lixiviat va migrer dans ce dernier (tiré de Christensen et al. 2001) processus d’immobilisation de plusieurs contaminants dans les sols. Ils concluent que la présence de plusieurs des composés investigués dans leur étude a un impact significatif sur l’évaluation du risque et le développement des méthodes de traitements des eaux de lixiviation dans les sites d’enfouissement. Ces composés ont le potentiel de migrer avec les eaux souterraines ou d’être remobilisés par les sols dans le cas des LES. Dans le cas des LET, les eaux de lixiviation captées et traitées risquent de contenir plusieurs de ces composés nocifs et peuvent potentiellement être rejetées dans l’environnement s’ils ne font pas l’objet d’un suivi analytique. Une liste plus exhaustive de composés dangereux et nocifs devrait donc être prise en considération lors de l’évaluation des paramètres à traiter ou à analyser pour la gestion du risque environnemental. Conséquemment, une bonne compréhension de la composition des eaux de lixiviation est critique pour prédire les effets à long terme et l’impact des anciens LES (van Breukelen 2003; Kjeldsen et al. 2002; Christensen et al. 2001).

Une étude écotoxicologique conduite en Colombie pour évaluer la toxicité des lixiviats sur la base de divers paramètres démontre que la toxicité du lixiviat au site étudié est accrue par la présence de carbone organique dissous (COD) et de cadmium (Olivero-Verbel et al. 2008). Selon cette étude, la toxicité dépendrait davantage de la formation de complexes organiques que de la concentration d’ions ou d’éléments libres qui seraient émis par le site. Les eaux de lixiviation peuvent également contenir une variété de bactéries. L’identification de famille de bactéries peut être utile pour détecter la présence de lixiviat dans les eaux souterraines ou pour identifier les voies de migration et la dégradation du lixiviat (Mouser et al. 2010; van Breukelen 2003).

2.2.3 Évolution géochimique et atténuation naturelle du lixiviat

Les sites d’enfouissement sanitaire sont des milieux très réducteurs alors que les aquifères les entourant sont généralement des milieux relativement oxydants, surtout s’ils sont en condition libre. L’entrée de lixiviat très réducteur dans un aquifère oxydant amène la création de zones ou d’environnements d’oxydoréduction très systématiques (Figure 2.1). Ainsi, autour d'un LES, il se formera une série de zones d’environnements réducteurs distincts à partir de la source de contamination jusqu’à l’extrémité du panache lors de la migration et de la dégradation des contaminants (Figure 2.2). Les réactions impliquées sont les suivantes : biodégradation de la matière organique, processus abiotiques d’oxydoréduction, réactions de précipitation et de dissolution de minéraux, formation de complexes, échange ionique et adsorption (Figure 2.3).

Dans les sites d’enfouissement, la migration du panache de lixiviat dans l’aquifère provoque l’atténuation des contaminants par des processus tels que la dégradation, la sorption, la volatilisation, la précipitation et l’échange ionique. Certaines de ces réactions sont décrites à la Figure 2.3. Tous les composés du lixiviat qui entrent dans l’aquifère sont sujets à la dilution lorsque les eaux de lixiviation se mélangent à l’eau souterraine. Pour les composés non réactifs, comme les chlorures, la dilution sera le seul mécanisme d’atténuation.

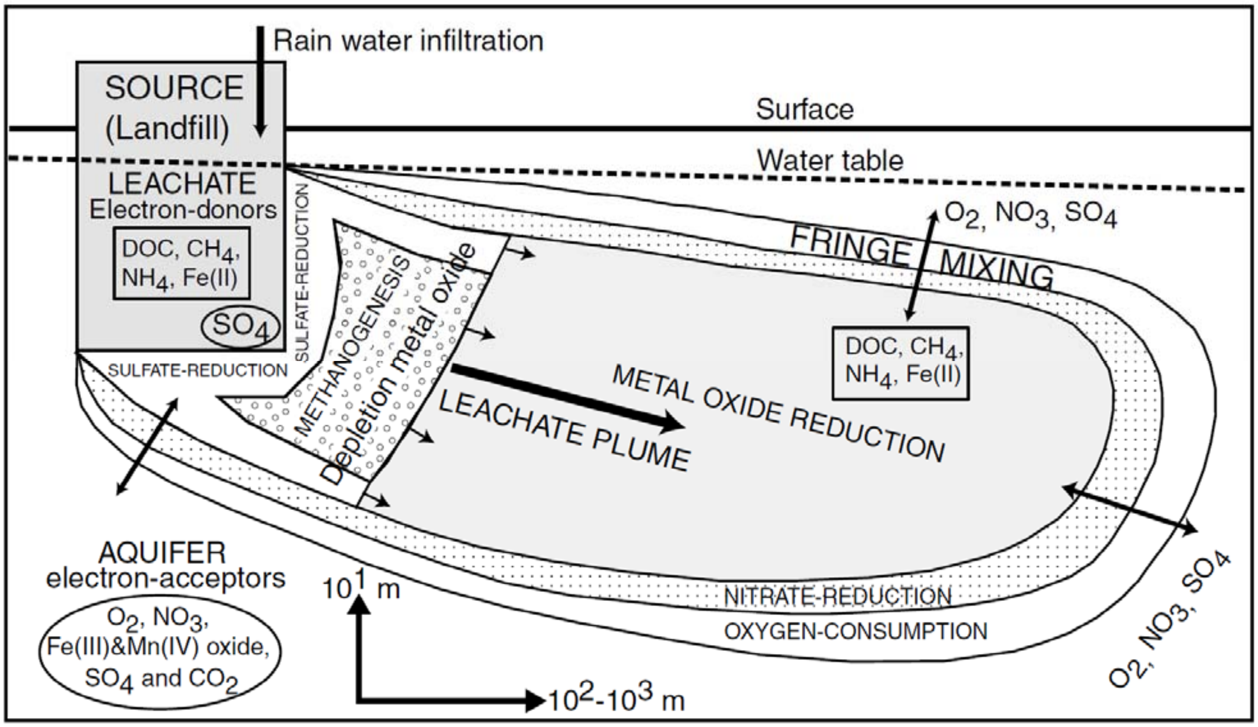


Figure 2.2 : Distribution spatiale des processus de d’oxydoréduction.
Les processus d’oxydoréduction sont couplés avec la dégradation du carbone organique dans un site d’enfouissement typique (tiré de van Breukelen 2003)

Les environnements d’oxydoréduction ont une grande influence sur la composition biochimique, organique et inorganique de l’aquifère et en même temps servent d’assise pour la compréhension des processus d’atténuation dans le panache. Les zones d’oxydoréduction influencent la composition biochimique de l’aquifère et, par la même occasion, créent un environnement géochimique particulier qui permet d’étudier et de comprendre les processus d’atténuation dans le panache (Bjerg et al. 2011; Cozzareli et al. 2011; van Breukelen 2003; Christensen et al.

2001). Dans un aquifère subissant l'apport constant de lixiviat, une zone méthanogène prévaut tout près du site (Figure 2.2). À partir de cette zone, des zones réductrices vont se former jusqu'à

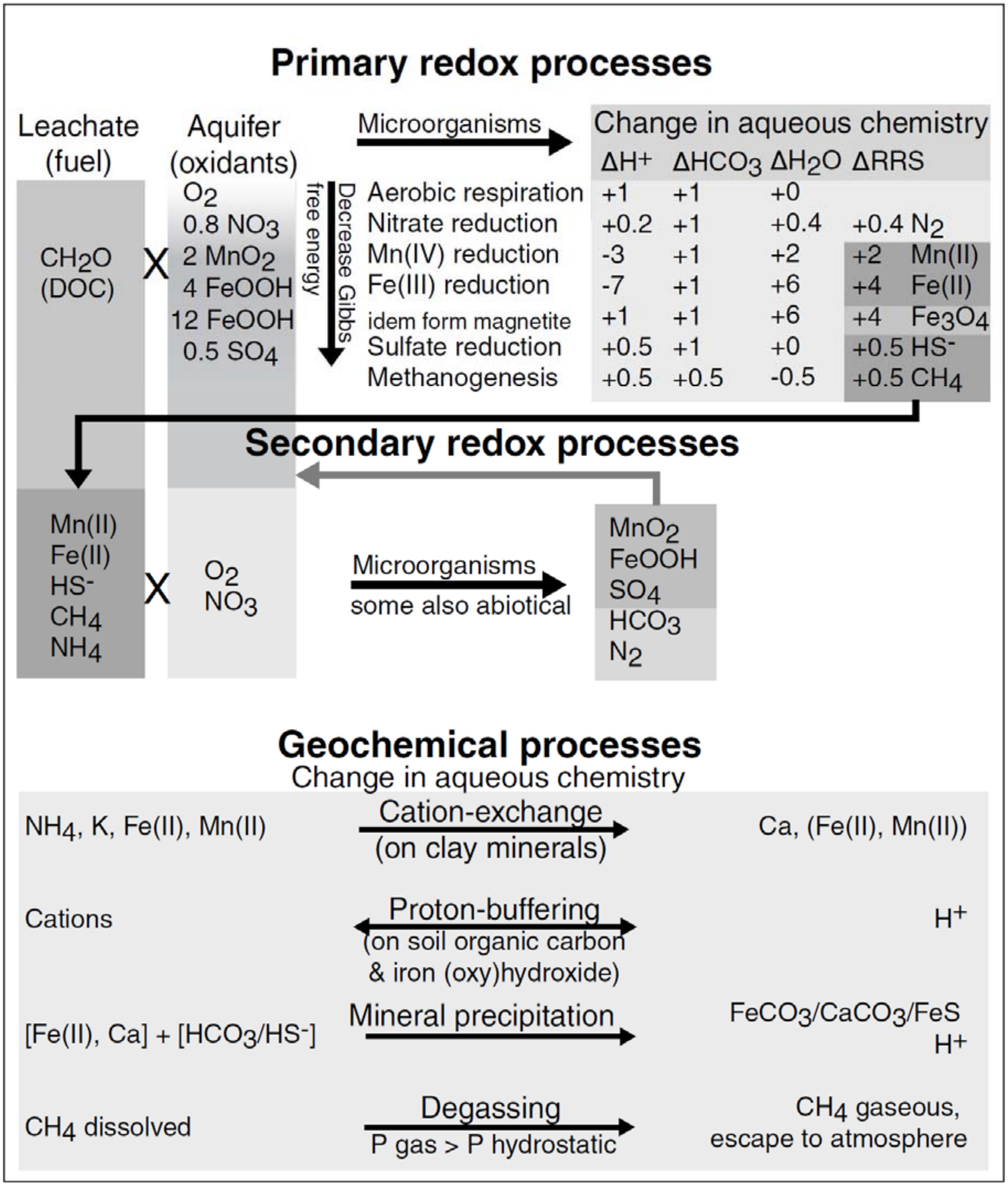


Figure 2.3 : Réactions biogéochimiques liées aux panaches de lixiviat.
Les réactions biogéochimiques prennent place dans les panaches de lixiviat qui migrent dans les eaux souterraines (tiré de van Breukelen 2003)

ce que des conditions aérobies s'imposent à la limite du panache (si l'aquifère est oxydant et possède à cet endroit une concentration significative d'oxygène). Les contaminants qui quittent le site vont migrer à travers cette série de zones réductrices et éventuellement rejoindre un environnement plus oxydant. Le long d'une ligne d'écoulement d'eau souterraine, le contenu d'espèces réductrices dans l'eau décroît. La composition des minéraux en phase solide change aussi avec la distance parcourue depuis le site.

Les zones d'oxydoréduction vont être influencées par plusieurs facteurs, comme les fluctuations de la nappe, l'infiltration, le changement de direction de l'écoulement, les variations naturelles dans les sédiments, etc. De plus, les zones d'oxydoréduction peuvent se chevaucher permettant ainsi à des réactions simultanées de se produire à l'intérieur d'une même zone, ce qui rend la compréhension des processus encore plus complexes. Dans un aquifère possédant un taux plus élevé de matière organique, certaines zones peuvent donc être méprises pour des zones d'oxydoréduction engendrées par les eaux de lixiviation. En effet, les hétérogénéités géologiques et géochimiques peuvent aussi causer des activités d'oxydoréduction (Anderson et McCray 2011; Allen-King et al. 2010). La disponibilité naturelle dans l'aquifère de certaines espèces sous forme solide, tels le fer et le manganèse, pour la réduction, est un facteur important à considérer, car elles pourraient constituer des accepteurs d'électrons dominants dans la plupart des panaches de lixiviat en agissant comme agents tampons (Christensen et al. 2001; Medina Lopez 2001). De plus, les zones d'oxydoréduction affectées par l'effet des eaux de lixiviation démontrent un nombre de bactéries relativement élevé en comparaison aux eaux naturelles et chacune des zones d'oxydoréduction est associée à une variété de bactéries spécifiques à chacune des zones.

Belevi et Baccini (1992) ont estimé par des tests de lixiviat provenant de déchets municipaux que les eaux de lixiviation peuvent contenir certains composés en grande concentration pendant des centaines d'années, en particulier le carbone organique et l'azote qui vont être significatifs en terme de quantité. Les cations sont énormément influencés par l'échange ionique avec les sédiments, même lorsque les sédiments sont grossiers (ex.: sable). Les réactions d'échanges et de dilution vont ainsi créer un patron de migration très complexe pour les cations dans le panache. Tous les composés contenus dans le lixiviat vont être atténués différemment et divers facteurs vont influencer la dégradation de chaque type de composé. Le Tableau 2.3, adapté de Christensen et al. (2001), résume les principales réactions d'atténuation associées à chaque type de composés. Le type de déchets aura des conséquences sur les bactéries disponibles et sur les conditions physicochimiques (pH, oxydoréduction) au cours de la dégradation. A leur tour, les conditions physicochimiques auront un impact sur le type de réactions chimiques, telles que la précipitation, la dissolution, l'oxydoréduction et l'adsorption.

Tableau 2.3 : Sommaire des réactions d'atténuation

(Christensen et al. 2001)

Composés chimiques	Caractéristiques	Mécanismes d'atténuation et réactions principales
Matière organique dissoute	Difficile de différencier les composés organiques présents dans la matière organique dissoute. Il est donc difficile d'être spécifique à propos de l'atténuation de ces contaminants. La portion de carbone organique dissous de la phase acide se dégrade facilement (acides gras volatils). Par contre, la fraction de matière organique dissoute présente dans la phase méthanogène du lixiviat est récalcitrante et ne se dégrade pas facilement (composés réfractaires, acide humique et fulvique).	Dilution, sorption, dégradation
Composés organiques xénobiotiques	La plupart de ces composés semble se dégrader dans le panache. Tous les composés de ce groupe se dégradent de manières différentes et des études spécifiques à la dégradation de certains composés sont disponibles.	Dilution, sorption, dégradation
Éléments inorganiques présents en grande quantité; i.e. plusieurs mg/l	La signification de ces processus dépend de la concentration des espèces et du pH. Les anions sont importants à cause de leur capacité à former des complexes et à participer aux réactions de précipitation et de dissolution. Par contre, la formation de complexes peut augmenter la mobilité des cations et des métaux lourds. L'atténuation des cations se fait principalement par échange ionique.	Dilution, oxydoréduction, échange ionique, précipitation
NH_4^+	Participe à des réactions d'oxydoréduction complexes, les mécanismes d'atténuation de ce composé ne sont pas bien compris. La concentration de ce composé ne décroît pas beaucoup dans le temps et il demeure un polluant à long terme.	Échange ionique (sorption), oxydation anaérobie
Fe, Mn, S	Impliqués activement dans les réactions d'oxydoréduction. Fe et Mn sont associés aux sédiments sous forme d'oxyde et d'hydroxyde (Fe(III) et Mn(IV)). Fe réduit sous la forme Fe(II) représente un agent tampon dominant dans les zones d'oxydoréduction.	Oxydoréduction, dilution, dissolution abiotique des minéraux du sédiment, échange ionique, précipitation, complexation avec du C organique
Métaux lourds	Ne représentent pas un problème au niveau de la contamination de l'eau souterraine dans les sites d'enfouissement car les métaux lourds participent activement à des réactions de sorption et de précipitation et parce qu'ils n'existent pas en grande quantité dans les sites. Par contre, la présence de ces composés dans les lixiviats complique la compréhension de l'atténuation car les divers métaux vont réagir différemment et vont se redistribuer dans le panache selon les conditions de pH (peuvent augmenter la solubilité ou la mobilité des métaux).	Sorption, précipitation, dilution

Dans les eaux de surface, on peut observer une diminution de la concentration en oxygène, des changements au niveau de la flore et de la faune des cours d'eau et l'apparition d'une toxicité par la présence d'ammonium produit de la dégradation des protéines (Kjeldsen et al. 2002). La pollution de l'eau de surface causée par les eaux de lixiviation a été observée, mais peu de cas réels ont été étudiés et décrits en détail dans la littérature (Milosevic et al. 2012). Les effets potentiels prédominants du lixiviat rejeté dans les eaux de surface seraient la réduction d'oxygène dans une partie du cours d'eau, la toxicité induite par l'ammonium et des changements au niveau de la flore et de la faune (Haile et Abiye 2012; Milosevic et al. 2012; Kjeldsen et al. 2002).

Tableau 2.4 : Principales études sur l'atténuation naturelle des panaches de lixiviat

Études sur l'atténuation naturelle des contaminants de panaches de lixiviat: mécanismes, taux de dégradation et principaux traceurs.		
Titre	**Auteurs**	**Date**
Identification of discharge zones and quantification of contaminant mass discharges into a local stream from a landfill in a heterogeneous geological setting	Milosevic et al.	2012
Natural attenuation processes in landfill leachate plumes at three Danish sites	Bjerg et al.	2011
Biogeochemical evolution of a landfill leachate plume, Norman, Oklahoma	Cozzarelli et al.	2011
Biochemistry at a wetland sediment-alluvial aquifer interface in a landfill leachate plume	Lorah et al.	2009
The groundwater geochemistry of waste disposal facilities	Bjerg et al.	2003
Biogeochemistry and isotope geochemistry of a landfill leachate plume	van Breukelen et al.	2003
Natural attenuation of landfill leachate: a combined biogeochemical process analysis and microbial ecology approach	van Breukelen	2003
Present and long-term composition of MSW landfill leachate: a review	Kjeldsen et al.	2002
Biogeochemistry of landfill leachate plumes	Christensen et al.	2001
Attenuation of landfill leachate pollutants in aquifers	Christensen et al.	1994
Landfilling of waste: leachate	Christensen et al.	1992

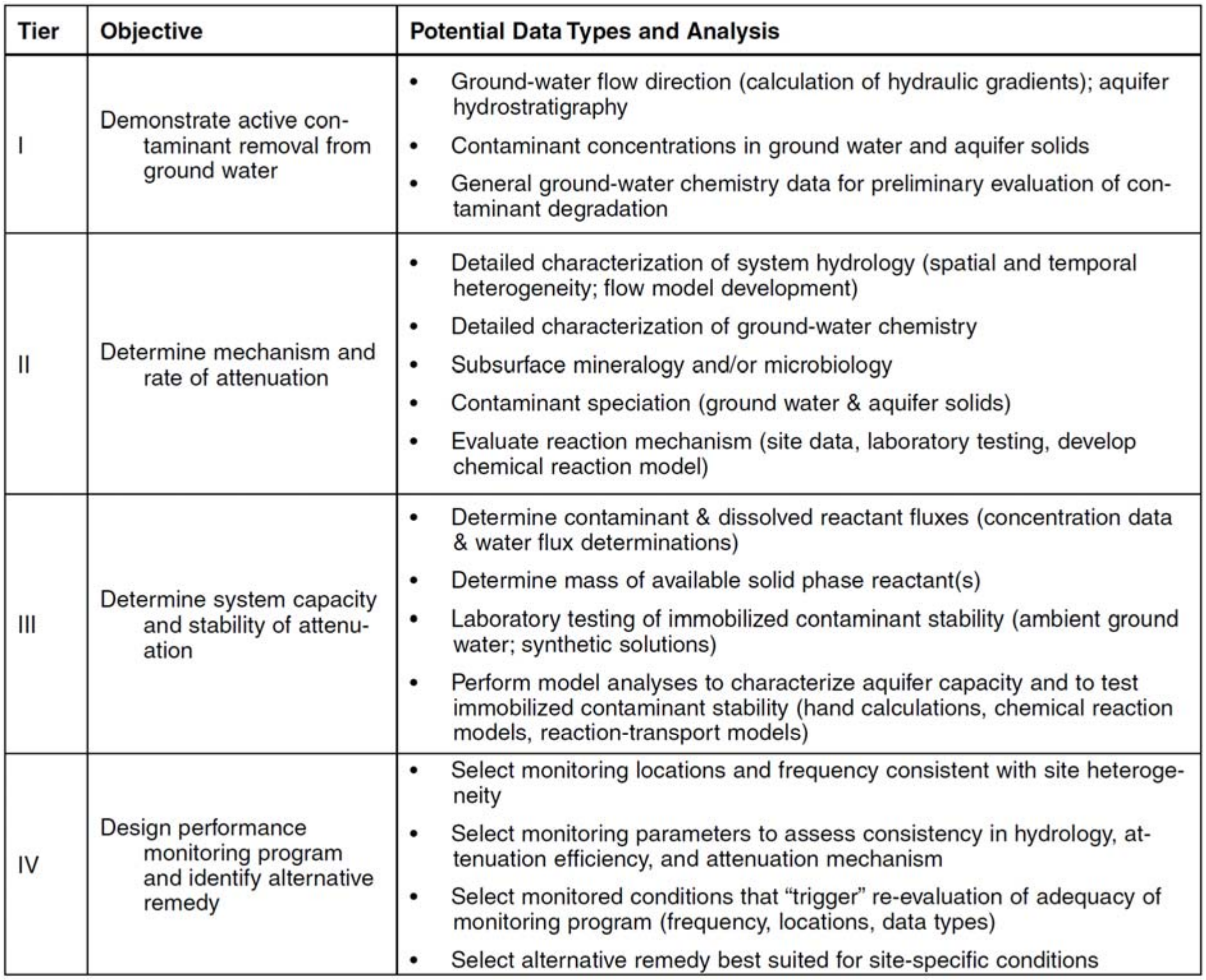
Au cours de la dernière décennie, plusieurs avancés scientifiques ont été réalisés par rapport à la fréquence et aux taux de dégradation de divers composés dans les panaches ou au niveau de l'interface des panaches avec l'aquifère. Le Tableau 2.4 fait état des principales études. De plus, certaines études ont évalué l'impact de panaches qui migrent dans des environnents hétérogènes et naturellement réducteurs, contrairement aux études classiques considérant un milieu homogène et oxydant (Milosevic et al. 2012; Cozzarelli et al. 2011; Lorah et al. 2009). Toutes ces études démontrent la complexité et la grande variabilité des réactions qui prennent place alors que le panache migre dans des contextes géologiques différents (Robertson et al. 2012; Bjerg et al. 2011, 2003; Cozzarelli et al. 2011; Sudicky et Illman 2011; Sudicky et al. 2010; Gutierrez-Neri et al. 2009; Lorah et al. 2009; USEPA 2008, 2007; van Breukelen et al. 2003; van Breukelen 2003; Kjeldsen et al. 2002; Christensen et al. 2001, 1994). Malgré ces nombreuses avancées scientifiques sur les mécanismes et les taux d'atténuation des panaches de lixiviat, un débat persiste dans la communauté de la réhabilitation de sites contaminés en ce qui concerne les critères d'évaluation nécessaires et suffisants d'un site contaminé. Une évaluation adéquate devrait offrir les réponses nécessaires, avec peu d'incertitude, sur le mode de réhabilitation requis pour un site spécifique (USEPA 2007; Rapti-Caputo et Vaccanro 2006; Rapti-Caputo et al. 2006; Wilson et al. 2005; Vangelas et al. 2005; Norris et al. 2004; Daugherty et al. 2004; Rittmann et al. 2004; Crumbling et al. 2003).

2.3 Évaluation de l'atténuation naturelle pour la gestion des anciens LES

L'atténuation naturelle comprend tous les processus physiques, chimiques et biologiques qui agissent sans l'intervention humaine et qui contribuent à la réduction de la masse, de la toxicité, de la mobilité, du volume ou de la concentration des contaminants (USEPA 2007). Dans les sites d'enfouissement, les contaminants du panache de lixiviat sont atténués naturellement par des processus tels que la dégradation, la sorption, la volatilisation, la précipitation et l'échange ionique. Selon la littérature scientifique consultée, les processus d'atténuation naturelle dans les panaches d'eau de lixiviation sont généralement efficaces et permettent la protection des aquifères. Les effets du panache sur l'eau souterraine seraient normalement limités à environ 1000 m des sites d'enfouissement (Christensen et al. 1994). En revanche, les LES peuvent émettre des contaminants dans l'environnement pour des décennies jusqu'à des siècles, et plusieurs contaminants peuvent être remobilisés. Les processus d'atténuation naturelle dépendent de l'importance de la source de contamination (en terme de concentration et de superficie) et de la composition chimique et minéralogique des matériaux de l'aquifère. La compréhension d'un site et de ses processus d'atténuation naturelle est nécessaire afin d'évaluer le risque environnemental pour les milieux récepteurs entourant un site d'enfouissement.

L'évaluation de l'atténuation naturelle des contaminants organiques et inorganiques d'un site d'enfouissement sanitaire est complexe puisque de nombreuses réactions chimiques, physiques et biologiques agissent simultanément au cours de la décomposition des déchets et de la migration du lixiviat. De plus, l’atténuation naturelle dépend de plusieurs facteurs : dimension du site, hétérogénéité physique et chimique du milieu, conditions d’écoulement, variété de contaminants émis (Christensen et al. 2001). Plusieurs techniques d’analyses doivent être combinées pour parvenir à la caractérisation complète des processus et des réactions en fonction de l’espace et du temps (USEPA 2007). En plus des données hydrogéochimiques, tous les processus géochimiques dominants doivent être considérés, car l’occurrence simultanée de certaines réactions peut masquer l’action d’importants mécanismes (Christensen et al. 2001). Il est donc important de développer de nouvelles approches de caractérisation et de suivis pour comprendre l’évolution future de la composition des eaux de lixiviation à travers le temps, car les eaux de lixiviation peuvent être rejetées dans l’environnement à n’importe quel moment dans le futur.

Tableau 2.5 : Étapes d’évaluation de l’atténuation naturelle
(USEPA 2007)



Tier	Objective	Potential Data Types and Analysis
I	Demonstrate active contaminant removal from ground water	• Ground-water flow direction (calculation of hydraulic gradients); aquifer hydrostratigraphy • Contaminant concentrations in ground water and aquifer solids • General ground-water chemistry data for preliminary evaluation of contaminant degradation
II	Determine mechanism and rate of attenuation	• Detailed characterization of system hydrology (spatial and temporal heterogeneity; flow model development) • Detailed characterization of ground-water chemistry • Subsurface mineralogy and/or microbiology • Contaminant speciation (ground water & aquifer solids) • Evaluate reaction mechanism (site data, laboratory testing, develop chemical reaction model)
III	Determine system capacity and stability of attenuation	• Determine contaminant & dissolved reactant fluxes (concentration data & water flux determinations) • Determine mass of available solid phase reactant(s) • Laboratory testing of immobilized contaminant stability (ambient ground water; synthetic solutions) • Perform model analyses to characterize aquifer capacity and to test immobilized contaminant stability (hand calculations, chemical reaction models, reaction-transport models)
IV	Design performance monitoring program and identify alternative remedy	• Select monitoring locations and frequency consistent with site heterogeneity • Select monitoring parameters to assess consistency in hydrology, attenuation efficiency, and attenuation mechanism • Select monitored conditions that “trigger” re-evaluation of adequacy of monitoring program (frequency, locations, data types) • Select alternative remedy best suited for site-specific conditions

L'USEPA (2007) propose 4 étapes pour l'évaluation de l'atténuation naturelle comme mode de gestion environnementale. Le Tableau 2.5 donne des exemples d'objectifs et de données requises pour statuer sur l'évaluation de l'atténuation naturelle en tant que mode de gestion (USEPA 2007). Les étapes d'évaluation sont les suivantes : 1) démontrer la décroissance de contaminants de l'eau souterraine et du panache; 2) déterminer les mécanismes et taux d'atténuation; 3) déterminer la capacité à long terme pour l'atténuation et l'immobilisation des contaminants et; 4) concevoir un suivi spécifique au site à l'étude et établir un plan en cas d'échec. Pour chacune des étapes, l'USEPA (2007) propose des sous objectifs et des analyses spécifiques. Ce document démontre que des études locales sont nécessaires pour déterminer les conditions d'atténuation naturelle spécifiques à un site en tenant compte de la distance d'un LES avec les milieux récepteurs qui l'entourent. La compréhension des processus d'atténuation naturelle spécifiques à un site est requise pour l'interprétation géochimique des échantillons de l'eau souterraine et pour la détermination d'actions de réhabilitation appropriées (Christensen et al. 1994). Enfin, deux éléments sont indispensables dans l'évaluation de l'efficacité de l'atténuation naturelle : des données chimiques de l'eau souterraine et du sol qui démontrent le déclin de la concentration des contaminants à travers le temps et des données hydrogéologiques et géochimiques qui permettent la compréhension des processus d'atténuation naturelle spécifiques au site à l'étude (USEPA 2011, 2008, 2007). Enfin, la compréhension des processus d'atténuation des contaminants du lixiviat est essentielle à l'évaluation du risque environnemental. D'autre part, la planification d'un réseau de suivi efficace dans le temps et le choix des méthodes de réhabilitation reposent sur cette compréhension des mécanismes et des réactions qui contribuent à la diminution des concentrations ou des flux de masse des contaminants du lixiviat.

Pour bien comprendre tous les processus, une étude détaillée incluant la quantification des processus de recharge, l'évaluation de l'écoulement de l'eau souterraine dans un milieu hétérogène et des voies de migration du lixiviat doit être réalisée en trois dimensions (USEPA 2011, 2007). Afin de déterminer les vitesses et les principales voies d'écoulement de l'eau souterraine et du panache, une telle étude doit également considérer les données sur les formations géologiques, les gradients, les perméabilités aussi en 3-D, ainsi que les limites physiques comme les unités géologiques imperméables, l'infiltration, les rivières et les puits. La modélisation numérique est surtout utilisée pour comprendre l'écoulement d'un système aquifère et aussi pour prédire comment ce même système va se comporter dans le futur. De plus, les modèles (n'importe quelle représentation d'un système réel) peuvent être utilisés pour analyser ou valider des hypothèses. De plus, les microorganismes semblent être importants pour faciliter l'oxydoréduction de panache (van Breukelen 2003; Kjeldsen 2002; Christensen et al. 2001). Il a été démontré que la détermination des processus microbiens peut accroître les connaissances sur la capacité d'atténuation d'un aquifère et permet d'identifier les eaux souterraines atteintes par le panache, car les communautés microbiennes sont plus sensibles que l'hydrogéochimie de l'eau pour détecter des concentrations faibles de lixiviat (Mouser et al, 2010). La quantification des taux de dégradation des composés du lixiviat par la reconnaissance de certaines bactéries par rapport aux conditions d'oxydoréduction le long des voies d'écoulement peut donc aider à statuer sur l'efficacité de l'atténuation naturelle (USEPA 2007).

Enfin, selon la littérature, les panaches de lixiviat demeurent étroits et la dilution est principalement influencée par les variations hydrogéologiques locales. Il est généralement reconnu que la dispersion transversale demeure faible pour des aquifères relativement homogènes (Christensen et al. 2001). Par contre, la dispersivité est aussi un facteur important à considérer dans l'évaluation de l'atténuation naturelle. La dispersivité est le terme dans l'équation de transport qui prend en compte la dilution ou le mélange en relation avec les gradients de concentration. La dispersivité a des composantes horizontale, verticale et transversale. La dispersivité est un paramètre difficile à évaluer quantitativement sur le terrain à cause du manque d'information sur la variabilité spatiale des conductivités hydrauliques (Christensen et al. 2001). Cette composante est souvent utilisée comme valeur de calibration dans les modèles numériques de transport de masse, tout en tenant compte de l'échelle considérée (Gelhar et al. 1992). Les variations de dispersivité reflètent l'influence de l'hétérogénéité dans un aquifère, ce qui rend la définition de l'hétérogénéité d'autant plus importante (USEPA 2011; Christensen et al. 2001).

2.4 Caractérisation des systèmes aquifères

La compréhension de l'écoulement de l'eau souterraine et de la migration des contaminants repose sur les deux aspects suivants : 1) la capacité de caractérisation des aquifères ; et 2) la représentation des processus à l'aide de simulateurs numériques. La description de l'hétérogénéité d'un aquifère est un des défis importants de la recherche en hydrogéologie (De Marsily et al. 2005). L'hétérogénéité d'un aquifère contrôle en grande partie la migration des contaminants et sa méconnaissance est souvent à l'origine des échecs des mesures de restauration. Au cours des dernières décennies, le développement continu des capacités de simulation numérique a permis de faire une représentation de plus en plus détaillée des systèmes d'écoulement complexes. Cependant, les méthodes conventionnelles de caractérisation hydrogéologique ne permettent pas l'acquisition de toutes les données pouvant être gérées par les simulateurs pour produire des modèles représentatifs de l'hétérogénéité des aquifères. Une meilleure caractérisation des aquifères exige l'utilisation de sources indirectes d'information sur la conductivité hydraulique qui sont plus nombreuses et plus continues que ce que fournissent les forages conventionnels, notamment les descriptions de forages (Ouellon et al. 2008) ou les levés géophysiques (Rubin et Hubbard 2005). Il est aussi souhaitable de développer les techniques permettant de générer un champ continu et détaillé des propriétés nécessaires aux modèles d'écoulement (Ouellon et al. 2008; Koltermann et Gorelick 1995).

Plusieurs méthodes géophysiques ont le potentiel d'évaluer la variabilité spatiale et l'anisotropie du milieu géologique en plus de contraindre les paramètres hydrauliques et améliorer les prédictions des modèles numériques (Sandberg et al. 2002). Le guide méthodologique proposé par Ressources naturelles Canada pour la caractérisation régionale des aquifères granulaires propose plusieurs types de levés géophysiques (Boivert et al. 2008). Contrairement aux méthodes de caractérisation conventionnelles, en plus de produire une image quasi continue des conditions souterraines, les méthodes géophysiques sont rapides, non invasives et peu coûteuses (Perri et al. 2012; Dogan et al. 2011). Par contre, chaque méthode a des avantages et des limites. Le Tableau 2.6 fait état d'études de caractérisation d'aquifères avec des méthodes géophysiques. Certaines des méthodes les plus communes sont discutées ci-dessous. Malgré son application pratique, la résolution verticale et latérale du géoradar (GPR) est limitée, réduite en fonction de la profondeur et contrainte par les matériaux conducteurs tels que l'argile (Turesson 2006). De plus, une corrélation entre les réponses du géoradar et les faciès hydrogéologique doit être établie (Niwas et al. 2011; Turesson 2006; Mosey et Knight 2002). La tomographie électrique (ERT) est fréquemment utilisée comme complément des levés géoradars (Chambers et al. 2006; Bentley et Gharibi 2004). Par contre, l'interprétation adéquate des levés ERT nécessite la disponibilité d'autres levés, tel que les sondages par enfoncement ou des analyses géochimiques de l'eau et du sol pour établir des valeurs de base. (Acworth et Jorstad 2006; Zume et al. 2006). La combinaison de la polarisation induite (IP) et le ERT a été testée pour leur capacité

Tableau 2.6 : Exemples de caractérisation avec méthodes géophysiques

Méthodes géophysiques: Détection d'anomalies qui pourraient indiquer la présence de contamination par du lixiviat par rapport aux conditions qui prévalent pour des eaux naturelles. Détermination des propriétés du matériel, de certaines conditions de l'aquifère, ou zonation d'une source de contamination.

Titre	**Auteurs**	**Date**
Application of time domain induced polarization to the mapping of lithotypes in a landfill site	Gazoty et al.	2012
Environmental impact and vulnerability of the surface and groundwater system from municipal solid waste disposal site: Koshe, Addis Ababa	Haile et Abiye	2012
Geophysical technique and groundwater monitoring to detect leachate contamination in the surrounding area of a landfill-Londrina (PR-Brazil)	Lopes et al.	2012
Three-dimensional hydrostratigraphic model from ground-penetrating radar and direct-push data	Schmelzbach et al.	2011
Lessons learned from a suite of CBC Borden experiments	Sudicky et Illman	2011
Detecting leachate plumes and groundwater pollution at Ruseifa municipal landfill utilizing VLF-EM method	Al-Tarazi et al.	2008
Detection and characterization of preferential flow paths in the downstream area of a hazardous landfill	Perozzi et Holliger	2008
Dense resistivity and induced polarization profiling for a landfill restoration project at Härlöv, Soutern Sweden	Leroux et al.	2007
Electrical resistivity imaging of conductive plume dilution in fractured rock	Nimmer et al.	2007
Integration of multi-channel piezometry and electrical tomography to better define chemical heterogeneity in a landfill leachate plume within a sand aquifer	Acworth et Jorstad	2006
Electrical resistivity tomography applied to geologic, hydrogeologic, and engineering investigation at a former waste-disposal site	Chambers et al.	2006
Mapping groundwater contamination around a lanfill facility using the VLF-EM method: a case study	Monteiro Santos et al.	2006
Subsurface imaging of an abandoned solid waste landfill site in Norman, Oklahoma	Zume et al.	2006
Overview of geological techniques to measure moisture content in landfills	Catley et al.	2006
Geochemical characterisation and geophysical mapping of Landfill leachates: the Marozzo canal case study (NE Italy)	Abu-Zeid et al.	2004
Integrated geophysical surveys on waste dumps: evaluation of physical parameters to characterize an urban waste dump (four case studies in Italy)	Cardarelli et Di Flippo	2004
Groundwater redox conditions and conductivity in a contaminant plume from geoelectrical investigations	Naudet et al.	2004
Two and three-dimensional electrical resistivity imaging at a heterogeneous remediation site	Bentley et Gharibi.	2004
Monitoring of Landfill Leachate Dispersion Using Reflectance Spectroscopy and Ground-Penetrating Radar	Splajt et al.	2003
Hydrogeology and leachate plume delineation at a closed municipal landfill, Norman, Oklahoma	Becker	2001
DC resistivity and induced polarisation investigations at a waste disposal site and its environments	Aristodemou et al.	2000

d'investigation de l'hétérogénéité de la sous-surface (Gazoty et al. 2012; Perri et al. 2012; Leroux et al. 2007; Chambers et al. 2006; Sandberg et al. 2002). De plus, la bonne corrélation entre divers levés géophysiques a facilité le développement de modèles conceptuels détaillés du transport de contaminants à plusieurs sites (Gasoty et al. 2012; Dogan et al. 2011; Acworth et Jorstad 2006).

Les méthodes géophysiques sont aussi utilisées pour détecter des anomalies qui pourraient indiquer la présence de contamination par du lixiviat par rapport aux conditions qui prévalent pour des eaux naturelles. Un panache de lixiviat peut être assez facilement détecté par différentes méthodes géophysiques à cause de la concentration élevée des eaux de lixiviation qui augmente considérablement sa conductivité électrique par rapport à celle de l'eau souterraine. D'autres travaux de géophysique ont visé à déterminer les propriétés du matériel, certaines conditions de l'aquifère, ou le zonage d'une source de contamination (Gasoty et al. 2012; Lopez et al. 2012; Al-Tarazi et al. 2008; Perozzi et Holliger 2008; Cardarelli et Di Filippo 2007; Acworth et Jorstad 2006; Monteiro Santos et al. 2006; Zume et al. 2006; Abu-Zeid et al. 2004; Naudet et al. 2004; Splajt et al. 2003; Becker 2001; Aristodemou et Thomas-Betts 2000).

Enfin, les études intégrées font appel à plusieurs méthodes qui, combinées, permettent de mieux définir les conditions rencontrées dans l'aquifère et d'identifier les processus géochimiques qui prévalent. La combinaison de plusieurs méthodes est utile pour imager la géologie de la sous-surface. Par contre, des mesures directes de la conductivité hydraulique sont aussi nécessaires pour définir les hydrofacies et établir les relations qui existent entre les mesures indirectes et les propriétés hydrauliques. Cela est important, car ce sont les valeurs de conductivité hydraulique qui sont requises pour le développement de modèles numériques (Ruggeri et al. 2013b; Paradis et al. 2011a; Ouellon et al. 2008). Plusieurs études suggèrent l'utilisation de la tomographie hydraulique pour fournir de l'information sur la variabilité spatiale des propriétés hydrauliques (e.g., Berg et Illman 2011; Zhu et Yeh 2005; Brauchler et al. 2003; Bohling et al. 2002). Par contre, la tomographie hydraulique n'est pas entrées dans la pratique et est demeurée dans le domaine de la recherche, car elle implique des opérations de terrain relativement complexes, longues et coûteuses, elle nécessite des spécialistes pour l'interprétation des données en plus de seulement permettre de caractériser un volume restreint de l'aquifère (Berg et Illman 2013 Bohling et Butler 2010). Comme toutes autres méthodes géophysiques, la tomographie

Tableau 2.7 : Exemples d'étude utilisant des traceurs environnementaux 1

Méthodes géochimiques: Utilisation de traceurs géochimiques pour identifier la provenance de l'eau, ses voies de migration, les transformations géochimiques qui l'affectent ainsi que pour estimer l'âge de l'eau souterraine ou pour ajuster un modèle d'écoulement.		
Titre	**Auteurs**	**Date**
Identifying the recharge sources and age of groundwater in the Songnen Plain (Northeast China) using environmental isotopes	Chen et al.	2011
Calibration of models using groundwater age	Sanford	2011
Age structure and recharge conditions of a costal aquifer (northern Germany) investigated with ^{39}Ar, ^{14}C, ^{3}H, He isotopes and Ne	Sültenfuβ et al.	2011
Using geochemical indicators to investigate groundwater mixing and residence time in the aquifer system of Djeffara of Medenine (southeastern Tunisia)	Zouari et al.	2011
On some methodological problems in the use of environmental tracers to estimate hydrogeologic parameters and to calibrate flow and transport model	Zuber et al.	2011
Tritium-helium groundwater age used to constrain a groundwater flow model in a TCE-contaminated valley fill aquifer	Murphy et al.	2011
Identification of groundwater recharge sources and processes in a heterogeneous alluvial aquifer: results from multi-level monitoring of hydrochemistry and environmental isotopes in a riverside agricultural area in Korea	Choi et al.	2010
Ambient well-bore mixing, aquifer cross-contamination, pumping stress, and water quality from long screened wells: what is sampled and what is not?	Mayo.	2010
Direct-push profiling of isotopic and hydrogeochemical vertical gradients	Hofmann et al.	2010
Groundwater flow regime, recharge and regional-scale solute transport in the semi arid Kalahari of Botswana derived from isotope hydrology and hydrochemistry	Stadler et al.	2010
Forensic isotope analysis to refine a hydrologic conceptual model	Bassett et al.	2008
Distinguishing sources of ground water recharge by using δ^{2}H and δ^{18}O	Blasch et al.	2007
Isotope application in environmental investigation part II :groundwater age dating and recharge processes, and provenance of sulfur and methane	Sueker	2003
Groundwater recharge and agricultural contamination	Böhlke	2002
The use of simulation and multiple environmental tracers to quantify groundwater flow in shallow aquifer	Reilly et al.	1994
A validation of the ^{3}H/^{3}He method for determining groundwater recharge	Solomon et al.	1993
Tritium and Helium as groundwater age tracers in the Borden aquifer	Solomon et al.	1992

hydraulique ne peut donc à elle seule fournir les données requises pour la caractérisation d'un aquifère à l'échelle régionale. À part les données hydrauliques, les données géochimiques peuvent fournir des indications sur les voies d'écoulement et aider à comprendre l'évolution de l'eau souterraine et l'atténuation des contaminants (Glynn et Plummer 2005; Edmunds 2009). Les traceurs géochimiques peuvent ainsi fournir des données indépendantes, qui complémentent les données hydrauliques, pour correspondre aux données historiques et contraindre un modèle numérique pour obtenir une représentation plus réaliste de l'écoulement de l'eau souterraine avec de meilleures capacités de prédiction (Murphy et al. 2011; Sanford 2011; Zuber et al. 2011; Reilly et al. 1994). En retour, la modélisation numérique hydrogéologique est utilisée pour mieux comprendre les conditions d'écoulement, interpréter les données géochimiques et représenter les transformations qui se produisent dans un système d'écoulement. L'application des traceurs géochimiques est discutée plus en détail dans la section suivante.

2.5 Traceurs géochimiques

De nos jours, les isotopes environnementaux sont utilisés non seulement pour retracer l'origine de l'eau souterraine, mais aussi pour identifier les processus et les mécanismes de recharge, l'évolution naturelle de l'eau souterraine, les réactions géochimiques et les taux de réactions en relation avec une source de contamination. Le Tableau 2.7 énumère quelques études qui ont utilisé les traceurs environnementaux pour comprendre le cycle naturel de l'eau, tandis que le Tableau 2.8 rapporte certaines études qui ont utilisé les traceurs environnementaux pour identifier une source de contamination ou définir les processus de réactions/dégradation.

Dans les systèmes d'écoulement où on retrouve des eaux souterraines de différentes origines et possédant des signatures géochimiques distinctes, la caractérisation de paramètres géochimiques peut donner des informations complémentaires sur les conditions de l'écoulement. Les études géochimiques font généralement l'utilisation de traceurs géochimiques pour identifier la provenance de l'eau, ses voies de migration, les transformations géochimiques qui l'affectent ainsi que pour mesurer l'âge de l'eau souterraine et estimer les taux de transformation des composés chimiques. Les traceurs géochimiques peuvent être utilisés afin de préciser certains paramètres d'écoulement de l'eau souterraine tels que l'âge de l'eau, le temps de résidence de l'eau dans les différentes formations géologiques, les zones de recharge et les principales voies d'écoulement de l'eau (Clark et Fritz 1997). Les isotopes radioactifs comme le tritium peuvent être utilisés pour la datation de l'eau souterraine et l'estimation de la recharge (Solomon et al. 1993). La composition des isotopes stables dans l'eau souterraine reflète leur origine et peut donner une indication des conditions de recharge (Clark et Fritz 1997). Certaines caractéristiques de la géochimie de l'eau (ions majeurs, pH, Eh et alcalinité) peuvent aussi fournir de l'information sur l'origine de l'eau et les principales voies de migration. Donc, un modèle conceptuel du système d'écoulement de l'eau souterraine peut être mieux défini par la composition isotopique et les informations géochimiques. De plus, la disponibilité de paramètres

Tableau 2.8 : Exemples d'étude utilisant des traceurs environnementaux 2

Méthodes géochimiques: Utilisation de traceurs géochimiques pour détecter ou identifier une source de contamination ou des processus de dégradation dans l'eau souterraine		
Titre	**Auteurs**	**Date**
Identification of discharge zones and quantification of contaminant mass discharges into a local stream from a landfill in a heterogeneous geological setting	Milosevic et al.	2012
Natural attenuation of septic system nitrogen by anammox	Robertson et al.	2012
Natural attenuation processes in landfill leachate plumes at three Danish sites	Bjerg et al.	2011
Biogeochemical evolution of a landfill leachate plume, Norman, Oklahoma	Cozzarelli et al.	2011
Bioattenuation in groundwater impacted by landfill leachate traced with 13C	Mohammadzahed et al.	2011
Biochemistry at a wetland sediment-alluvial aquifer interface in a landfill leachate plume	Lorah et al.	2009
Statistical evaluation of groundwater quality around an uncontrolled landfill: implication for plume migration pathways	Park et al.	2008
The use of tritium content as an indicator of the groundwater contamination by sanitary landfills leachates in the region of Belo Horizonte City, Brazil	Bandeira et al.	2008
Origin and fate of industrial ammonium in anoxic ground water – 15 N evidence for anaerobic oxidation (Anammox)	Clark et al.	2008
Human impacts on groundwater flow and contamination deduced by multiple isotopes in Seoul City, South Korea	Hosono et al.	2008
Monitored natural attenuation forum: the use of carbon isotope analysis at MNA sites	Philip et al.	2007
Ammonium transport and reaction in contaminated groundwater: application of isotopic tracers and isotope fractionation studies	Böhlke et al.	2006
Geochemical evidences of landfill leachate in groundwater	Rapti-Caputo et al.	2006a
Can stable isotopes be used to monitor landfill leachate impact on surface water?	North et al.	2006
Recharge processes drive sulfate reduction in an alluvial aquifer contaminated with landfill leachate	School et al.	2006
Compound specific isotopic analysis (CSIA) of landfill leachate DOC components	Mohammadzahed et Clark	2005
Isotope and dissolved gas evidence for nitrogen attenuation in landfill leachate dispersing into a chalk aquifer	Heaton et al.	2005
The groundwater geochemistry of waste disposal facilities	Bjerg et al.	2003
Biogeochemistry and isotope geochemistry of a landfill leachate plume	van Breukelen et al.	2003
Natural attenuation of landfill leachate: a combined biogeochemical process analysis and microbial ecology approach	van Breukelen et al	2003
In situ biodegradation determined by carbon isotope fractionation of aromatic hydrocarbons in an anaerobic landfill leachate plume (Vejen, Denmark)	Richnow et al.	2003
Biogeochemistry of landfill leachate plumes	Christensen et al.	2001
A multitracer study in a shallow aquifer using age dating tracers ^{3}H, ^{85}Kr, CFC-13 and SF-6 – Indication for retarder transport of CFC-13	Bauer et al.	2001

hydrauliques et géochimiques peut permettre de valider un modèle numérique et le rendre plus représentatif des conditions d'écoulement de l'eau souterraine (Reilly et al. 1994).

Les traceurs géochimiques peuvent être d'une grande utilité dans une étude de caractérisation hydrogéologique. Ils ont la capacité de révéler des informations qui permettent de mieux comprendre les dynamiques d'écoulement d'un système hydrogéologique. Par contre, ils doivent être choisis soigneusement. Les traceurs doivent être sélectionnés en fonction des réponsescherchées et par rapport au contexte du milieu hydrogéologique investigué. Par exemple, dans les sites d'enfouissement sanitaire la compréhension du cycle de l'azote est primordiale. L'ammonium (NH_4) est l'un des constituants majeurs retrouvés dans les eaux de lixiviation qui proviennent de la dégradation des protéines retrouvées dans la matière organique contenue dans les déchets. Plusieurs études hydrogéologiques effectuées sur les sites d'enfouissement sanitaire rapportent des concentrations d'ammonium dans l'eau souterraine de 500 à 2000 mg/L et démontrent que ces concentrations ne décroissent pas de façon significative sur une période de 30 années. Pour cette raison, l'ammonium a été identifié comme l'un des contaminants les plus persistants dans l'environnement, et ce même dans les anciens sites d'enfouissement qui contiennent désormais de faibles teneurs en matière organique (Kjeldsen et al. 2002). En contrepartie, l'ammonium peut aussi être présent naturellement dans l'eau souterraine, mais en très faible quantité, de l'ordre de moins de 1 mg/L, et potentiellement à des concentrations de plus de 2 mg/L dans l'eau de certains types de paléosol (Glesser et Roy 2009). Le transport de l'ammonium dans les aquifères par l'entremise de l'eau souterraine est complexe, et malgré plusieurs études récentes sur le sujet, les mécanismes d'immobilisation et les processus de dénitrification de l'ammonium dans les environnements réducteurs dus à la contamination anthropique ne sont pas entièrement compris (Robertson et al. 2012; Cozzarelli et al. 2011; Lorah et al. 2009; Böhlke et al. 2006; Heaton et al. 2005). L'ammonium peut être retardé par des processus physiques telle l'adsorption (incluant l'échange cationique) ou par la transformation par les bactéries (Cozzarelli et al. 2011). De plus, l'ammonium peut-être oxydé conjointement à la réduction de l'oxygène, produisant nitrite et nitrate. Finalement, l'oxydation de l'ammonium peut aussi s'effectuer en condition anaérobie (réaction anammox avec l'aide de bactérie de la division des planctomycétales) par la réduction du nitrite et du nitrate produisant ainsi du N_2 (Robertson et al. 2012). Lorsque cette réaction a lieu, un enrichissement de ^{15}N est observé dans l'eau souterraine. L'abondance naturelle des isotopes stables de l'azote en combinaison avec d'autres analyses chimiques peut procurer des informations valables sur les processus qui affectent le transport de l'ammonium. Selon les études de Cozzarelli et al. (2011), Lorah et al. (2009), Böhlke et al. (2006) et Heaton et al. (2005), la masse principale de NH_4 qui voyage le long des lignes d'écoulement était retardée par un facteur de 2.5 à 6 fois par rapport à d'autres composés plus mobiles dans l'eau souterraine. Certaines études concluent qu'il n'y a pas de fractionnement isotopique du ^{15}N en condition anaérobie, et les valeurs reportées de $\delta^{15}N$ varient entre +4 et + 12 pour les eaux avec des concentrations de plus de 100 mg/L d'ammonium. D'autres études démontrent qu'il y a un faible fractionnement négatif lors de l'adsorption par échange cationique de l'ammonium sur les sédiments (Cozzarelli et al. 2011). La Figure 2.4 montre la variabilité des valeurs de $\delta^{15}N$ en relation avec les concentrations d'ammonium dans l'eau souterraine. Cette figure illustre les effets isotopiques contrastants entre de l'eau souterraine échantillonnée à faible profondeur par rapport à des profondeurs plus importantes dans un aquifère contaminé par du lixiviat (Cozzarelli et al. 2011).

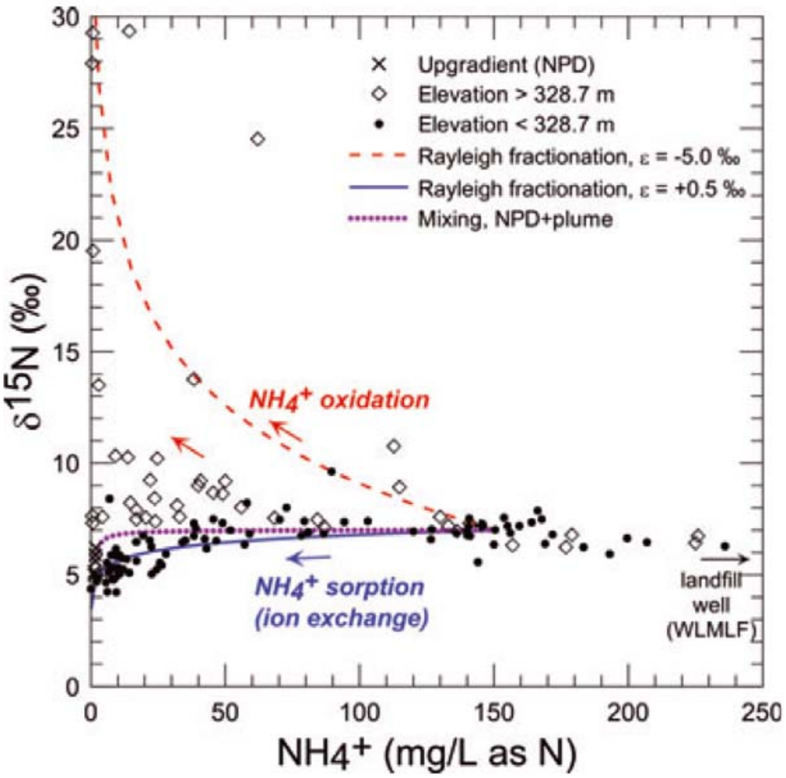


Figure 2.4 : Variabilité du $\delta^{15}N$ de l'ammonium émis par un site d'enfouissement. Cette figure illustre les effets isotopiques sur le $\delta^{15}N$ de l'ammonium dans l'eau souterraine échantillonné à faible et grande profondeur dans un aquifère contaminé (Cozzarelli et al. 2011).

Chapitre 3

RÉGION D'ÉTUDE

3.1 Contexte général

Ce chapitre fait la synthèse des différentes caractéristiques de la région d'étude. Les conditions décrites sont basées sur des informations tirées de documents publics et privés disponibles avant ou en cours de réalisation du projet doctoral. Ces divers documents fournissent des données de types variées qui ont été intégrées et validées à la suite de leur synthèse afin d'illustrer ce portrait général de la région d'étude. La synthèse de ces informations permet de se familiariser avec les conditions retrouvées dans la zone investiguée et de faire le bilan des connaissances actuelles par la même occasion. Les articles de la partie 2 de la thèse vont faire la description des connaissances acquises sur la région d'étude à la suite des travaux réalisés. Cette étape de synthèse de l'information disponible réalisée en début de projet a permis de mieux planifier le type et la localisation des travaux de caractérisation requis pour parfaire la compréhension du système hydrogéologique à l'étude.

La région d'étude est située dans le sous-bassin versant de la rivière Beaurivage qui fait partie du bassin versant de la rivière Chaudière de la région administrative Chaudière-Appalaches. Le bassin versant de la rivière Chaudière s'étale du fleuve St-Laurent, au nord, jusqu'aux limites du Québec avec les États-Unis, au sud (Figure 3.1). Cette étude vise spécifiquement la région entourant l'ancien lieu d'enfouissement sanitaire (LES) de St-Lambert-de-Lauzon localisé dans le sous-bassin versant de la rivière Beaurivage en bordure de la limite de partage des eaux avec le bassin versant de la rivière Chaudière (Figure 3.1). La région d'étude délimitée autour de l'ancien site couvre une superficie de 12 km^2 et chevauche la MRC de Lévis (Concessions de Pétrée) et la MRC de la Nouvelle-Beauce (Concession de Belvèze).

3.2 Physiographie, localisation et accès

Le site d'enfouissement sanitaire de Saint-Lambert-de-Lauzon se situe à environ 35 km au sud-ouest du centre-ville de Québec. L'entrée principale est localisée au 517, rue St-Aimé à Saint-Lambert-de-Lauzon. Le site d'enfouissement sanitaire est facilement accessible par les routes primaires et la région d'étude est accessible par des routes secondaires et par des chemins

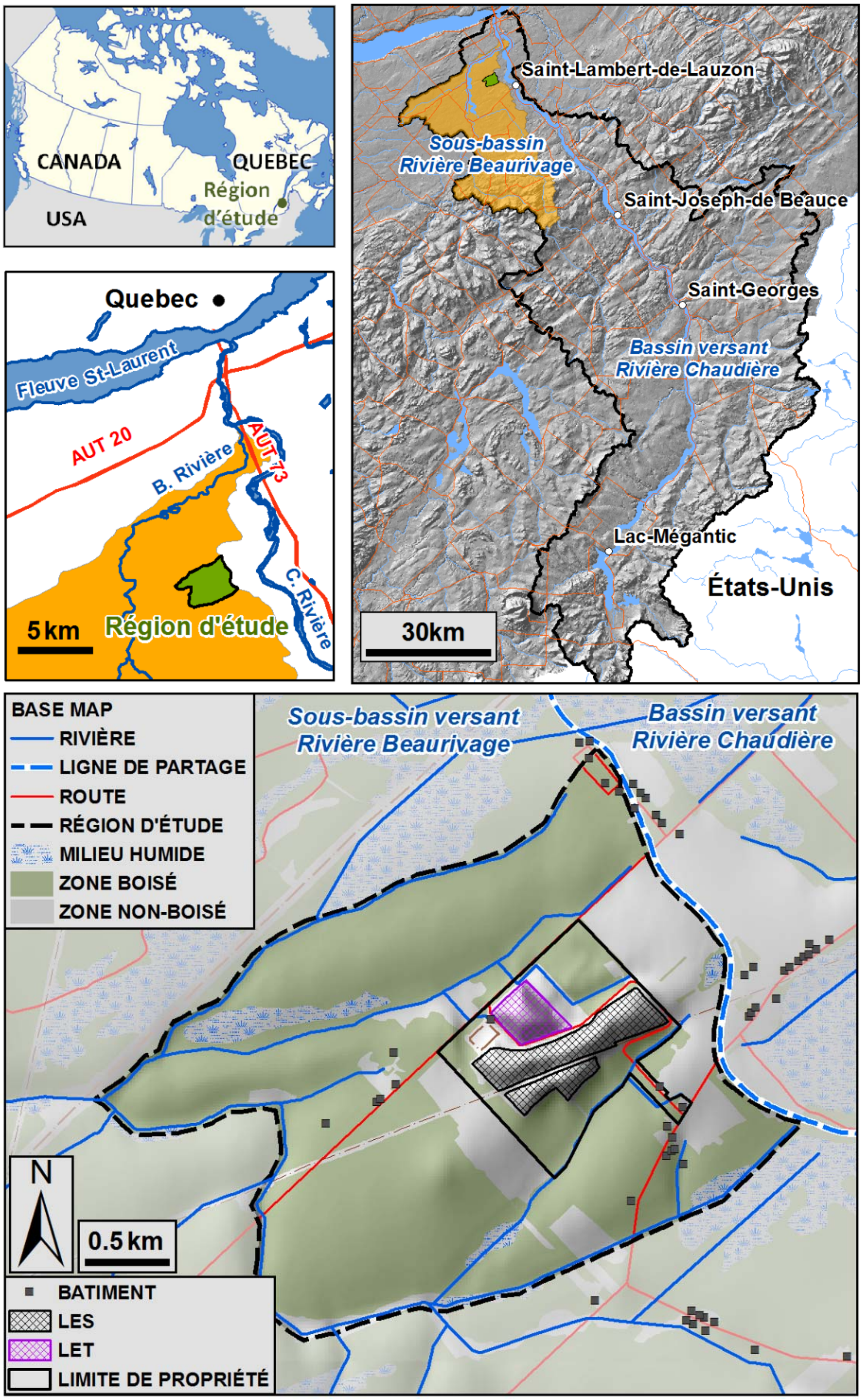


Figure 3.1 : Physiographie, localisation et accès de la région d'étude

forestiers privés. La Figure 3.1 présente la physiographie, les limites du LES fermé, du lieu d'enfouissement technique (LET) actuellement en exploitation, les limites du territoire à l'étude ainsi que les cours d'eau localisés dans le sous-bassin.

La zone investiguée d'une superficie d'environ 12 km^2 est délimitée par la ligne de partage des eaux entre le bassin versant de la rivière Chaudière et le sous-bassin versant de la rivière Beaurivage à l'est, et selon la physiographie des cours d'eau environnants partout ailleurs. Au sud et au nord, la zone d'étude est délimitée par de petits cours d'eau et la limite ouest est définie par la rivière Cugnet. La rivière Cugnet reçoit l'eau de tous les ruisseaux de la zone investiguée, son exutoire est localisé quelques km au nord-ouest où elle se déverse dans la rivière Beaurivage. La topographie générale du secteur est régulière et relativement plane entre 109 m et 124 m d'altitude. La zone d'enfouissement modifie la topographie locale, avec une hauteur atteignant 130 m d'altitude. L'utilisation du territoire est majoritairement l'exploitation forestière, mais il y a aussi quelques champs qui servent à l'agriculture. Entre autres, une ferme porcine est située à l'extrémité est des limites de propriété du site. Enfin, à l'extérieur de la zone investiguée, il y a des zones de tourbières et plusieurs zones sable et de gravier en exploitation.

3.3 Origine et type de données disponibles sur le sous-bassin versant

Le Tableau 3.1 résume les sources d'information initialement consultées. Plusieurs études et cartes géologiques à diverses échelles portant sur la géologie du socle rocheux et sur la géologie du Quaternaire. L'annuaire des granulats de la rive sud et la base de données du SIH ont aussi été des ressources pour l'étude. De plus, une étude hydrogéologique régionale sur le bassin versant de la rivière Chaudière a été réalisée en 2007-2008 (COBARIC et UPA 2008), et suivie de travaux complémentaires d'hydrogéochimie (Benoit et al. 2012, 2011, 2009) et de modélisation numérique (Brun Koné 2012; Brun Koné et al. 2011). De nombreuses études environnementales réalisées au site d'enfouissement au cours des années d'exploitation et d'autres sur les tourbières et les gravières en exploitation dans les environs nous ont également été fournies (Tableau 3.1). Celles-ci cumulent de nombreuse données de forages, des perméabilités pour les matériaux composant l'aquifère granulaire et des données sur la géochimie de l'eau souterraine sur la propriété du site d'enfouissement. L'étude a aussi pu bénéficier de la collaboration des municipalités pour l'accès à l'information des cadastres et aux photos aériennes les plus récentes.

Tableau 3.1 : Synthèse des données disponibles sur la région à l'étude

Sujet	Documents consultés
Géologie du Roc	1. Carte de la géologie du roc (St-Julien 1995) 2. Carte de compilation géologique, Québec-Chaudière.(Castonguay et al. 2002) 3. Histoire géologique et stratigraphie (Lavoie 2002) 4. Mineralogie (Ogunyomi et al. 1980)
Hydrogéologie du Roc	Benoit et al. (2012, 2011, 2009); Brun Koné (2012); Brun Koné et al. (2011); COBARIC et UPA (2008); Gauthier et al. 1993.
Géologie du Quaternaire	1. Carte de la géologie du Quaternaire (Bolduc 2003) 2. Variabilité de la composition lithologique des nappes de till sur le piémont appalachien, Saint-étienne-de Lauzon, Québec (Ruest 2002) 3. Late Wisconsinian Deglaciation and Champlain Sea Invasion in the St-Lawrence Valley, Québec. (Parent et Occhietti 1988) 4. Inventaire des ressources en granulats de la région de Charny 921L/11 (Brazeau 1999)
Pédologie	Étude pédologique du comté de Lévis (Laplante 1962). Feuillet pédologique de l'IRDA 1:50 000 (MAPAQ 2008).
Puits d'approvisionnement en eau potable	Système d'information hydrogéologique (SIH)
Études hydrogéologiques sur le LES	Gouvernement du Québec (1971) ; Gouvernement du Québec (1974) ; Carrier et al. (1978); Geomines Ltée (1981) ; Georoche Ltée (1985) ; Géohydrotek inc. et Robert Hamelin & Associes Inc. (1989) ; Gauthier & Guillemette Consultant Inc., Dessau Environnement Limitée et Serrener Consultation Inc. (1993a) ; Laboratoire de matériaux de Québec Inc. (1993); Gauthier & Guillemette Consultant Inc., Dessau Environnement Limitée et Serrener Consultation Inc. (1993b) ; Consultants H.G.E Inc. (1998) ; LVM Technisol (2008) RIGDCC-Régie municipale de gestion des déchets des Chutes-de-la-Chaudière(2012).
Études hydrogéologiques sur la tourbière des portugais	Envir'eau PUITS (2012)
Photos aériennes	Photocartothèque : (1965) Q65352-94-98; Q65359-38 et ortho photos Nouvelle-Beauce (2007) : Q01102-037, 039, 055, 057, 059, 061, 137, 139, 141, 143

3.4 Données météorologiques

Plusieurs stations météorologiques d'Environnement Canada sont situées dans un rayon de 30 km entourant la région d'étude (Figure 3.2). Il y a aussi une ferme expérimentale de l'Institut de recherche et de développement en agroenvironnement (IRDA) qui possède une station météorologique depuis 2006 dans la ville de St-Lambert-de-Lauzon. La station est localisée à moins à 5 km directement à l'est du site d'enfouissement de la région d'étude. Par contre, il y a beaucoup de données de précipitation et de température manquantes et la station ne fait pas le suivi des données des précipitations hivernales. En ce qui concerne les stations météorologiques d'Environnement Canada, la station de Beauséjour (Identification : 7020567) est la plus proche, au nord-est à 10 km du site d'enfouissement, et elle possède les données les plus complètes, sur la plus longue période que les autres stations (depuis 1978). La station est située à une élévation de 106.7 m comparable aux élévations de la zone d'étude (130 m à 108 m). Les données sont facilement accessibles via le site internet d'archives nationales d'information et de données climatologiques d'Environnement Canada (Environnnement Canada 2013). Les données mensuelles journalières sont habituellement disponibles dans les quatre mois suivant les dates d'enregistrement.

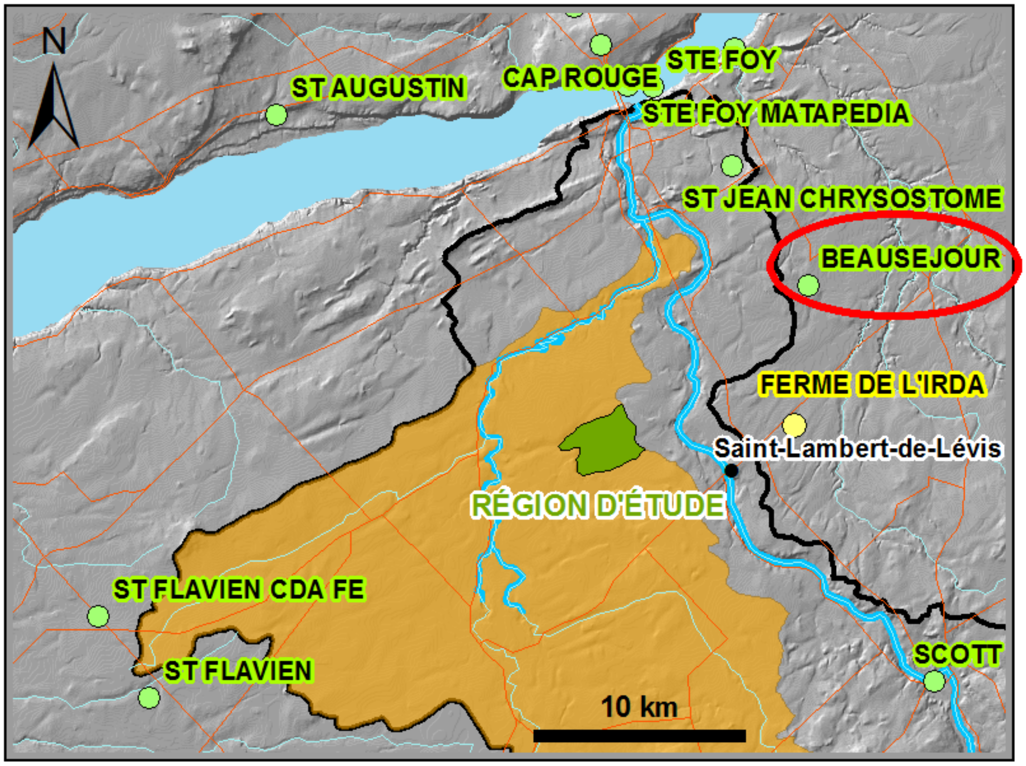


Figure 3.2 : Localisation des stations météorologiques à proximité de la région d'étude Les stations d'Environnement Canada sont en vert et celle de la ferme de l'IRDA en jaune. La station de Beauséjour est la plus proche à 10 km et possède les données les plus complètes sur la plus longue des périodes d'observation des stations du secteur, soit depuis 1978.

3.5 Pédologie

Dans cette région relativement plane, les sols sont de podzols mal drainés d'origine marine. L'écoulement de l'eau souterraine est influencé par des cours d'eau naturels ainsi que par un réseau de fossés de drainage agricole et forestier. Selon les études pédologiques réalisées par l'IRDA, il y aurait cinq types de sols présents dans la région d'étude (Figure 3.3). La figure illustre les types prédominants : sables de Saint-Jude (sols sableux d'origine marine), suivis des sols graveleux nommés Beaurivage, de loam sableux, de la tourbe grossière, des sables de Sorel fin et de la terre noire bien décomposée.

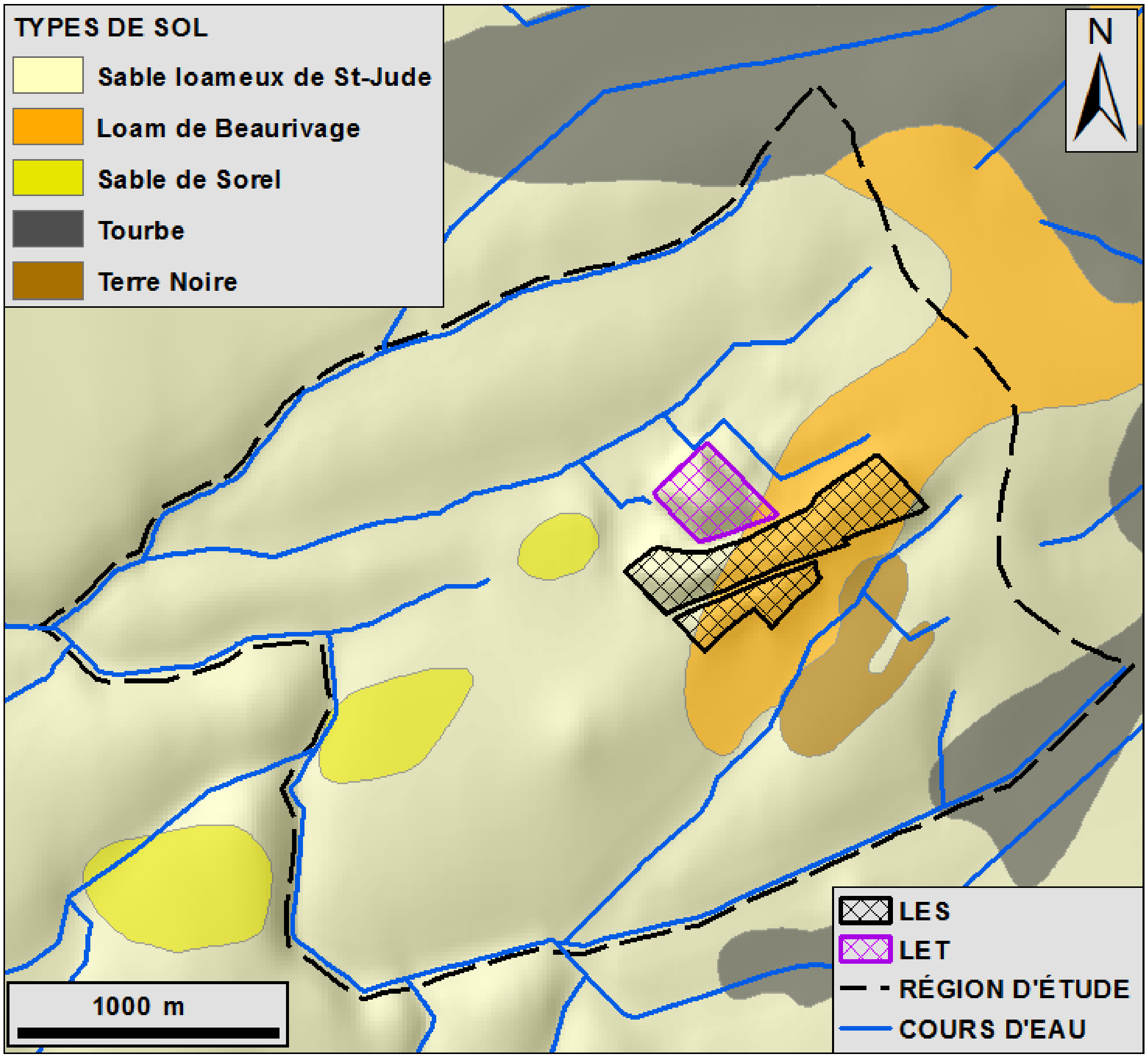


Figure 3.3 : Carte pédologique

Modifié des cartes pédologique 1:50000 (MAPAQ 2008)

3.6 Géologie et hydrogéologie

Il y existe deux types d'aquifères dans la région d'étude, un aquifère régional dans le roc fracturé qu'on retrouve à environ 20 m de profondeur et un aquifère dans les dépôts granulaires en surface. Ces deux aquifères sont séparés par des dépôts glaciaires peu perméables (aquitard). Les dépôts quaternaires dans la région de St-Lambert reposent sur un socle rocheux particulièrement accidenté composé principalement de roches sédimentaires (grès, schistes à blocs, conglomérats et calcaires) du Paléozoïque inférieur et de l'Ordovicien inférieur et moyen (Hocf et Dubé 1994; Landry et Mercier 1983). L'orogenèse appalachienne a modelé ce socle en imposant un alignement NE/SW dans la structure du roc. Les nombreuses périodes glaciaires qui ont suivi ont érodé ce socle rocheux jusqu'à son élévation actuelle. Il est recouvert de sédiments mis en place lors de plusieurs épisodes glaciaires, du Wisconsinien à l'Oligocène, appartenant aux domaines glaciaires, marins, lacustres et fluviaux. Dans le secteur de St-Lambert, ce sont les sédiments liés à la dernière glaciation qui dominent. L'aquifère sableux est à nappe semi-captive et la nappe est située très près de la surface du sol, souvent à moins de 1 m de profondeur (Gauthier et al. 1993).

3.6.1 Dépôts meubles

Les dépôts meubles dans la région d'étude sont principalement d'origine glaciaire et marine. La Figure 3.4 présente la géologie du Quaternaire du site. La carte a été entièrement révisée en fonction des besoins et de l'échelle de la région d'étude (Lamarche et Tremblay 2012). La carte est sous la responsabilité de Lamarche et Tremblay; à l'extrémité est de la région d'étude, près de l'ancien site d'enfouissement sanitaire, des remontées de till ont été identifiées sur les photos aériennes. Quoiqu'il existe d'autres indications de ces remontées de matériaux glaciaires (réflecteurs sur les lignes géoradars), des forages récents ont démontré l'absence de till un peu plus au sud. Les interprétations pour la partie est de la région d'étude doivent être davantage investiguées.

En général, les formations superficielles correspondent à des tills et sédiments fluvioglaciaires (sables et graviers), des sédiments marins (argiles de la Mer de Champlain, sédiments littoraux) et à des sédiments fluviatiles de plaine alluviale. Des tourbières se sont développées sur certaines de ces surfaces. De plus, l'incision du réseau de drainage actuel a laissé de nombreuses marques dans le paysage sous forme de terrasses le long des cours d'eau principaux (Ruest 2002). Des dépôts plus anciens sont également observables à proximité de la zone d'étude, mais ce n'est que

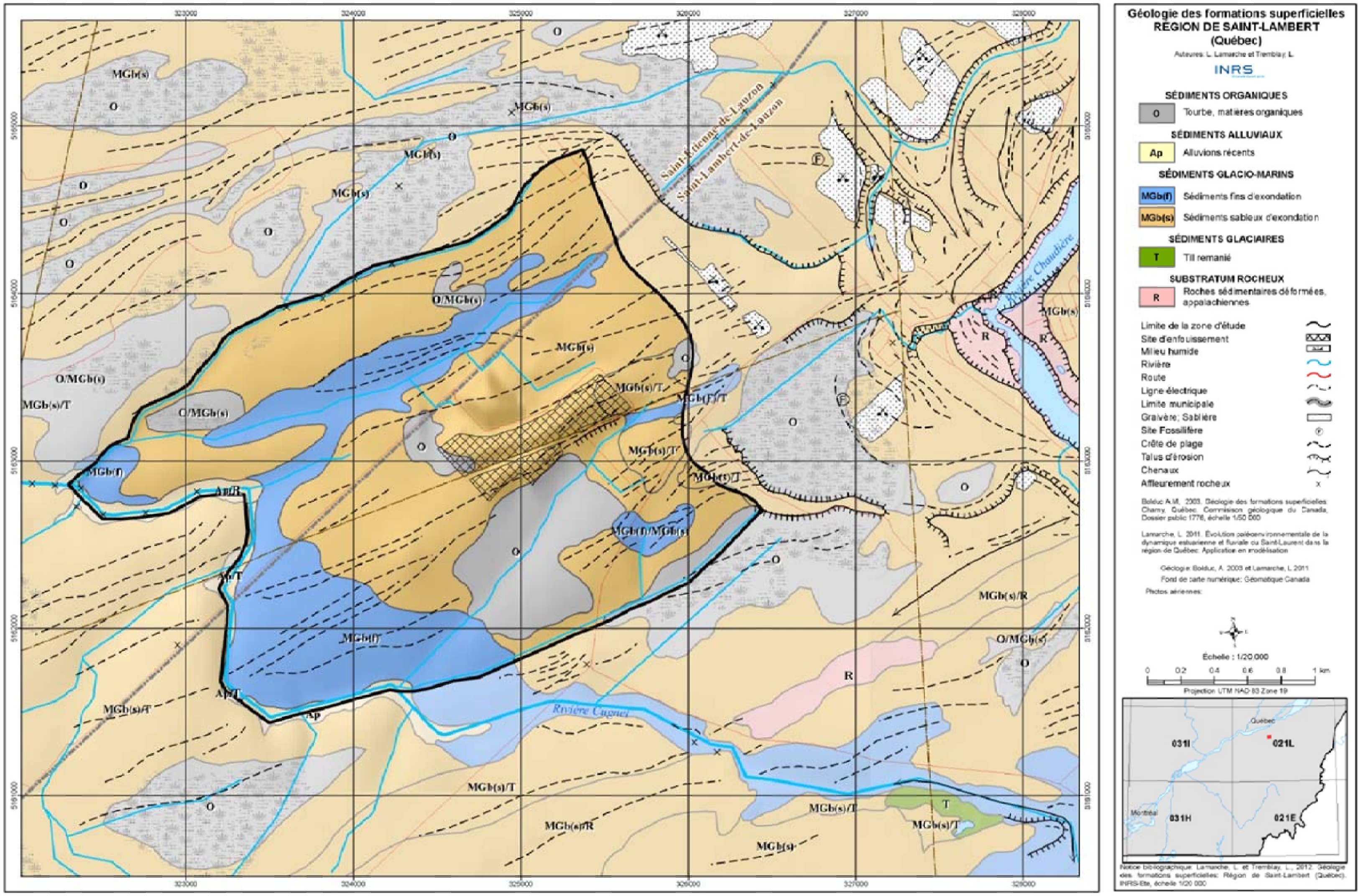


Figure 3.4 : Carte du Quaternaire refaite à l'échelle de la zone d'étude

très localement. Au site de St-Lambert, les conditions sont très peu favorables à la conservation des dépôts plus anciens. Toutefois, il faut envisager la possibilité de rencontrer des sédiments plus anciens qui ont été protégés de l'érosion lors de l'interprétation des différentes données de forages. Des forages récents ont démontré la présence de matériaux fluvioglaciaires très grossiers et bien délavés en profondeur et l'absence du till compact à certaines localisations dans la région d'étude.

La topographie de la région a été affectée par des mouvements tectoniques ainsi que par de forts agents d'érosion fluviale et littorale préglaciaire et interglaciaire. Les dépressions sont partiellement comblées par les sédiments glaciaires et marins datant du Wisconsinien. Les différents mouvements glaciaires peuvent être interprétés à partir de la composition lithologique du till. Les fragments composant les tills sont dérivés des roches des Appalaches, de la Plate-forme du Saint-Laurent et du Bouclier laurentien. Il existe une variation dans les proportions des fragments rocheux contenus dans les tills, ce qui implique une réorganisation des écoulements glaciaires majeurs sur le piémont appalachien. Quatre écoulements principaux auraient affecté la rive sud du Québec au Wisconsinien supérieur: la phase initiale de glaciation a engendré un écoulement vers le sud-ouest. Un autre vers le sud-est associé au maximum glaciaire. Un vers le nord-est exprime un courant glaciaire dans l'axe du fleuve St-Laurent. Enfin, un écoulement tardiglaciaire est dirigé vers le nord. Une étude effectuée sur les dépôts glaciaires exposés sur les berges de la rivière Beaurivage à St-Etienne-de-Lauzon a révélé deux unités de tills distinctes qui auraient été déposées par différents mouvements glaciaires au Wisconsinien (Ruest 2002).

La dernière glaciation a pris place entre 80 ka et 18 ka avec un maximum d'extension glaciaire à 21 ka. Jusqu'à présent, aucune évidence ne suggère la déglaciation du site de site de Saint-Lambert durant cette période. Cependant, dans le secteur plus au sud de la rivière Chaudière, la formation de Gayhurst montre un recul de la glace au Wisconsinien moyen (≈ 60 ka à 30 ka) (Shilts 1981). Les répercussions de ce réchauffement ne sont pas encore très bien établies dans la vallée du St-Laurent. Les dépôts associés à cette période sont un till de fond compact avec un écoulement vers le sud-sud-est qui peut avoir été remanié ultérieurement. La présence de sédiments fluvio-glaciaires à l'est du secteur suggère que les dépôts de même origine peuvent également se retrouver enfouis dans le secteur de St-Lambert. Une attention particulière devra être portée aux sédiments grossiers dans les descriptions de forage.

Tableau 3.2 : Stratigraphie et conductivité hydraulique mesurée sur le terrain et en laboratoire (Gauthier et al. 1993)

Horizon	**Lithologie**	**Élevation moyenne (m)**	**K_h (cm/s)**
Terre végétale		118-120	
Horizon 1	Sable fin à moyen, assez homogène	109-113	1×10^{-3}
Horizon 2	Sable fin silteux et argileux	103-112	1×10^{-4}
Horizon 3	Sable fin avec un peu de silt et des traces de graviers	100-105	3.4×10^{-3} à 2.9×10^{-4}
Horizon 4	Till: silt argileux d'origine glaciaireintercalé entre le niveau sabloneux et le socle rocheux. Absent dans les forages 92-F3, 92-F4, 92-F5, 92-F6, son épaisseur est de l'ordre de 3m sauf au forage 92-F2 avec plus de 7m.	97-102	1×10^{-7}
Roc fracturé	Shale gris avec veinules et microveinules carbonatées, épaisseur de quelques mètres et fracturation intense	96-101	$2.4\text{-}5.7 \times 10^{-5}$
Roc fracturé	Grès gris avec veinules et microveinules carbonatées dont les caractéristiques sont très variables. À titre exceptionnel, des bancs gréseux sont parfois rencontré au-dessus du niveau de shale fracturé	96-101	1.3×10^{-3}

La déglaciation qui a suivi a été ponctuée par une dynamique glacière particulière liée à la topographie du Québec méridional. Au Sud, le recul du front glaciaire se fait de façon régulière (environ 200 m/an) avec des périodes de stagnation associées à la formation des différentes moraines (Parent et Occhietti 1988). À la même époque, la formation d'une baie de vêlage dans l'estuaire a modifié l'écoulement glaciaire au nord du site. La formation d'un courant de glace au niveau du fleuve actuel provoque un rééquilibre de l'écoulement glaciaire à Saint-Lambert. Durant ce temps, l'écoulement de la glace est alors dévié vers le nord (Lasalle 1984; Shilt 1981; Gadd 1976). L'étape de la déglaciation complète jusqu'à l'invasion marine successive est très peu documentée puisque les évidences ont été enfouies sous les dépôts marins. Présentement, plusieurs interrogations persistent au sujet de la dynamique de la déglaciation finale du détroit de Québec et l'invasion de la Mer de Champlain. Toutefois, les échanges entre les masses d'eau de la Mer de Champlain et l'océan devaient être très importants dans le secteur provoquant une dynamique intense dans le secteur de St-Lambert et des environs.

Au maximum marin ($\approx$ 12,5 ka), le niveau de l'eau sur la rive sud a atteint une élévation d'environ 185 m (Parent et Occhietti 1999). À cette époque le site de St-Lambert était donc sous au plus 80 m d'eau. La proximité de l'embouchure de la rivière Chaudière de même que les forts courants et les vagues dans le détroit de Québec à ce moment évoquent un système deltaïque fortement perturbé. Le site de St-Lambert a été exondé dès le début de l'Holocène (10 ka).

Sur la propriété de la RIGDCC de 1 km^2, les propriétés hydrauliques historiques mesurées par des essais de perméabilité démontrent une grande hétérogénéité au niveau des matériaux (hydrofaciès) et peu de continuité horizontale au niveau des unités (Tableau 3.2). Le LES est situé près d'une ligne de partage des eaux souterraines.

3.6.2 Socle rocheux

La région à l'étude se situe dans la zone délimitée au sud par la faille du Foulon et au nord par la ligne de Logan (Figure 3.5). Le socle rocheux de la région d'étude est constitué de schiste argileux vert et rouge, de grès quartzeux et de conglomérat de la formation de Breakeyville du Groupe de Sillery (Castonguay et al. 2002). Cinq lithofaciès principaux ont été reconnus au sein de cette formation et peuvent se répèter à différents niveaux stratigraphiques (Figure 3.6). Les shales forment 65 % de l'ensemble, les grès 30 % et les microconglomérats 5% pour une épaisseur de formation estimée à 2400 m. Cette formation fait partie d'une nappe de charriage allochtone de l'époque du Cambrien. La Figure 3.5 décrite illustre et décrit en détail les structures et la minéralogie de ces roches.

Par définition, une nappe de charriage est un assemblage géologique caractérisé par une fabrique tectonique qui lui est propre et qui contraste avec celles qui servent à définir les nappes avoisinantes. La nappe de charriage représente donc le résultat de la remobilisation imposée de roches sédimentaires et volcaniques lors de la migration de l'arc volcanique développé au-dessus de la zone de subduction lors de la fermeture océanique. Les successions sont remobilisées le long de grandes failles de chevauchement dont les déplacements cumulatifs sont plurikilométriques. La Nappe de la Chaudière est constituée de roches sédimentaires d'âge Cambrien précoce à tardif qui furent déposées sur une marge continentale tectoniquement inactive (marge passive) peu de temps après l'ouverture de l'Océan Iapetus. Les successions rocheuses furent remobilisées près de 40 millions d'années plus tard, lors de la migration d'une série d'arcs volcaniques Ordoviciens lors du premier épisode orogénique majeur reconnu sur la marge de l'ancien continent Laurentia, soit l'orogénie Taconique de l'Ordovicien médian-tardif. La Nappe de la Chaudière, et les autres nappes formées de roches cambriennes à ordoviciennes inférieures présentes dans les Appalaches canadiennes, forment une zone tectonique identifiée comme la Zone de Humber (Williams 1976). Cette zone renferme diverses nappes dont l'origine des sédiments est associée à cet ancien craton Laurentia, les autres zones tectoniques de l'Orogénie Taconien sont formées de roches d'origine exotique et non- Laurentienne. Les nappes de chevauchement de la zone de Humber se sont mises en place successivement vers le nord-ouest, dans une séquence d'empilement qualifié "en séquence" (roches plus vieilles sur plus jeunes et failles tardives à niveaux structuraux inférieurs). Cependant, un empilement "hors séquence" est probable pour la Nappe de la Chaudière caractérisée par une position structurale au sommet de l'empilement tectonique, malgré tout, l'évolution tectonique de cette nappe demeure encore mal comprise (Castonguay et al. 2002).

La Nappe de la Chaudière a été cartographiée en détail par St-Julien (1995) qui a mis en carte divers éléments structuraux complexes incluant des plis d'amplitude et de longueur d'onde variable (de métriques à kilométriques) ainsi que des failles complexes dominées par des mouvements inverses (de chevauchement) avec des fractures tardives de nature extensionnelle. Le Groupe de Sillery de 5000 m d'épaisseur constitue la Nappe de la Chaudière et est divisé en trois formations, de la base au sommet, celle de Sainte-Foy, de Saint-Nicolas et de Breakeyville d'âge Cambrien précoce à tardif (Lavoie 2002). La surface de charriage à la base de la nappe est connue comme la Faille du Foulon et les données de terrain et de sismique réflexion suggèrent que la Nappe de la Chaudière est entièrement déracinée et forme donc une klippe tectonique au sommet de l'empilement tectonique des nappes de la région de Québec. En détail, la structure interne de la Nappe de la Chaudière est caractérisée par plusieurs failles de chevauchements à vergence nord-ouest et à pendage SE dans la partie nord du la klippe et à pendage faible généralement vers le NO dans la partie SE de la klippe. L'étude de la distribution des plis dans le Groupe de Sillery indique que les plis de type isoclinaux sont serrés avec des axes orientés grossièrement NE-SW et même renversés vers le nord-ouest. Ce système de plis est relativement ouvert dans la partie sud-est de la klippe à proximité de la région à l'étude. Ces plis se présentent

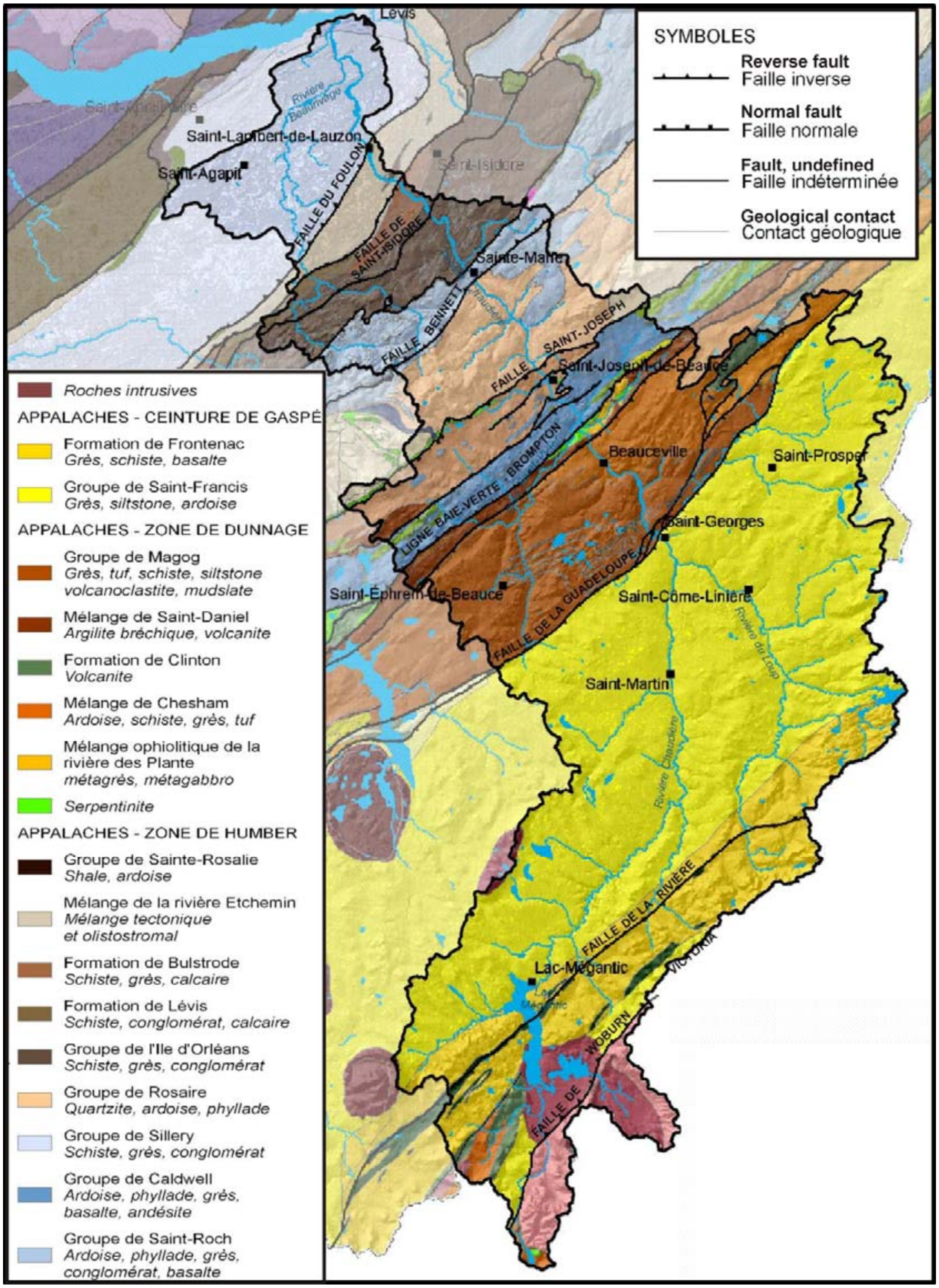
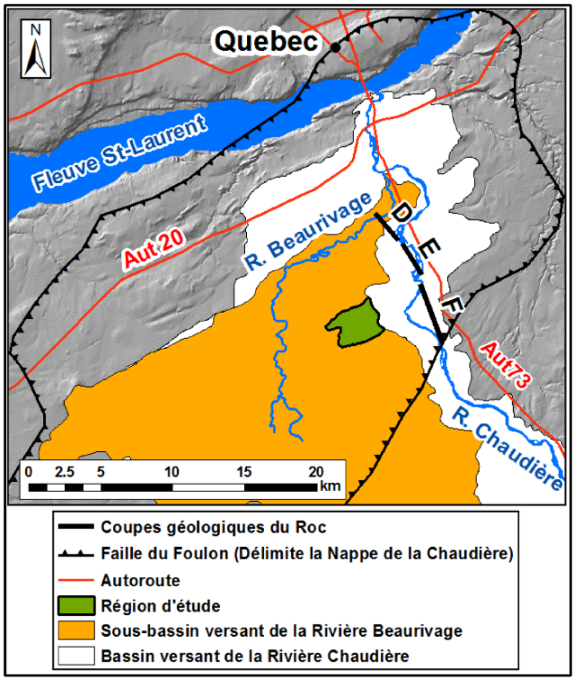


Figure 3.5 : Géologie simplifiée du socle rocheux pour le bassin versant de la rivière Chaudière, tirée de Benoit et al. (2012). La zone à l'étude est située dans la formation de Breakeyville (Figure 3.6) du Groupe de Sillery, au nord de la Faille du Foulon indiqué sur la carte



Description des unités lithostratigraphiques composant la formation de Breakeyville

6. Shales gris foncés avec quelques interlits de grès quartzeux et de silstones de 5 cm à 1 m d'épaisseur

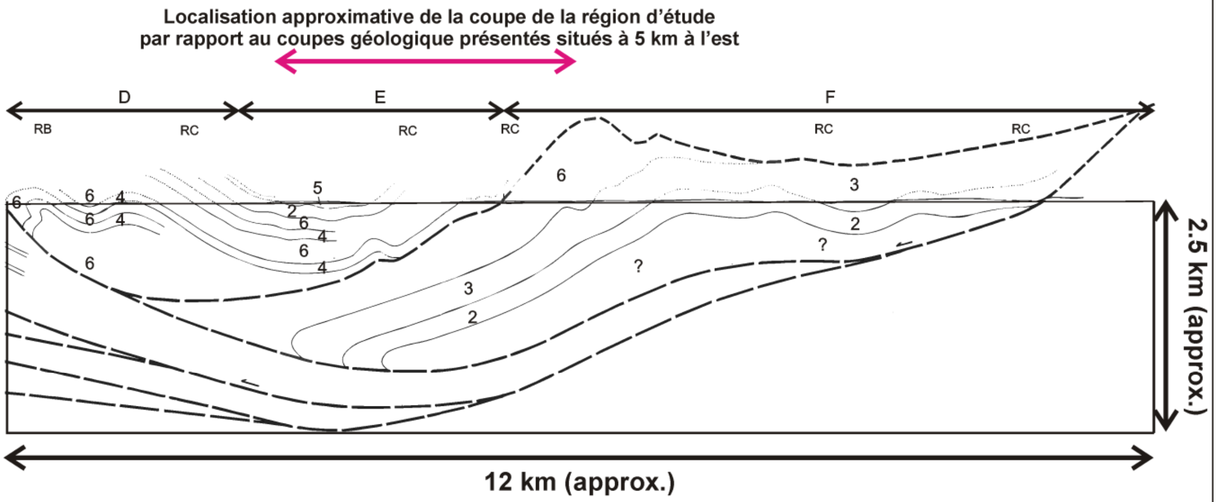
5.Interstratification de microconglomérats et de shales gris foncé

4.Grès quartzeux et d'orthoquartzites avec interlits de shales gris foncés

3. Mudstones et shales rouges avec quelques interlits de silstone et de grès quartzeux

2. Alternance de grès felspathiques et de mudstones et shales rouges, vert et gris

Les informations sur la géologie du socle rocheux et la coupe géologique sont tirés de l'étude de St-Julien 1995.



Description des grès et des shales de la formation de Breakeyville

Les grès sont composés de 10-40% de feldspath potassique gris ou blanc (orthose, perthite, microcline et albite en moindre proportion). Le quartz forme le reste de la portion gréseuse avec 1% de muscovite, biotite, zircon, apatite et minéraux opaque. Les grès sont granuloclassés et possèdent des microcomglomérats à leur base composé de 80% de quartz et 10% de feldspath. Les microconglomérats, composés majoritairement de particules de quartz, contiennent d'autres constituants majeurs tels le feldspath blanc, le calcaire et le chert. La matrice des microconglomérats est composé de shale ou de ciment carbonaté plus ou moins recristallisé et les fragments de roches sont composés de: grès, quartzite, chert, shale et localement un peu de glauconie.

Les mudstones et les shales rouges possèdent des lits de 30 cm à 5 m d'épaisseur sans stratification interne apparente. Ils sont composés de 3% de grain de quartz uniformément distribués et la matrice est constituée d'hématite, de chlorite et d'illite. Il y a aussi des flocons de mica parallèles à la stratigraphie. Les shales et les mudstones verts et gris possèdent des lits plus fins et ont des grains de quartz et de plagioclase et de glauconie de 0.04-0.006 mm. La matrice est formé de chlorite et d'illite. Les mudstones et les shales semblent tous fissiles parallèle à leur stratification.

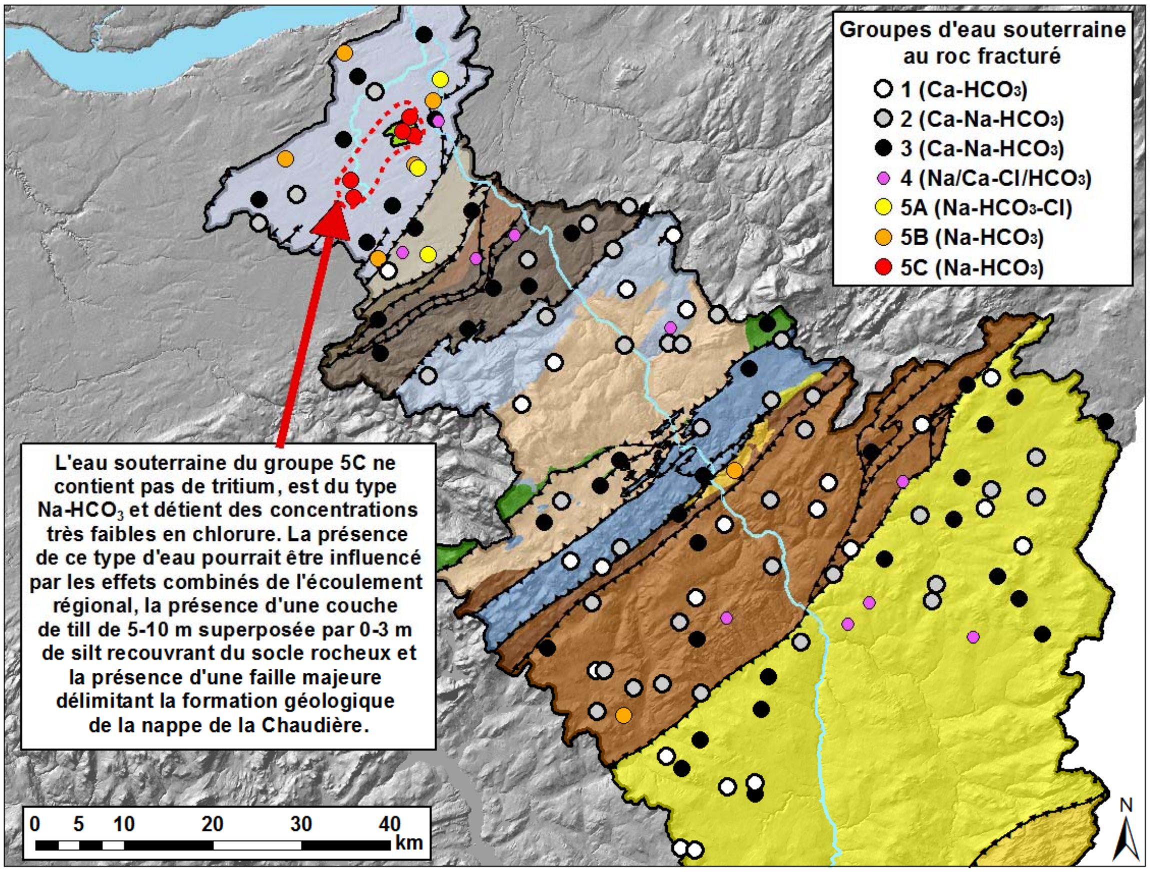
Figure 3.6 : Description de la géologie du socle rocheux

sous trois échelles de longueur d'onde : 5-25m, 250-750 m et dans la partie SE des plis de 1 à 5km. Un clivage ardoisier est localement développé dans les roches pélitiques de la région axiale des plis et les joints de tension sont communs dans les grès et certains de ces joints contiennent de l'anthraxolite, un témoin de la migration d'hydrocarbures dans ces fractures ouvertes. De plus, des fractures orientées à 30 degrés par rapport à la verticale ont été observées dans les échantillons de roc prélevés dans le sous-bassin versant étudié, ce qui démontre la plausibilité de la présence de faible pendage (Gauthier et al. 1993).

La roche sédimentaire shale-grès-microconglomérat a une porosité primaire négligeable. Le réseau de fractures et leur connectivité contrôlent la perméabilité de la roche (Benoit et al. 2009). Une valeur de conductivité hydraulique moyenne de 10^{-6} m/s montrant une faible décroissance jusqu'à une profondeur de 100 m a été mesurée pour la formation de Breakeyville (Brun Koné 2012). Un modèle numérique de l'écoulement a été développé pour le bassin versant de la rivière Chaudière et 127 échantillons d'eau souterraine ont été prélevés dans la roche sédimentaire pour corroborer avec le modèle d'écoulement (Figure 3.7) (Benoit et al. 2012). Le type d'eau retrouvé au centre de la formation de Breakeyville est Na-HCO_3, et il ne contient pas de tritium et a une concentration moyenne de chlorure sous 1 mg/L. La présence de ce type d'eau semble influencée par l'effet de l'écoulement régional contrôlé par les structures (failles et plis) de la roche ainsi que par la présence d'un couvert plus épais de l'aquitard. Basée sur des forages réalisés sur la propriété de la RIGDCC (Gauthier et al. 1993b), la roche sédimentaire se trouve à une élévation moyenne de 100 m et est connue pour affleurer à plusieurs localisations dans la région d'étude ou à s'élever à une élévation de 110 m. Les perméabilités au roc moyennes enregistrées sous la propriété de la RIGDCC à des profondeurs de 100 à 90 m sont de 10^{-7} m/s pour le shale noir et de 10^{-5} m/s pour le grès (Tableau 3.2). De plus, les études indiquent des fractures à 60 degrés et des veinules carbonatées (Gauthier et al. 1993b). Le suivi des niveaux d'eau à plusieurs niveaux démontre que la nappe du roc fracturé est possiblement reliée à celle de l'aquifère granulaire.

3.7 Données complémentaires, puits résidentiels, gravières, et tourbières

La rive ouest de la rivière Chaudière, entre les municipalités de Saint-Etienne-de-Lauzon et de Saint-Lambert-de-Lauzon, est caractérisée par la présence de nombreuses sablières et gravières. À St-Lambert, sur quelques kilomètres seulement, il y a des dépôts de sable grossier, de sable fin



L'eau souterraine du groupe 5C ne contient pas de tritium, est du type Na-HCO_3 et détient des concentrations très faibles en chlorure. La présence de ce type d'eau pourrait être influencé par les effets combinés de l'écoulement régional, la présence d'une couche de till de 5-10 m superposée par 0-3 m de silt recouvrant du socle rocheux et la présence d'une faille majeure délimitant la formation géologique de la nappe de la Chaudière.

	1	2	3	4A	5A	5B	5C
Ca (mg/L)	7.9-75	7.1-76	26-130	0.08-160	1.3-4.5	0.35-11	2.1-27
Na (mg/L)	1.2-4.4	4.6-45	3.8-56	7.9-290	62-180	33-91	26-97
Cl (mg/L)	0.26-8	0.3-9	10-93	94-591	31-52	1.5-8.5	0.37-0.8
SO_4 (mg/L)	2.6-35	2.6-67	6.6-81	20-95	1.8-19	11-38	3.4-24
pH	5.8-8.4	6.5-9.5	6.5-8	6.5-8.5	>9	>8	>8
^3H (TU)	12-14	9-10	9-10	9.3	3.4-5	4-5	0-0.8 (d.l.)
^{14}C (pmC)	80-95	72	72	48	7-25	40	26-30
Type	Ca-HCO_3	Ca-Na-HCO_3	Ca-Na-HCO_3-Cl	Na/Ca-Cl/HCO_3	Na-HCO_3-Cl	Na-HCO_3	Na-HCO_3
Context	recharge	intermediate	mixed-med Cl	mixed-high Cl	mixed	mixed	discharge

Only 1 to 3 analysis of ^3H and ^{14}C were available for each water group. d.l. stands for detection limit.

Figure 3.7 : Géochimie de l'eau souterraine de l'aquifère régional de roc fracturé. La légende de la géologie simplifiée du socle rocheux est indiquée à la Figure 3.5

plus ou moins silteux, de minces couches d'argile laissée en petits lacs fossiles sous les tourbières, des tills compacts constitués d'argile et de blocaux aux roches variées sur des épaisseurs considérables, mais sans étendue. Ce genre de dépôt est caractéristique de toute la région des Basses-terres qui bordent la rive sud du fleuve Saint-Laurent dans les environs de

Québec. Dans les terrains mal drainés, des matières organiques se sont accumulées pour former des dépôts de tourbes. On note la présence de plusieurs zones marécageuses et de tourbières dont certaines sont en exploitation (Laplante 1962). La Figure 3.8 illustre la localisation des tourbières et gravières actuellement en exploitation près de la zone à l'étude et les données de forages et de puits disponibles dans les environs à l'extérieur des limites de propriété du site de la RIGDCC. Les gravières et tourbières ont aussi été utiles pour faire l'observation de coupes géologiques.

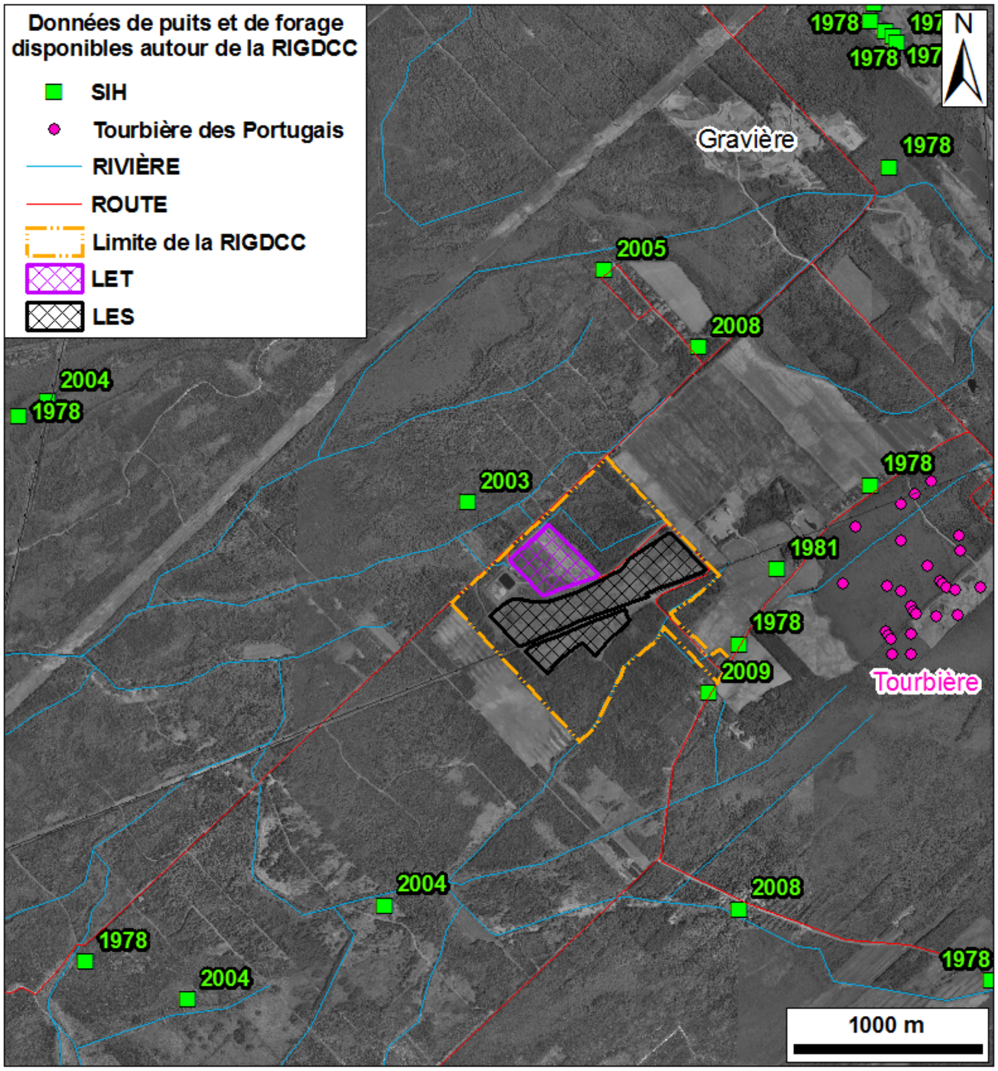


Figure 3.8 : Localisation de données de puits complémentaires
(puits résidentiels disponibles sur le SIH et puits localisés sur la propriété de la compagnie de Parterres Portugais Ltée.)

3.8 Contexte du site d'enfouissement sanitaire

La propriété de la Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière (RIGDCC) a une superficie d'environ 1 km^2. La RIGDCC exploite depuis novembre 1998 un lieu technique d'enfouissement (LET) conforme aux normes les plus strictes de gestion (MDDEP 2012a). Aménagé selon les règlements sur la mise en décharge et l'incinération des déchets, il est constitué d'un système d'imperméabilisation séparé en cellules et d'un système de collecte et de traitement des eaux de lixiviation. La Figure 3.9 illustre l'emplacement de l'ensemble des infrastructures sur la propriété et la répartition des zones boisées et les terres agricoles qui entourent le site. Au nord, le territoire est zoné "forestier" avec seulement des terres à bois.

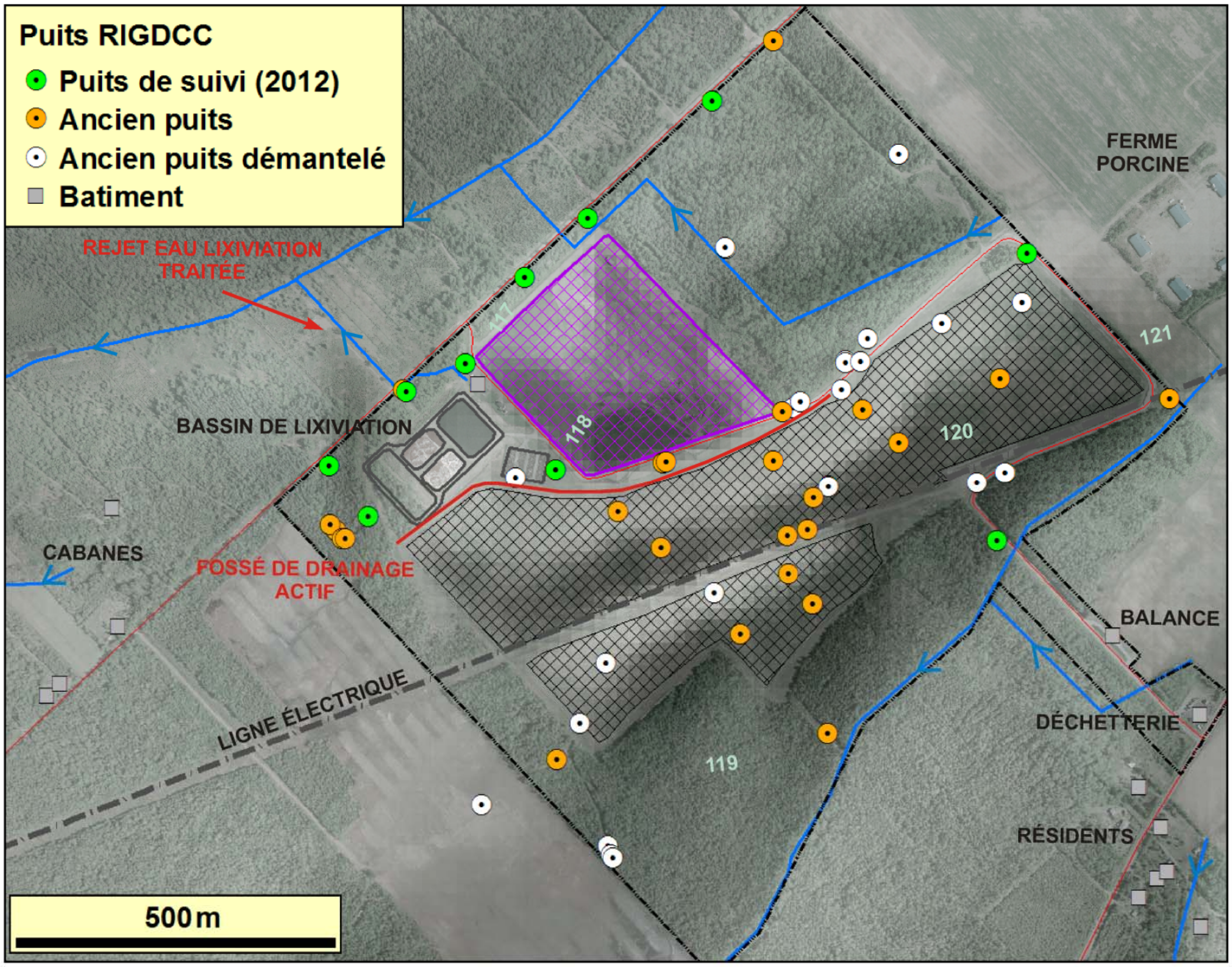


Figure 3.9 : Localisation des puits historiques disponibles au LES

L’ancienne piézométrie découlant d’une combinaison de plusieurs rapports de consultants montre l’élévation de la nappe sur la propriété du site (Simard 2007; Gauthier et al. 1994, 1993). Des résidents possèdent des puits d’approvisionnement en eau potable dans les aquifères au roc et granulaires à moins de 500 m au sud de l’ancien site d’enfouissement. Dans le site actuellement en opération, les eaux de lixiviation récupérée sont acheminées dans des bassins d’aération. Les eaux aérées sont ensuite acheminées dans des lits de tourbe puis rejetées dans l’environnement dans un cours d’eau adjacent (Figure 3.9). Les opérations sont saisonnaires. La performance du traitement dans les lits de tourbe est influencée par la température et par la charge des eaux du lixiviat. Au début du traitement, vers la mi-mai, un débit est de 50 m^3/j est acheminé vers les bassins de tourbes. Ensuite, au début du mois de juin, le débit est augmenté jusqu'à 140 m^3/j dans les 15 premiers jours et ensuite à 235 m^3/j à partir de la seconde moitié du mois de juin. Ce débit est maintenu jusqu'à la mi-septembre. Le débit est alors diminué, jusqu'à 50 m^3/j à la mi-novembre. Le système est en mesure de traiter un débit d’été pouvant aller jusqu'à 275 m^3/j.

Sur la même propriété, un ancien lieu d’enfouissement (LES) a été exploité de 1974 à 1997. L’ancien site d’enfouissement sanitaire couvre une surface d’environ 500 m par 1000 m. Le site repose sur une ancienne zone d’exploitation de granulat. En 24 ans, 900 000 tonnes de déchets de sources municipales, agricoles et industrielles, provenant de 15 municipalités différentes, ont été enfouies (RIGDCC 2013). Les détails de l’opération de l’ancien lieu et la proportion de la provenance des déchets sont inconnus. Les déchets auraient été placés directement à la surface du sol, après qu’une couche superficielle de terre et de sédiments d’épaisseur non précisée ait été enlevée de la zone d’enfouissement. Les déchets étaient ensuite compactés et recouverts d’une couche d’une mince couche de terre. Les déchets ont été placés directement sur les dépôts meubles composés de sable et gravier qui couvrent des silts et des argiles peu perméables recouvrant les tills et le roc. L’émission dulixiviat par l’ancien site d’enfouissement sanitaire de St-Lambert est uniquement gérée par atténuation naturelle, sans que l’efficacité de cette approche ait été vérifiée. Pour l’obtention de permis d’agrandissement et afin de respecter les règlements sur l’enfouissement des matières solides résiduelles de l’époque, plusieurs caractérisations géotechniques et hydrogéologiques ont été faites au cours des années d’opération du site. Avant que le site soit aménagé d’un recouvrement imperméable et de fossés, des eaux de lixiviation ont fait résurgence dans les ruisseaux avoisinants (Gauthier et al 1993; Dessau et al.

1993). La réhabilitation de l'ancien lieu d'enfouissement sanitaire de Saint-Lambert-de-Lauzon a débuté à l'été 1997. Les déchets localisés à l'extérieur des limites de propriété ont été déplacés et compactés. La surface couvrant une superficie 80 150 m^2 a été ensuite recouverte d'une membrane étanche composée de bentonite et recouverte de sable et de terre végétale. Les travaux ont été exécutés en novembre 1997 et l'ensemencement au cours du printemps 1998.

L'article 160 du Règlement sur l'enfouissement et l'incinération de matières résiduelles (REIMR) précise que les lieux d'élimination qui ont été fermés définitivement avant l'entrée en vigueur du REIMR en 2006 demeurent régis par les dispositions du Règlement sur les déchets solides (RDS) et par celles de leurs certificats d'autorisation ou de conformité. Comme le LES de Saint-Lambert a été définitivement fermé avant l'entrée en vigueur du REIMR, il demeure ainsi régi par le RDS et ses certificats d'autorisation ou de conformité. Le RDS ne prévoit pas de suivi des eaux souterraines avec des valeurs maximales à respecter. Il permettait que les eaux de lixiviation soient infiltrées dans le sol si les conditions hydrogéologiques particulières étaient remplies (art. 29 du RDS). Le LES de St-Lambert a reçu un certificat de conformité le 25 juillet 1996.

Préalablement à l'émission du ce certificat de conformité en 1996, l'étude de Gauthier et al. (1993) a tiré les conclusions suivantes :

- Les paramètres physicochimiques et les analyses chimiques préliminaires montrent que les eaux souterraines naturelles ont une signature géochimique particulière et présentent des conditions très variables dans l'espace. Les eaux de lixiviation ont aussi une signature spécifique.
- La contamination des piézomètres en bordure des cellules d'enfouissement s'accorde avec la direction de l'écoulement souterrain, perpendiculaire à l'axe des cellules d'enfouissement (Figure 3.10). La DCO, le fer et les phénols excèdent largement les normes du RDS (MDDEP 2012b). Les autres puits de suivi sur la propriété ne démontrent pas de concentration au-delà des normes prescrites. De l'eau de lixiviation se dilue dans les ruisseaux au nord et sud du LES.
- Lorsque les eaux sont hautes, la nappe phréatique s'élève dans un dépôt granulaire plus grossier, ce qui favorise une meilleure circulation des eaux souterraines et réduit le temps de migration. Il y avait un abaissement des concentrations de certains éléments chimiques à la fin de l'essai de pompage.

- Tous les échantillons d'eau potable recueillis dans les puits résidentiels situé à l'extérieur des limites de propriété de la RIGDCC montrent que les paramètres analysés respectent les normes réglementaires sur l'eau potable. Par contre, les échantillons des puits résidentiels installés dans l'aquifère granulaire montrent des concentrations de fer et manganèse supérieures aux normes pour l'eau potable et un pH sous la norme de 6.

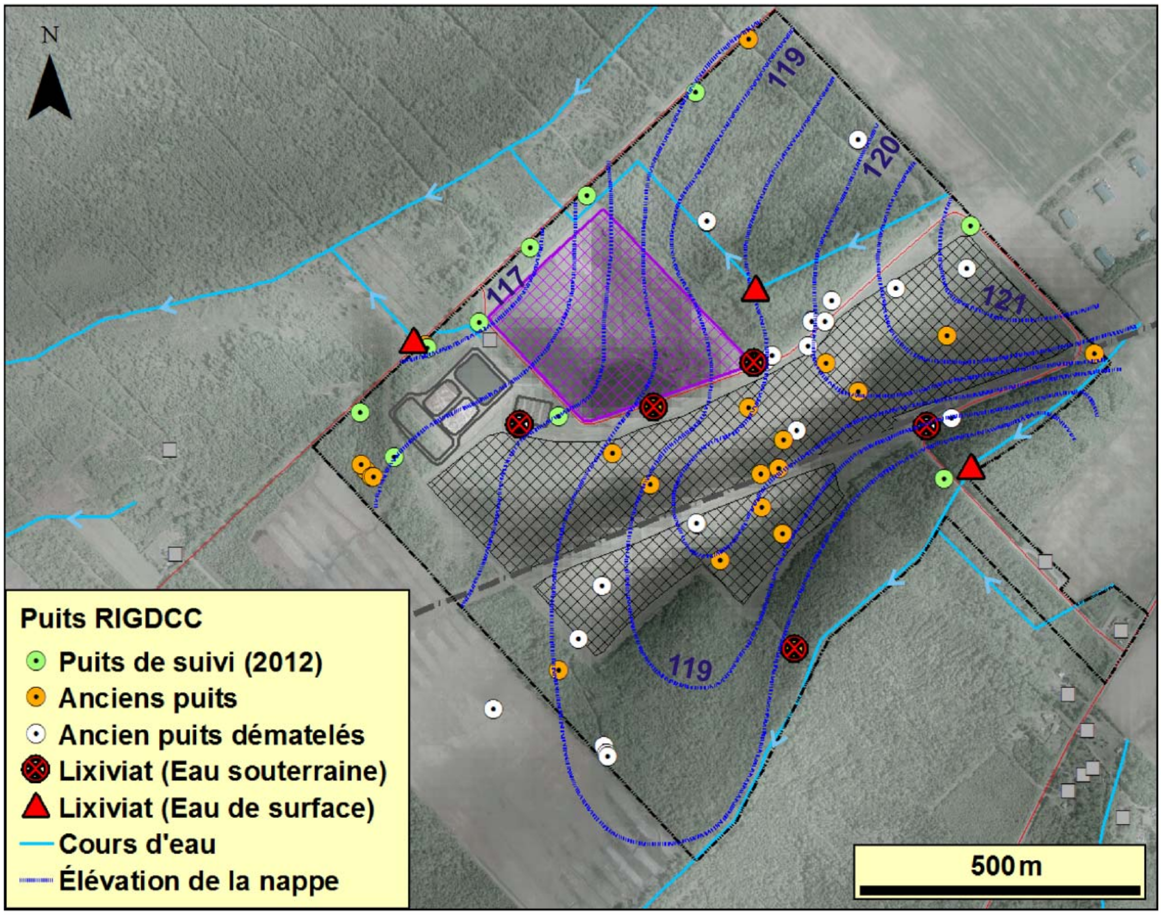


Figure 3.10 : Données historiques des dépassements géochimiques au LES
Pour l'eau souterraine, les X rouges indiquent où le DCO, le fer et les phénols dépassent largement les normes du RDS (MDDEP 2012b). Pour l'eau de surface, les triangles rouges indiquent où l'ammonium, la DBO_5, la DCO et les composée phénoliques dépassent les normes du RDS (MDDEP 2012b). La piézométrie à l'intérieur du site et à sa périphérie est dérivée de données de rapports de consultants. Partie nord (Simard 2007; Gauthier et al. 1994); partie sud (Gauthier et al. 1993).

Le niveau imperméable à la base des dépôts meubles s'est avéré n'être présent que localement sous le site d'enfouissement sanitaire et son épaisseur est nettement inférieure à celle proposée dans l'article 29 du Règlement sur les déchets solides (Q-2, r.14); sa discontinuité la discalifie comme couche imperméable d'ancrage. De plus, une continuité hydraulique entre les dépôts meubles et le shale sous-jacent est observée d'après le suivi des niveaux d'eau et les gradients verticaux.

Gauthier et al. (1993) recommandaient de compléter le réseau de surveillance de la qualité des eaux souterraines par l'installation de puits d'observation à l'extérieur de la propriété, dans le prolongement de la direction de l'écoulement, afin de quantifier les impacts actuels et éventuellement de dépister les impacts futurs. Il n'existe cependant pas de suivi à long terme dans les mêmes puits d'observation. Au cours des années d'exploitation, plusieurs puits ont été démantelés et installés, et certains rapports sont manquants pour les années 1994 et 1995. Actuellement, le réseau de puits d'observation installé pour satisfaire la règlementation concernant les LET est exploité. Pour la période de 2007 à 2012, le suivi des puits a porté sur seulement 7 paramètres, et ce, 3 fois par an.

Récapitulation des conditions reliées à l'ancien LES :

- Opération de 1974 à 1997, avec déchets placés directement sur des sables et graviers.
- Pas de traitement sauf l'atténuation naturelle sans suivi. Suivi environnemental non exigible, cas fermé avec certificat de conformité respectent les exigences de l'époque.
- Couche de sédiments imperméables sous le LES discontinue et épaisseur insuffisante pour assurer la protection de l'aquifère sous-jacent. La continuité des aquifères et aquitards mal comprise et connectivité possible entre l'aquifère rocheux et l'aquifère granulaire.
- Conditions d'écoulement dans l'aquifère granulaire inconnues à l'échelle du sous-bassin versant.
- Étendue du panache est inconnue, mais impact constaté dans les cours d'eau à proximité avant recouvrement final du LES.
- Aquifère granulaire composé de sable et de silt, généralement hétérogène, avec perméabilités généralement faibles, et naturellement réducteur.

CARACTÉRISATION INTÉGRÉE D’UN AQUIFÈRE GRANULAIRE POUR L’ÉVALUATION DES PROCESSUS GÉOCHIMIQUES INFLUENÇANT L’ATTÉNUATION NATURELLE D’UN PANACHE DE LIXIVIAT

Partie 2

Articles

Chapitre 4

Conceptual Model of Leachate Migration in a Granular Aquifer from the Integration of Detailed Multi-source Characterization Data

TITRE FRANÇAIS :

Modèle conceptuel de la migration d’un panache de lixiviat dans un aquifère granulaire grâce à l’intégration des données d’une caractérisation multidisciplinaire détaillée.

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CONTRIBUTIONS:

Laurie Tremblay: a planifié et coordonné l’acquisition des données de terrain, a contribué à l’acquisition, au traitement et à l’interprétation des données. Était personnellement responsable de l’acquisition des données superficielles (sols et cours d’eau) et géochimiques. Au niveau de l’article, a réalisé les figures et écrit l’article.

René Lefebvre : a coordonné le projet, encadré les étudiants impliqués et révisé l’article.

Daniel Paradis : a contribué à l’acquisition, au traitement et à l’interprétation des données. Était personnellement responsable de l’acquisition des données hydrauliques. Au niveau de l’article, a révisé la version préliminaire.

Erwan Gloaguen : a supervisé l’acquisition et l’interprétation des levés géophysiques. A contribué aux orientations du projet. A révisé la version préliminaire de l’article.

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MODIFICATIONS EXIGÉES PAR LE COMITÉ DE LECTURE DE LA REVUE :

Les deux évaluateurs de l'article et l'éditeur associé de la revue ont demandé que des corrections importantes soient faites au manuscrit initial, particulièrement au niveau des points suivants : a) expliquer clairement les aspects novateurs de l'approche proposée, b) faire le lien entre les travaux décrits dans l'article avec les études existantes dans ce domaine, et c) donner une perspective claire à l'article. Des demandes ont aussi été faites de réviser la structure de l'article dans son ensemble et de mettre davantage l'accent sur l'intégration des données plutôt que de détailler les étapes de la caractérisation. L'éditeur de la revue a accepté cette version révisée du manuscrit en demandant seulement d'en réduire la taille et de considérer l'enlèvement de deux figures. Les deux figures suggérées ont été retirées et le manuscrit final a été réduit de 10 pages par rapport à la version révisée.

RÉSUMÉ

Les modèles numériques possédant les deux caractéristiques suivantes sont requis pour comprendre le devenir et la migration des panaches de contamination: 1) ces modèles doivent couvrir un territoire incluant les zones sources de contamination ainsi que les milieux récepteurs et 2) ils doivent être basés sur des modèles conceptuels représentatifs qui tiennent compte de l'hétérogénéité des aquifères. Malheureusement, les méthodes de caractérisation actuelles des aquifères ne fournissent pas les données requises pour développer ce type de modèle. Les objectifs de cette étude étaient de développer des procédures de caractérisation et d'intégration des données hydrogéologiques: 1) adapté à la définition régionale de l'hétérogénéité des aquifères (excédant quelques km^2) et 2) fournissant les données complémentaires requises pour supporter le développement de modèles numériques détaillés de l'écoulement de l'eau souterraine et du transport de masse. Cette étude comprend la caractérisation de terrain multidisciplinaire d'un aquifère granulaire peu profond dans un sous-basin versant de 12 km^2 à l’intérieur duquel ce trouve un ancien site d’enfouissement sanitaire qui émet un panache de lixiviat géré par atténuation naturelle. Utilisant des méthodes éprouvées, la séquence de caractérisation a été conçue pour optimiser chacune des méthodes de terrain en termes de localisation, d’échelle d’acquisition, de densité et de qualité. L’emphase a été mise sur l’acquisition de données hydrogéophysiques indirectes détaillées qui ont été intégrées avec des données hydrauliques et géochimiques de haute résolution. Le programme de travaux a fourni un important jeu de données de sources variées bien distribuées spatialement dans la région d’étude. L’intégration de ces données, grâce à la géostatistique, a permis de définir la géométrie de l’aquifère en 3D, l’hétérogénéité des matériaux granulaires de l’aquifère, les voies d’écoulement de l’eau souterraine, les contextes de recharge et d’émergence et l’étendue du panache de lixiviat. Cet article met l’emphase sur les premières étapes d’intégration qualitatives et géostatistiques des procédures développées, ce qui permet le développement d’un modèle conceptuel hydrogéologique détaillée. Cet étape initiale est un prérequis à d’autres étapes d’intégration, telles que la prédiction d’hydrofacies et de valeurs de conductivité hydraulique (*K*), des simulations géostatistiques de *K* et l’étude des processus géochimiques et la modélisation numérique de l’écoulement et du transport de masse. Ce niveau détaillé de caractérisation n’aurait pas été atteint dans le même laps de temps et avec les mêmes ressources en utilisant des méthodes conventionnelles. Cette méthodologie a été efficace en termes de coûts et de temps et elle peut être adaptée à tout type d’environnement hydrogéologique et d’échelle de région d’étude.

MOTS-CLÉS : aquifère granulaire, caractérisation, intégration de données, hétérogénéité, modèle conceptuel.

ORIGINALITÉ ET PERTINENCE:

Plusieurs études ont abordé la question de l’hétérogénéité des aquifères ou avaient pour but d’offrir une compréhension plus approfondie des sites contaminés, ce qui démontre que ces problématiques sont importantes et requièrent un développement scientifique. Selon les auteurs, les approches décrites dans l’article ont les aspects novateurs et originaux suivants par rapport aux études antérieures portant sur ce sujet :

Échelle d'étude

Les études sur l'hétérogénéité, avec les approches hydrauliques ou hydrogéophysiques, ont mis l'emphase sur l'étude de sites locaux de dimensions restreintes (moins de 50 m x 50 m). L'étude documentée dans cet article couvre un territoire de quelques km^2 dont la taille permet de considérer la zone source de contamination (l'ancien site d'enfouissement) et les milieux récepteurs (principalement les cours d'eau). Cette échelle d'investigation est aussi requise pour la gestion des sites contaminés et pour offrir la possibilité de développer les modèles numériques nécessaires pour supporter ce type de gestion.

Les données acquises

Plusieurs études sur l'hétérogénéité des aquifères sont basées sur des données synthétiques, souvent seulement en coupe 2D. Cette étude est l'une des rares tentatives de produire les données requises pour un site réel afin de représenter l'hétérogénéité à l'échelle d'un sous-bassin versant en 3D. Cet objectif de couvrir un véritable territoire de la taille de la région d'étude a eu d'importantes implications par rapport à l'approche séquentielle suivie pour l'acquisition des données requises pour les fins de cette étude.

L'utilisation de plusieurs méthodes géophysiques

Plusieurs études mettent l'emphase sur l'application d'une seule méthode. Même si certaines études font l'utilisation de multiples méthodes, l'approche décrite dans cet article est originale par la séquence des méthodes utilisées et le rôle assigné à chacune des méthodes, et ce dès le début de l'acquisition des données de terrain.

Combinaison originale des mesures directes et indirectes.

Pour produire des données directes qui puissent être comparées aux données hydrogéophysiques indirectes sur la gamme de matériaux et de perméabilités retrouvées dans une zone d'étude hétérogène, des puits crépinés sur toute l'épaisseur de l'aquifère ont été localisés à partir des levés géoradars et installés par enfoncement sur la base des sondages CPT/SMR donnant les informations sur la séquence de matériaux en temps réel sur le terrain. De plus, plusieurs méthodes hydrauliques ont été testées et même développées afin de produire les données hydrauliques permettant l'établissement de relations entre les données géophysiques indirectes et les mesures hydrauliques.

Intégration originale des données

Les méthodes d'intégration emploient une variété d'approches, de façon qualitative à quantitative, notamment les coupes géologiques, l'intégration géostatistique par krigeage, et la conversion des valeurs de résistivité des sondages par enfoncement en estimés de matière dissoute dans l'eau souterraine. Les méthodes d'intégration utilisées dans l'article permettent de développer un modèle conceptuel du système d'écoulement, en définissant les conditions hydrogéologiques, en identifiant les zones de recharge et d'émergence et en délimitant l'étendue du panache de lixiviat. Ces éléments sont essentiels pour la compréhension du système d'écoulement et le développement de modèles numériques représentatifs.

Implications scientifiques

Cette étude aborde des problématiques importantes en hydrogéologie par rapport aux développements de modèles conceptuels représentatifs pour guider des études plus spécifiques sur la recharge, la géochimie, la vulnérabilité, et la modélisation numérique.

Pertinence socio-économique

Ce projet démontre que l'approche de caractérisation utilisée 1) permet de définir efficacement et de façon détaillée les conditions rencontrées dans un aquifère, 2) utilise des méthodes éprouvées déjà disponibles commercialement et 3) peut être réalisée à des coûts moindres que les approches conventionnelles. Ces nouvelles connaissances ont permis de transférer des techniques de caractérisation hydrogéologique novatrices aux partenaires privés du projet. Ces travaux devraient aussi faciliter la gestion environnementale du site par la RIGDCC et la compréhension du site par tous les citoyens concernés, grâce notamment aux modèles conceptuels qui synthétisent la connaissance du système aquifère et illustrent les conditions de façon concrète.

ABSTRACT

Numerical models encompassing source zones and receptors, based on a representative conceptual model and accounting for aquifer heterogeneity are needed to understand contaminant migration and fate. However, aquifer characterizations seldom provide the data needed to develop such models. The objective of the study was to develop a workflow for field characterization and data integration 1) adapted to the definition of subwatershed-scale aquifer heterogeneity (over 10 km^2) and 2) adequately supporting mass transport model development. The study involved the field investigation of a shallow granular aquifer in a 12 km^2 sub-watershed in which a decommissioned landfill is emitting a leachate plume managed by natural attenuation. Using proven field methods, the characterization sequence was designed to optimize each method in terms of location, scale of acquisition, density and quality. The emphasis was put on the acquisition of detailed indirect geophysical data that were integrated with direct hydraulic and geochemical data. This paper focuses on the first qualitative and geostatistical data integration steps of the workflow leading to the development of a hydrogeological conceptual model. This is a prerequisite for further integration steps: prediction of hydrofacies and hydraulic conductivity (*K*), geostatistical simulations of *K*, studies of geochemical processes and numerical modeling.

KEYWORDS: granular aquifer, characterization, data integration, heterogeneity, conceptual model

4.1 Introduction

Historically, abandoned gravel pits often served as a repository for municipal and industrial waste (Roy 1994). Nowadays, many of these decommissioned and unlined landfills remain solely managed by monitored natural attenuation (MNA), relying on processes such as dilution, sorption, and microbial degradation to reduce the mass of contaminants to less harmful forms and concentrations (Christensen et al. 2001, 1994). Since not all contaminants can be remediated by natural attenuation or their concentration decreased to acceptable levels, contaminant mass emissions from such unlined waste disposal sites can pose a threat to groundwater quality in shallow aquifers for decades to centuries (Milosevic et al. 2012; Bjerg et al. 2011, 2003; Cozzarelli et al. 2011; Park et al. 2008; Kjeldsen et al. 2002; Christensen et al. 2001). In the last decade, numerous scientific advances have been made regarding the occurrences and rates of several attenuation processes within plumes or at their fringe (physical, chemical and microbial), which control the fate and transport of contaminants in aquifers. These studies demonstrated the complexity and the variability of the reactions occurring as plumes migrate through different types of geological conditions (Robertson et al. 2012; Bjerg et al. 2011, 2003; Cozzarelli et al., 2011; Sudicky and Illman 2011; Sudicky et al. 2010; Gutierrez-Neri et al. 2009; Lorah et al., 2009; USEPA 2008, 2007; van Breukelen et al. 2003; Kjeldsen et al. 2002; Christensen et al. 2001, 1994). However, despite advances on the scientific understanding of mechanisms and rates of contaminant attenuation in leachate plumes, a debate still exists within the remediation community regarding what constitutes an adequate assessment of a contaminated site. Such adequate assessment should provide the answers needed, with an acceptable level of uncertainty, to define the type of remediation required (USEPA 2007; Rapti-Caputo et al. 2006a; Wilson et al. 2005; Vangelas et al. 2005; Norris et al. 2004; Daugherty et al. 2004; Rittman et al. 2004; Crumbling et al. 2003). Still, among the various national and state (or provincial) environmental agencies throughout the world, there is no unanimity on parameters to be monitored or on maximum concentrations acceptable in groundwater (MDDEP 2012; Butt et al. 2008; USEPA 2007; Rapti-Caputo et al. 2006b; Lee et al. 2003; EA 2003, 1999; EC 2000, 1999).

Effective management and protection of water resources is a major multifaceted societal challenge. The responsible management of groundwater requires knowledge of the shallow subsurface, groundwater occurrence, flow dynamics, contamination from land surface, solute transport and the representation of these processes with numerical simulators (Nordstrom 2012; Schmelzbach et al. 2011; Konikow 2010; Janković et al. 2009; Maier and Grathwohl 2006; Zhu and Anderson 2002). Nowadays, it is recognized that much of the spatial variability in contaminant concentrations results from geological heterogeneity and, consequently, three dimensional characterization of the groundwater flow field is required to understand the transport and, ultimately, the fate of contaminants (USEPA 2008, 2007; de Marsily et al. 2005). Many authors have concluded that heterogeneity not only controls the physical transport of contaminants by dispersion and advection but also affects chemical reactions, bacterial attachment rates, and remediation effectiveness (Anderson and McCray 2011; Allen-King et al. 2010). In addition to the development of a detailed understanding about the hydrogeological system needed to delineate transport pathways, groundwater recharge and discharge conditions also have to be defined, since they exert major controls on flow and groundwater chemistry (Stephens 2009; Illman et al. 2008; USEPA 2008, 2007; Bredehoeft 2007; Healy and Cook 2002; Kim et al. 2000; Arnold and Allen 1999). There is thus a need in contaminant hydrogeology for the elaboration of systematic approaches leading to the development of representative conceptual models, based on a wide range of characterization data, acquired at a scale encompassing source zones and receptors, as well as data integration schemes maximizing the information derived from indirect and direct data. This paper aims to make a contribution to such developments, which would support contaminated site management.

Such developments are challenging since they address two elusive, and related, research topics in hydrogeology: 1) the development of representative hydrogeological conceptual models, and 2) the quantitative estimation of uncertainty (Bredehoeft and Konikow 2012; Sousa et al. 2012; Anderson and McCray 2011; Frind et al. 2006). A hydrogeological conceptual model can be defined as a qualitative, often graphical, description of a groundwater system, which is needed as a basis for the development of a numerical model. Conceptual models describe the aquifer geometry, lithology, hydrogeological parameters, initial and boundary conditions or hydrobiochemical reactions (Zuber et al. 2011). Although seldom explicitly acknowledged, uncertainty in hydrogeological predictions is in large part related to an incomplete conceptual model, due to either data scarcity, undefined heterogeneity of the subsurface geological materials, and uncertainties on boundary conditions such as recharge (Sousa et al. 2012). Scientists from diverse hydrogeological research specialties (recharge quantification, groundwater quality, wellhead protection areas, modeling of solute transport, etc.) advocate for the development of a conceptual model based on actual field data prior to efforts aiming to resolve issues related to groundwater management and protection (Nordstrom 2012; Sousa et al. 2012; Sudicky and Illman 2011; Clément 2010; Healy 2010, Crumbling et al. 2003). Regrettably, fewer resources and less efforts is given to the collection of necessary field data compared to those dedicated to the improvement of numerical modeling codes (Nordstrom 2012; Sousa et al. 2012; Yang et al. 2004). In recent decades, continuous development of numerical simulators has allowed the modeling of complex flow systems. However, quality assurance and hypothesis validation become more challenging as models increase in complexity (Nordstrom 2012). Also, the numerical model itself includes hydrogeological uncertainty, as well as the issue of calibration non-uniqueness (Sousa et al. 2012). Furthermore, conventional hydrogeological characterization methods generally do not provide the data necessary to represent complex aquifers in a model (Refsgaard and Henriksen 2004), whereas hydrogeophysical characterization has often focused on detailed assessment of small areas (Ruggeri et al. 2013). While it is recognized that aquifer heterogeneity exerts strong control on mass transfer and dispersion (Sudicky and Illman 2011; Frei et al. 2009; de Marsily et al. 2005; Fogg et al. 1998; Goode 1996), it is still not characterized at the larger scale. Although in numerous studies based on synthetic aquifers it is possible to assess the validity of the representation of heterogeneity, for actual field sites such representation relies on limited data and is usually based on multiple geostatistical realizations that cannot be fully verified (Blouin et al. 2013; Engdahl and Weissmann 2010; Illman et al. 2010; Dafflon et al. 2009; Mariethoz et al. 2009; Fleckenstein and Fogg 2008; Carle et al. 2006; Tronicke and Hollinger 2005). This issue has more than theoretical importance, since inadequately heeding aquifer heterogeneity is a common cause of remediation failure (Sudicky and Illman 2011; Maxwell et al. 2008; de Marsily et al. 2005).

Many environmental agencies and councils have identified the pressing need to develop better tools and approaches to characterize, monitor, and investigate hydrogeological systems and processes in the shallow subsurface at relevant spatial scales and in a minimally invasive manner (USEPA 2008, 2007; MDDEP 2008; Crumbling et al. 2003). In the petroleum and mining industries, as well as in recent groundwater studies, continuous indirect data often obtained from geophysical surveys have proved very efficient to adequately map the geometry of major lithological units and constrain the spatial modeling of subsurface heterogeneity based on sparse direct measurements (Gazoty et al. 2012; Schmelzbach et al. 2011; Dogan et al. 2011; Ouellon et al. 2008; Mosey 2002; Sandberg et al. 2002; Goovaerts 1997). Rubin and Hubbard (2005) have also described the potential for the emergent field of hydrogeophysics to enhance the characterization of aquifer heterogeneity. Many geophysical methods have the potential to assess spatial variability of anisotropy, constrain hydraulic parameters and improve groundwater-flow model predictions (Sandberg et al. 2002). In contrast to traditional field investigation, non-invasive hydrogeophysical methods are less costly, more efficient, and can provide higher spatial resolution sampling (Perri et al. 2012; Dogan 2011). Since each single method has advantages and limitations (Turesson 2006; Acworth and Jorstad 2006), the combination of multiple methods increases the level of confidence in the hydrogeophysical imaging of aquifers. In addition, direct measurements of hydraulic properties are needed to define hydrofacies and the relationships between indirect hydrogeophysical data and hydraulic properties, as values of hydraulic properties are needed to develop groundwater flow models (Paradis et al. 2011a; Ouellon et al. 2008). Hydraulic tomography can be used as part of detailed field characterization studies to provide insights on aquifer heterogeneity, or to establish relationships with indirect indicators of hydraulic properties, such as geophysical surveys (Brauchler et al. 2012; Caers 2005; Rubin and Hubbard 2005). However, hydraulic tomography, like any other hydraulic tests, has not, and cannot by itself, be used to provide the data needed for aquifer characterization at the regional scale. Besides direct hydraulic data, geochemical data can provide key indications of flow paths and help understand groundwater evolution and contaminant attenuation (Glynn and Plummer 2005; Edmunds 2009). Geochemical tracers can provide independent data, which can complement hydraulic data, to match the historical data of a site and fully constrain numerical models to obtain a more realistic representation of groundwater flow with reliable predictive capabilities (Murphy et al. 2011; Sanford 2011; Zuber et al. 2011; Reilly et al. 1994).

This study proposes a workflow for field characterization and data integration, adapted to the definition of aquifer heterogeneity at the subwatershed scale, and providing relevant information for contaminant studies and solute transport modeling. Very few studies have attempted to define heterogeneity at the subwatershed scale (5 to 50 km^2) (e.g. Ouellon et al. 2008). In the perspective of defining aquifer heterogeneity for contamination risk assessment, other studies have investigated areas of 500 m^2 or less (Berg and Illman 2013; Gazoty et al. 2012; Sudicky and Illman 2011, 2010; Dogan et al. 2011; Engdahl et al. 2010; Nyáry 2010; Illman et al. 2007). For the study reported in this paper, while utilizing proven field methods, the characterization sequence was designed to optimize each sampling technique in terms of location, scale of acquisition, density and quality. This paper thus makes a contribution in defining a sequence for fieldwork and a logical combination of multiple tools to optimize each survey acquisition, quality and relevance as a field campaign is carried out, insuring data set completeness. The multi-disciplinary characterization was designed to provide the data required for: 1) the development of a representative conceptual model of the aquifer system (aquifer-aquitard units and geometry, stratigraphy and heterogeneity, hydraulic properties, flow directions, boundaries including links between groundwater and surface water, recharge and discharge zones and natural geochemical conditions); 2) the understanding of the migration and fate of contaminants (source zones, geochemical conditions in the plume, geochemical processes and aquifer heterogeneities such as hydrofacies, hydraulic conductivities and natural geochemistry affecting plume migration, and links between plume and receptors); 3) the development and verification of numerical models (reduction of uncertainty by reproducing the conditions with numerical simulations, calibration with hydraulic heads, streamflow, groundwater age and plume tracers); 4) the support of environmental risk assessment (present and future concentrations at receptor) and; 5) the understanding necessary to manage contaminated sites-monitored natural attenuation (MNA), remediation, control. This study thus shows how to integrate key components of a hydrogeological system at the subwatershed scale within a conceptual model from detailed and varied data provided by a single characterization program which also aims to be efficient, cost effective and adaptable to other sites. Such a conceptual model is a prerequisite for the following quantitative data integration steps that are only briefly described in the paper: development of a learning machine workflow to recognize hydrofacies and predict *K* (Paradis et al. 2011b), multiple points geostatistical simulations to define a heterogeneous *K* field (Brunet et al. 2012), and studies of geochemical processes and numerical modeling to assess natural attenuation (Tremblay et al. 2012).

4.2 Study area

The study area covers a 12 km^2 sub-watershed encompassing a former landfill located in the municipality of Saint-Lambert-de-Lauzon, 35 km south of Quebec City, Canada (Figure 4.1). The study area is easily accessible by primary, secondary and forestry roads. The eastern limit of the zone investigated represents the groundwater and surface-water divide line between the Beaurivage River subwatershed and the Chaudière River watershed. Elsewhere, the boundaries of the investigated area were selected on the basis of the physiology of the creeks and rivers located in the subwatershed. The landfill is surrounded by forested land, peatland, agricultural fields and a few operational gravel pits. In this relatively flat region, the water table is very shallow, as it is found between 0.2 to 2 m below ground surface, and the surface soils are mostly composed of poorly drained podzols of marine origin (Laplante 1962). Due to relatively flat topography and imperfectly drained soil type, an important forestry and agricultural drainage network is in place and, in combination with natural streams, local hydrology strongly influences groundwater flow (Tremblay et al. 2008).

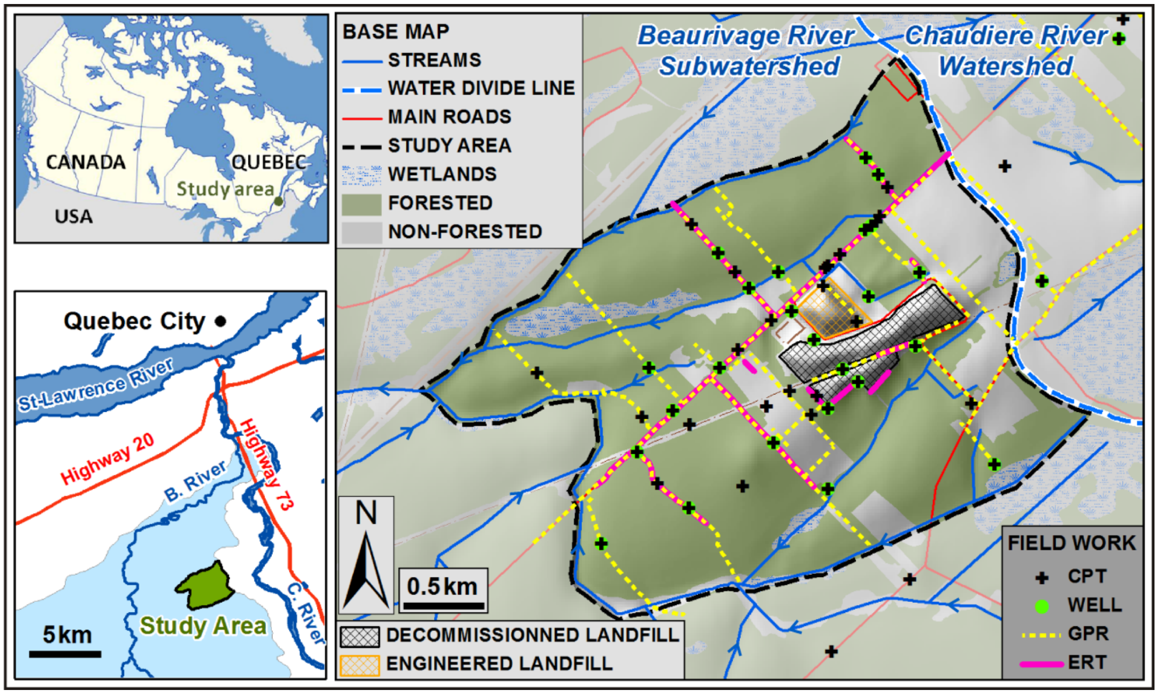

Figure 4.1 : Study area location and spatial distribution of characterization work
On the map at the bottom left: B. River and C. River respectively stand for Beaurivage River and Chaudiere River. On the same map, the Beaurivage River subwatershed is shaded in blue. The map on the right shows physiographic characteristics and land use of the study area

The “Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière” (RIGDCC) is the public organization currently operating an engineered landfill meeting current standards of waste management on 18% of its 1 km^2 property (Figure 4.1). However, 40% of the property is occupied by a decommissioned landfill. This former unlined landfill was in operation for 24 years and during that time 9 million tonnes of waste from different sources were buried directly over permeable sands. The former site was closed, reshaped and its surface was capped with an impermeable barrier in 1997 (RIGDCC 2013). Prior to site closure, landfill leachate was detected in the surrounding creeks and rivers (Gauthier 1993). Groundwater and surface water affected by the leachate plume showed mostly elevated concentrations of organic matter and inorganic macrocomponents such as chloride, ammonium, calcium, potassium, sodium, iron, manganese, and hydrogen carbonate. Aromatic hydrocarbons, phenols and heavy metals were detected close to detection limits only in observation wells located within 25 m of the former waste site. Currently, groundwater chemistry shows no concentrations of dissolved ions exceeding provincial standards (MDDEP 2012) in wells located outside the limits of the 1 km^2 landfill property. The decommissioned landfill is solely managed by natural attenuation. No studies have yet validated the effectiveness of this method of environmental management or attempted to predict the actual extent and fate of the plume.

The aquifer is under semi-confined conditions and is composed of 10 to 20 m of highly heterogeneous sand interbedded with silt and clayey silt lenses. The aquifer overlies discontinuous layers of glaciomarine silt and till forming the relatively impermeable base of the aquifer. The till deposited by the last glaciation overlies the bedrock aquifer composed of fractured shale and sandstone. The Quaternary fluvial and deltaic sediments making up the aquifer were deposited after the last glaciation (Bolduc, 2003). The unconsolidated material originated from sea-level changes during the regression of the Champlain Sea at the former Chaudière River outlet. At that time, the system was controlled by sea tides and current, followed by fluvial erosion and organic sedimentation. This littoral depositional environment is responsible for the high heterogeneity of the aquifer material (Parent and Occhietti 1999) as well as naturally occurring anoxic and oxidizing zones that influence the natural geochemical composition of groundwater (Tremblay et al. 2008). Added to this complex environment, is the presence of a leachate plume which undergoes important geochemical transformations along its path (Tremblay et al. 2008).

4.3 Characterization and methods

4.3.1 Characterization approach

The methodological workflow was designed to ensure the complete and proper acquisition of the data required to define, at the subwatershed scale, aquifer heterogeneity (geological, geochemical, recharge patterns, interrelation between aquifers, etc.). The characterization sequence was designed to optimize each field operation in terms of location, scale of acquisition, density and quality. Figure 4.2 summarizes the overall approach for the fieldwork sequence required for the subsurface characterization presented in this paper. The project relies on a wide range of characterization methods providing continuous indirect 1D, 2D, or 3D hydrogeophysical data, as well as continuous 1D and point direct data on hydraulic properties and groundwater geochemistry. Preliminary locations for surveys were selected based on a review of existing reports and ease of access through secondary and forestry roads. The subsurface characterization field work combined three approaches: 1) surface geophysics including a) ground penetrating radar (GPR) and b) electrical resistivity tomography (ERT); 2) direct-push methods including a) cone penetration tests (CPT) with soil moisture resistivity (SMR), b) soil sampling and c) installation of fully-screened observation wells in the granular aquifer; 3) borehole operations including a) flowmeter measurements while pumping, b) high vertical resolution (15 cm) packer slug tests, and c) multilevel measurement of geochemical parameters and groundwater sampling. The combination of indirect methods offered the flexibility to make decisions in the field, as data were acquired. For example, the previously acquired GPR data were used to choose the best locations and density of acquisition for the CPT/SMR soundings. CPT/SMR data available in the field allowed decisions to be made about the intervals for soil sampling or the design of direct-push observation wells.

To optimize each survey, the choice of the field work sequence was essential. GPR surveys are fast and provide reliable information on sedimentary structure and stratigraphic boundaries. However, the GPR has a limited capability to identify fluids or specific lithologies, hence the need for other surveys to provide this information. ERT surveys were thus carried out to investigate zones with attenuated signals found in some GPR profiles and provide information on the 2D lateral extension of electrical responses obtained by the CPT/SMR (high and low resistivity vertical profiles). ERT is a technique able to complement GPR by filling the gaps in zones where the electrical conductivity increases and renders the GPR penetration depth impractical. Following the interpretation of surface geophysical survey data, CPT/SMR sounding locations were chosen and wells were installed only at sites of interest for hydraulic conductivity measurements and fluid logging and sampling. Direct push operations were carried out to: 1) measure in situ CPT/SMR parameters in situ that are indicative of the heterogeneity of materials and hydraulic conductivity; 2) validate the stratigraphy of the aquifer (lithological changes and depth of the aquifer base); 3) sample soil at optimized intervals based on CPT/SMR responses; and 4) optimize well locations with CPT/SMR responses available “in situ” in real time, allowing the direct push installation of wells screened over the entire aquifer thickness without a sand pack. Fully-screened direct push wells were considered the most appropriate choice in the geological context of the study area, as they allow the acquisition of high-resolution hydraulic data (Paradis et al. 2011a). These measurements also provide continuous data of the subsurface stratigraphy in real time and identify thin layers of significantly different hydraulic conductivity. This methodology using continuous geophysical indirect measurements combined with direct high-resolution measurements ensures the coverage of the aquifer variability found in the study area.

High-resolution borehole measurements were carried out to measure directly, at numerous depth intervals, multiple physical and chemical properties of the aquifer material and groundwater. The objectives of this characterization are two-fold. The first objective was to acquire direct point data along vertical profiles to constrain 1D CPT/SMR soundings and 2D geophysical profiles previously obtained and to further assess the physical properties of the aquifer material under investigation. The second objective was to define discrete hydrofacies structures and properties within the heterogeneous aquifer. Finally, as a last step, high-resolution geochemical profiling and sampling were conducted in the wells to define geochemical conditions and delineate the leachate plume based on the results of the stratigraphic characterization program. As will be discussed later, these geochemical data were also used to correlate total dissolved solids with bulk conductivity measurements from CPT/SMR soundings (Figure 4.2).

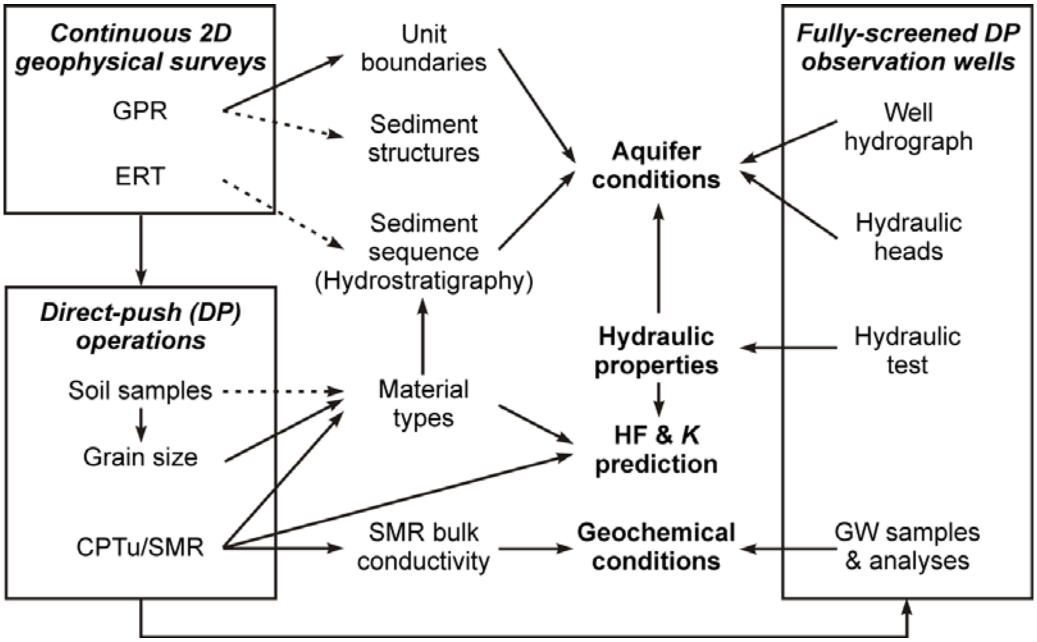

Figure 4.2 : Field data acquisition workflow for the subsurface characterization
The figure also illustrates the interrelation between the various components and the anticipated use of the field data

4.3.2 Data acquisition and field procedures

Table 4.1 summarizes the specifications of the instrumentation used for field characterization. Direct-push operations were carried out with a Geotech 605D drilling rig that is crawler-mounted for all-terrain capability. All operations followed standard techniques (ASTM 2000) and the approaches used are described in Fauveau et al. (2006) and Paradis et al. (2011a). CPTu/SMR survey locations were based on GPR data for seventy (70) of the eighty (80) surveys conducted throughout the study zone (Figure 4.1). A 15 cm^2 penetrometer cone with a 60° conical tip was used in accordance with ASTM D3441 standards (ASTM 2004). Most of the time, sediments were sampled in a different borehole, at a distance of 1 to 2 m from the CPT/SMR survey and the wells were installed at the same location as the CPT/SMR survey, providing co-located data from CPT/SMR and hydraulic tests. Occasionally, soil samples were collected below the maximum depth of the CPT/SMR surveys, where these surveys did not reach the aquifer base. Average sediment recovery was 83%. Cores were described and subsamples were used for grain size analyses, vertical permeability tests in laboratory permeameters (Paradis and Lefebvre 2013), X-ray diffraction for mineralogy (DRX), X-Ray Fluorescence (XRF) spectrometry and geochemistry.

Tableau 4.1 : Specifications of the field characterization operations

Survey type	Instrument	Configuration	Number of surveys	Total length (m)
GPR	Pulse Echo 100 volts	100 Hz antenna	43	28 000
ERT	Syscal Pro	96 electrodes 2-5 m wide	16	8 000
CPT/SMR	Geotech 605D	Vertek tool and data acquisition	80	660
Well installation	Geotech 605D	2 inch diameter PVC fully screened (0.024 mm openings)	29	305
Soil sample	GeoProbe Macro-Core -Soil sampler	PVC tubing 1.5 m long, 1 inch diameter	75	95
Flowmeter	EM Flowmeter 9721	15 cm measurement intervals	8	75
Geochemical profiles	YSI 556 MPS Troll 9000	Temperature, pH, Conductivity, Dissolved Oxygen, Redox Potential	70	600

At the same locations as the CPT/SMR soundings, fully-screened wells were installed over the entire thickness of the granular aquifer (Paradis et al. 2011a). Twenty-nine (29) observation wells were installed by hammering a 76-mm outside diameter (OD) metal casing equipped with an expendable point at its base that was hammered into the ground to the desired depth. Then, a 52-mm inside diameter (ID) and 60 mm OD fully-screened PVC tubing was inserted inside the metal casing before the outer metal casing was withdrawn. The thickness of the annulus between the OD of the metal casing and the OD of the PVC well was 8 mm. Slots in the PVC screen were 0.024mm (0.001 inch) wide, in accordance with the lower end of the grain size distribution of sediments. For well development, pumping and surging operations were performed with a Waterra inertial pump equipped with a foot valve and a surge ring having a diameter slightly smaller than the inside diameter of the PVC tubing. For each well, 0.5-m intervals were developed one at a time until no turbidity/sediments were observed in the pumped water, which often required development operations over more than a day at a given well. Slug tests were conducted before and after development in some of the wells to verify the efficiency of the development operation (Paradis et al. 2008 and 2011a).

Eighteen (18) flowmeter and eight (8) packer slug tests were carried out at 15-cm intervals into fully-screened wells (Paradis et al. 2011a). The intervals were selected based on CPT/SMR profiles to ensure the full coverage of geophysical responses by flowmeter and slug tests (Paradis et al. 2011b). Measurements of vertical hydraulic conductivity (K_v) were also obtained with laboratory permeameter tests conducted on soil samples, as well as from vertical interference slug tests (Paradis and Lefebvre 2013) and hydraulic tomography developed for that purpose (Paradis et al. 2012). Multilevel hydraulic head measurements were also made to help define the connectivity between hydrofacies identified along profiles. Distinct inflections in hydraulic head profiles are used to define hydrofacies (Meyer et al. 2008), whereas head differences between hydrofacies are used to define connectivity (Xu et al. 2013; Brunet et al. 2012). Those head measurements were made as part of the multilevel slug tests as well as in 5 additional wells to cover the whole study area.

Physicochemical profiles were measured in the wells as a first indicator of groundwater geochemical conditions found throughout the aquifer. The physicochemical parameters (temperature, redox potential (ORP), dissolved oxygen (DO), pH, and conductivity) were recorded with an YSI 556 probe. The probe was lowered in the well and measurements were taken at every meter. The probe was left stationary from 10 to 30 minutes at the same depth to allow parameters to stabilize before recording measurements. Profiling was carried out during three different seasons over a period of 4 years. Finally, groundwater geochemical sampling and analyses were carried out as described in Tremblay et al. (2012).

4.4 Results

4.4.1 Data processing and interpretation

4.4.1.1 Surface geophysics

GPR data processing included the following steps: 1) a dewow, 2) a static correction, 3) manual gain, 4) bandpass filtering, 5) background removal and 6) time to depth conversion. The availability of multiple geophysical surveys and CPT/SMR profiling also allowed the development of integrated geophysical data interpretation (Gloaguen et al. 2012). As shown on Figure 4.3, GPR surveys generally provided clear identification of the aquifer-aquitard (till) interface at the base of the granular aquifer, as well as sediment structures within the aquifer. These features were seen down to a depth of about 20 m at locations where the signal was not attenuated by the presence of clayey sediments that are electrically conductive (such as silts and tills). The impermeable base of the aquifer, corresponding to the top of the underlying till unit (confirmed by soil samples), is easily recognizable by the presence of strong GPR diffractors at depth, numerous hyperbolas due to the presence of till blocks and by the strong reflexion at the till surface. Throughout the study area, this strong interface reflector displayed important lateral topographic variations that could not have been inferred only from point data provided by drilled boreholes. In general, the GPR provided a high-resolution imaging of the heterogeneous internal structure of the sedimentary aquifer material. The numerous stacked reflectors are mainly due to contrasts in water content between layers, implying alternating coarse and fine sediments. In addition, within the study area, GPR sections often showed highly attenuated zones (e.g. bottom right of Figure 4.3). The conditions related to such zones had to be further investigated with ERT, CPT/SMR soundings, soil samples and groundwater analyses.

ERT data processing involved voltage inversion to infer the resistivity at the electrode locations. To guarantee proper inversion results, each electrical survey line was corrected for elevation.

The selected inversion algorithm used a smoothing constraint and the stopping criterion was reached in an average of five iterations. Furthermore, to ensure the data supported the final resistivity profiles, numerous inversion parameters were tested (Bélanger et al. 2010). ERT surveys provided a nearly continuous image of the subsurface in 2D and reduced the risk of undetected contaminants compared to local data such as observation wells. ERT surveys performed within the RIGDCC property limits showed low resistivity values compared to the relatively high resistivity values recorded outside the property or within the upstream part of the study area. Interpretation of ERT results benefited from the availability of other field data (CPT/SMR conductivity and groundwater geochemical analyses), which provided the ground “truth” to support ERT interpretation (Gasoty et al. 2012; Nyári et al. 2010; Zume et al. 2006).

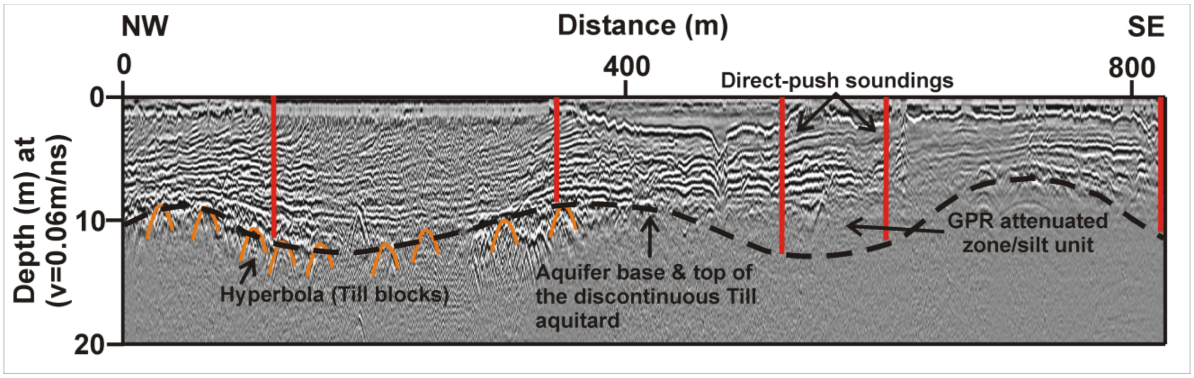

Figure 4.3 : Example of a GPR profile

Example of a GPR profile showing sediments structures (black and white contrasts) within the aquifer and indications of the surface of the till underlying the aquifer base. The nature of sediment in the aquifer and their thickness were confirmed by direct-push soundings illustrated in red, including CPT/SMR soundings (indicated by red lines) and soil sampling

4.4.1.2 CPT/SMR

CPT/SMR soundings provided a basis for the choice of observation well locations, allowed the definition of continuous 1D vertical stratigraphic profiles and the measurement of bulk electrical conductivity related to sediment types and the presence of the leachate plume. Sediment types present in the aquifer were identified by plotting the CPTu tip stress response versus sleeve stress on a Fellenius and Eslami (2000) profiling chart (Figure 4.4). The Fellenius and Eslami (2000) profiling chart was extended by Fauveau et al. (2005) and Ouellon et al. (2008), who added areas allowing the delineation of materials recognized in the study area on the basis of soil samples.

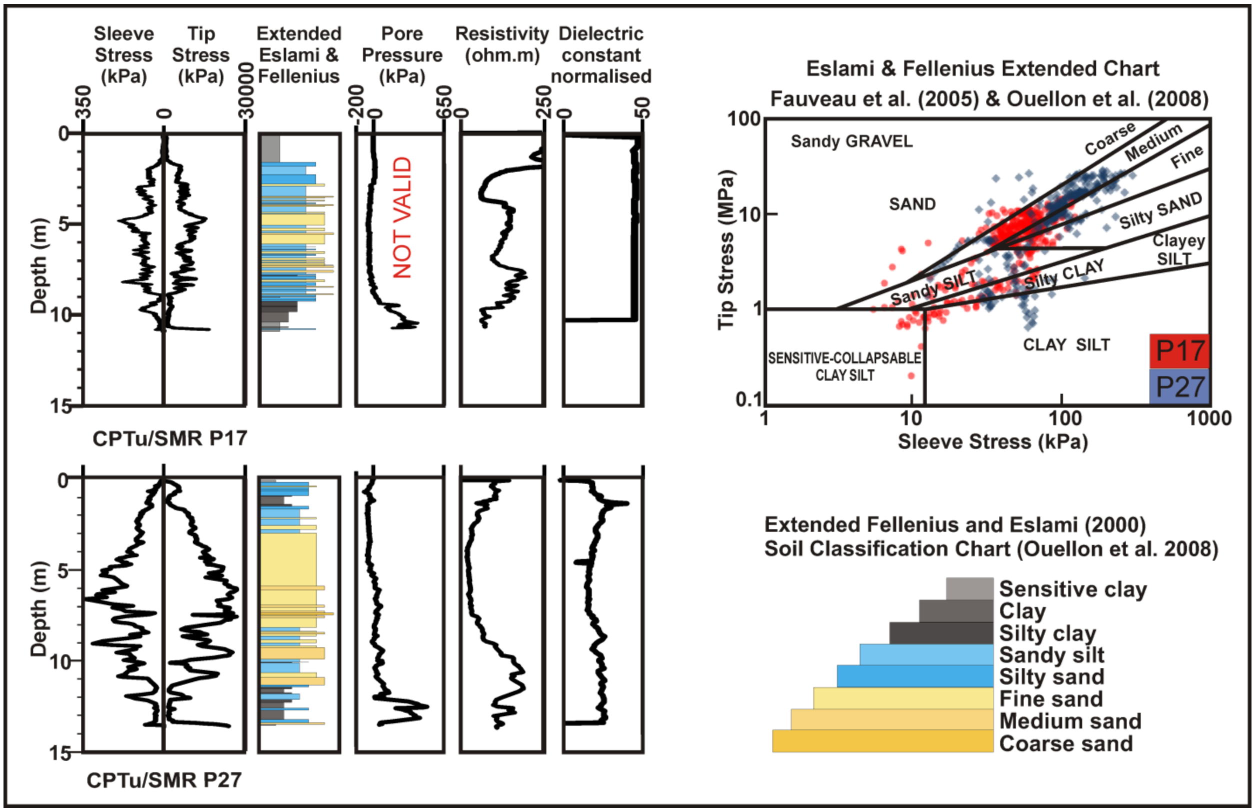

Figure 4.4 : Lithofacies interpretation from CPT/SMR soundings

Two examples of CPT/MSR soundings are shown on the left, including direct measurements and a pseudo-stratigraphic column of lithofacies interpreted from the Fellenius and Eslami (2000) CPT profiling chart extended after Fauveau (2006) and Ouellon et al. (2008). The chart to the right shows material classes interpreted for the two CPT soundings on the left

Soil samples were used to confirm soil types inferred from the chart. This chart provided a more coherent stratigraphic interpretation than other available charts (Lunne et al. 1997). An example of the real-time lithology data provided by CPT/SMR soundings in the field is illustrated in Figure 4.4. These vertical profiles of material types were extended in a lateral cross-section, at the correct elevation, to obtain a first indication of the spatial distribution and connectivity (lateral extent) of lithofacies (soil texture classes). Locations of CPT/SMR soundings, including those in Figure 4.4, are shown on the map of Figure 4.1, Figure 4.7 and on the geological section of Figure 4.12. Lithofacies identification based on the Fellenius and Eslami (2000) profiling chart is illustrated in Figure 4.4 for soundings P17 and P27. The two surveys are located on either side of the former landfill and are both down-gradient in relation to groundwater flow paths originating from the landfill. Based on the interpretation chart, on the upper right of Figure 4.4, P17 (located 300 m of the north side of the landfill) is composed of finer material than P27 (located right at the southern limit of the former landfill). Dielectric measurements from SMR were used to obtain sediment characteristics, such as porosity and water content. SMR also provide global electrical conductivity (including solid material and water).The resistivity value measured by the CPT/SMR was also useful to identify the presence of conductive fluids associated with the leachate plume migration. For instance, the SMR resistivity for P27 decreases significantly within the identified permeable sand from 2 to 10 m depth, indicating that the fluid saturating the aquifer at this location is conductive and most likely relates to the leachate plume.

4.4.2 Data Integration for the Development of a Conceptual Model

4.4.2.1 Definition of aquifer conditions

This section focuses on the first qualitative and quantitative data integration steps leading to the development of a representative hydrogeological conceptual model. The emphasis is placed on the interpretation of hydrogeological conditions. This is a prerequisite for other data integration steps as mentioned previously. Data integration steps beyond conceptual model development are illustrated in this paper but not discussed. The field data acquisition activities aimed to provide the basis needed to achieve the sub-objectives of all the components of the study illustrated on Figure 4.5, which also shows the interrelations between these components. The present section documents the integration of results allowing the definition of aquifer conditions, as well as the geochemical conditions that allowed the preliminary delineation of the leachate plume. These steps allowed the development of the conceptual model of the aquifer system that will also be described. Following the initial qualitative interpretation of the hydrogeophysical data, quantitative interpretation was done with a combination of data types and simple integration methods: image superposition of geophysical surveys, simple theoretical relationships between diverse parameters and basic statistics, application of a CPT soil classification chart and ordinary kriging. This shows that combining the various independent data provides a basis to develop a complete and representative conceptual model of the hydrogeological system.

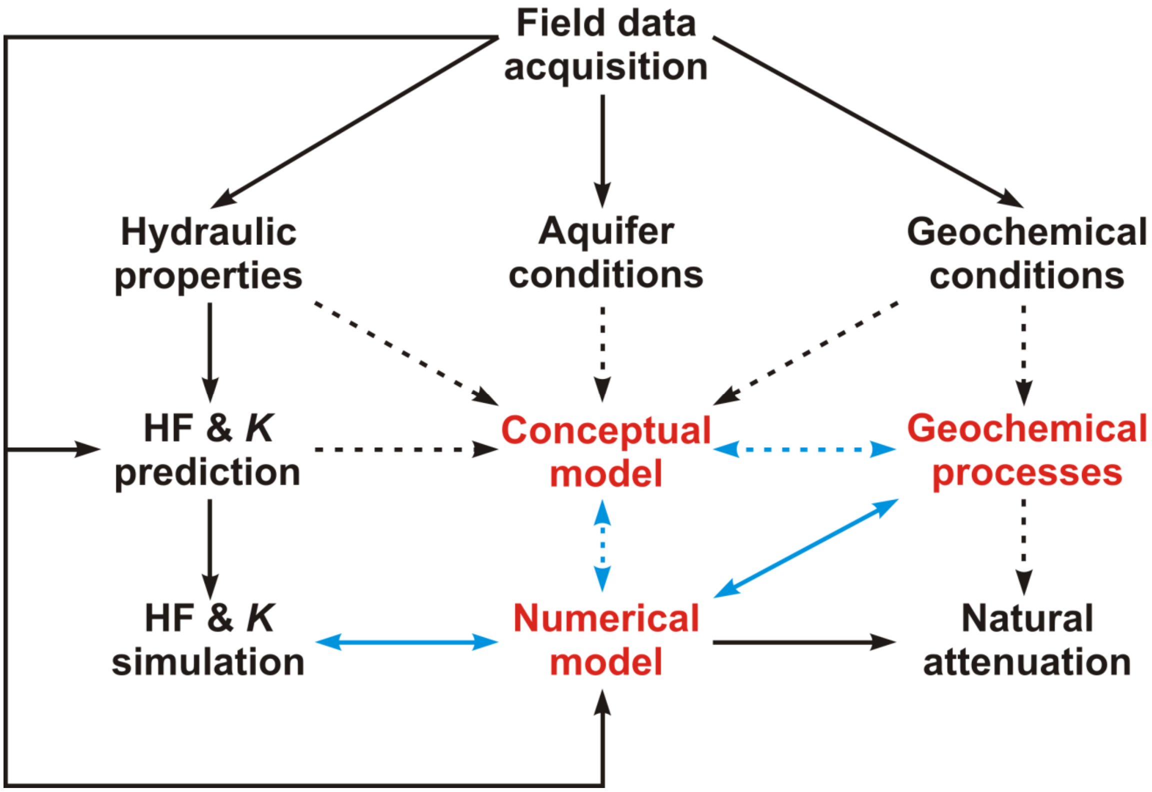

Figure 4.5 : General data integration workflow

Integration of the various types of data was an iterative process, especially between steps related to the conceptual model, numerical model, hydrofacies (HF) & hydraulic conductivity (*K*) simulations and interpretation of geochemical processes. Notes: continuous lines indicate quantitative relations, whereas dashed lines refer to qualitative relations. Lines with single and double arrows indicate uni- or bi-directional relations, respectively

4.4.2.2 *Hydrostratigraphy*

A qualitative visual comparison of hydrogeophysical data was first done through 3D data integration into Gocad (Paradigm 2010). The integrated data included GPR and ERT surveys as well as CPT/SMR soundings. 3D visualization helped the initial qualitative interpretation of geophysical data and an assessment of the completeness of the data set (Bélanger et al. 2010). Figure 4.6 provides an example of hydrogeophysical data integration, which is done by the superposition of sediment structures indicated by a GPR surveys over color shading indicating the electrical conductivity from ERT data inversion and 1D resistivity profiles from CPT/SMR soundings. 3D data integration improved the interpretation, as in Figure 4.6 where GPR data easily identifies the aquifer/till interface, which allows the till signature to be inferred on the

ERT data, thus also facilitating identification of the electrical response of the rock. Based on these results, local and discontinuous hydraulic links between the granular aquifer and the bedrock aquifer can be inferred, which has important implications for this study aiming to delineate the extent and natural attenuation of a leachate plume.

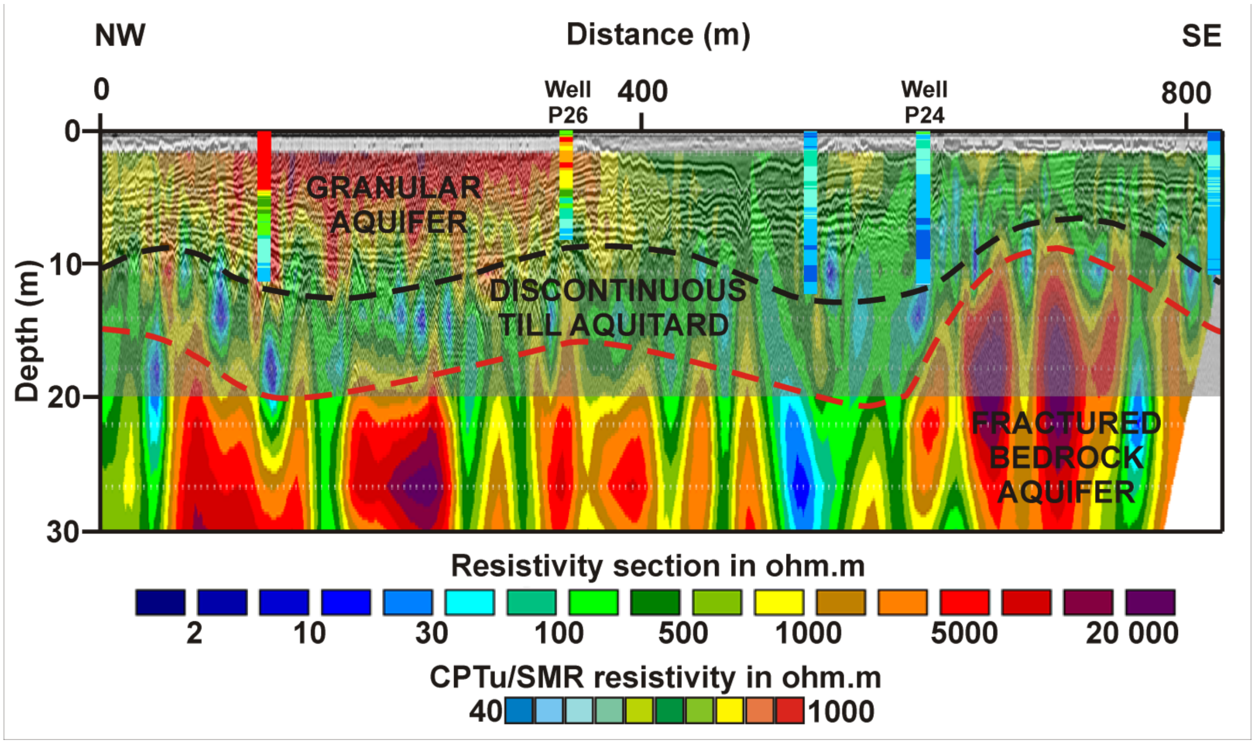

Figure 4.6 : Integrated geophysical data

2D GPR (shading) and inverted electrical resistivity (color scale) combined with 1D vertical CPT/SMR resistivity profiles. GPR data show sediments structures and variable thickness related to the aquifer base topography. ERT surveys provide the variation of the resistivity (confirmed by CPT/SMR) related to lateral changes in materials and fluids

Moreover, the highly interbedded stratigraphy seen on Figures 4.3, 4.4 and 4.6 (see also Figure 4.11) was confirmed by CPT/SMR mechanical responses and sediments cores. As mentioned earlier, within the study area, GPR sections often showed highly attenuated zones (e.g. bottom right of Figure 4.3 and Figure 4.6). In this example, the attenuated zones were found to correspond to 1) variations in groundwater conductivity related to an increase in total dissolved solids inducing higher bulk electrical conductivity that attenuates the GPR signal and 2) to thicker layers of fine material such as a compact silt layer of 1 m or more. On numerous surveys located in the mid-southern section of the study area, a laterally continuous conductive sediment layer (compact silt or clay) was found to highly attenuate the GPR signal starting at depths between 6 and 12 m, preventing the signal from reaching the reflector corresponding to the till at the base of the aquifer. Also, when the GPR reflectors were attenuated near the surface (depth of 0 to 2 m) of the survey, the attenuation was linked to layers of thick organic material. Overall, the GPR data were found to be quite useful to identify aquifer limits and sediment structures in this shallow semi-confined setting where sediments were mostly made up of sand. Since each method has its advantages and limitations, the qualitative interpretation of Figure 4.6 clearly illustrates the advantages of integrating multiple data sources in a hydrogeological characterization. Each method provides part of the answer, which leads to firmer conclusions on the nature of materials and fluids.

4.4.2.3 Aquifer limits and thickness and leachate extent

As part of the conceptual model development, the delineation of aquifer limits and thickness of the granular aquifer can be defined efficiently at the subwatershed scale by combining different sets of data acquired from the integrated characterization (Figure 4.2). The integration of multiple data sources can provide specific and representative insight on aquifer conditions as shown in Figure 4.7: 1) the top and base surfaces (the aquifer envelope/sediment thickness) and the nature of sediments within this envelope (inferred from CPT/SMR data and soil samples, see Figure 4.4); 2) the piezometric surface elevation and the flow directions from the landfill, and; 3) the delineation of the leachate plume and locations of potential leachate concentration “hot spots” indicated by SMR bulk conductivity (Figure 4.7d discussed later). All surfaces were interpolated using ordinary kriging (Davis 2000) in the geostatistical analysis tools of ArsGIS (ESRI 2008). The data points used for the interpolations included continuous indirect data and local direct data. Figure 4.7a shows higher elevation of the ground surface at the eastern boundary of the study area, along the groundwater divide oriented north-south, and shows, perpendicular to the divide, another topographic high running underneath the landfill location. Typically, small variations in topography and their related structures will affect recharge and runoff in areas where variability in ground elevation is low, where the water table is close to the ground surface, the aquifer is shallow and surface soil is composed of low permeability material (Frei et al. 2010). Therefore, the high-resolution DEM serves as a first indicator of potential recharge, discharge, and runoff zones and points out potential groundwater flow directions. The

DEM was also be used to correct GPR and ERT data and for the interpolation of aquifer thickness.

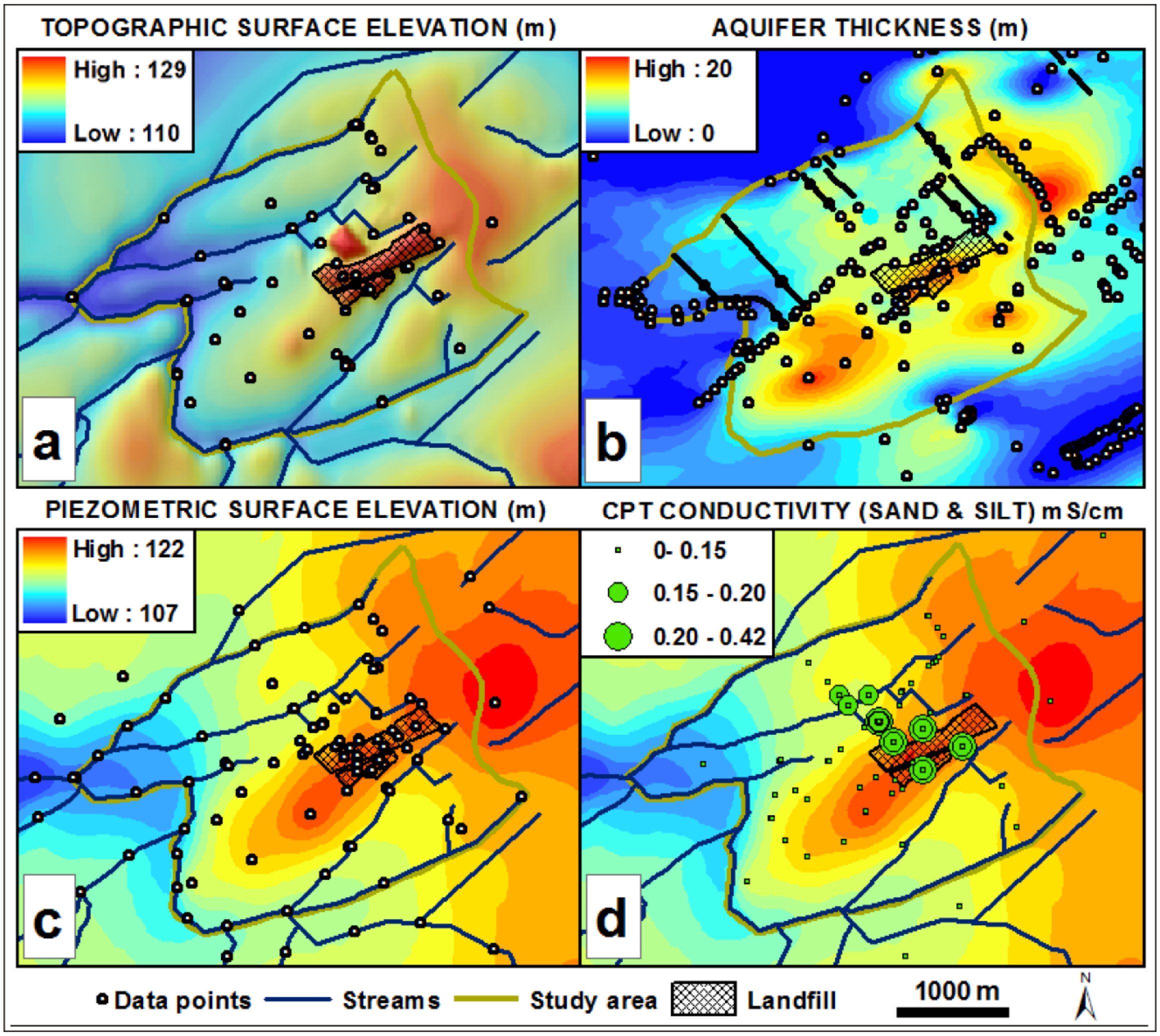

Figure 4.7 : Hydrogeological conditions

Hydrogeological conditions in the study area derived from characterization data: a) digital elevation model (DEM), b) aquifer thickness from the soil surface to the top of the till layer, which is the aquifer base, c) piezometric surface, and d) CPT/SMR maximum conductivity recorded in sands and silts superposed on the piezometric map

Secondly, the thickness of the aquifer material was interpolated from points derived by subtracting the elevation of the till surface from the DEM (Figure 4.7b). The till surface (base of the granular aquifer) was interpolated from the strong reflector located at the base from continuous indirect GPR data (as shown on Figure 4.3), point direct data from CPT/SMR and soil samples confirming the total thickness, borehole descriptions from private consultant reports, and rock outcrops. The map shown in Figure 4.7b indicates high variability in the base of the aquifer, which could not have been defined at this scale from the conventional hydrogeological characterization based solely on boreholes.

Finally, a piezometric surface (Figure 4.7c) was interpolated from water level measurements in wells combined with survey points along creeks at key locations. The piezometric surface shows stronger horizontal hydraulic gradients on both sides of the former landfill. At this location, the site acts as a local groundwater divide line and sediments are coarser and the aquifer is thicker (up to 20 m). It can be inferred that higher groundwater elevations at the landfill location are possibly the results of a local water table mound (Christensen et al. 2001) below the western end of the former waste pile. Groundwater flow starts somewhere along the groundwater divide line, however the local highest levels of the piezometric surface are observed below the former waste pile and the steepest gradients are seen on both the north and south sides of the waste pile.

Figure 4.7d shows the groundwater elevation map (same as Figure 4.7c) with CPT/SMR electrical conductivity data superimposed. High CPT/SMR conductivity values provide indications of the potential extent of the leachate plume. The lack of CPT/SMR data in the southern part of the study area, especially south of the landfill, is due to the inability to access this densely forested area. However, old observation wells were accessible in this area and groundwater was sampled at those wells for geochemical analyses to complement the data set. Figure 4.7d shows that CPT/SMR bulk electrical conductivity (for sand and silt recognized by the CPT chart of Fellenius and Eslami (2000), Figure 4.7) is a useful indicator of zones of higher conductivity within the aquifer and also reveal potential leachate migration paths. The delineation of higher conductivity zones could lead to the identification of "hotspot" locations within the waste pile. This is further discussed in the next section.

4.4.2.4 Hydrogeological conditions

Besides the recognition of hydrofacies, CPT/SMR-derived data also allow the identification of recharge and discharge zones as well as natural geochemical conditions versus anthropogenic conditions related to leachate emissions. Once high potential zones of leachate emissions are identified (highly conductive zones), further investigations can be done with CPT/SMR resistivity profiles. First, based on the same Fellenius and Eslami (2000) lithofacies interpretation

(Figure 4.4), CPT/SMR bulk conductivities can be classified according to material type using basic statistics. Figure 4.8a shows that extreme electrical conductivities are found in sand and silt materials, which implies that leachate migration occurs mostly in the more permeable hydrofacies. In addition, these extreme values are found between depths of 2 to 10 m (not shown). Moreover, Figure 4.8a shows that median values of bulk electrical conductivity for each type of material increases as the type of material becomes finer (higher porosity and more electrically conductive sediments, e.g. clayey silt), which is consistent with theoretical values found in the literature (Rubin and Hubbard 2005; Mussett and Khan 2000). This relationship between electrical conductivity and material types can be used to derive *K* estimates from ERT data at the regional scale (Ruggeri et al. 2011, 2013). Silt units (porosity of 0.44 derived from pycnometer measurements) have a higher electrical conductivity than medium to coarse sand (porosity of 0.35). A groundwater with higher dissolved solids concentration will increase the conductivity of the typical values obtained for a given material, therefore the conductivity of the fluid will drive the increase of the bulk conductivity of the CPT/SMR for a given material. Consequently, as shown on Figure 4.8b, a linear relation can be derived by plotting the median CPT/SMR bulk conductivity values recorded in sand and silt at each well site against the average conductivities of the groundwater measured in this well. As highly conductive fluid drives the increase in bulk conductivity of the CPT/SMR data, the wells affected by leachate can be identified with a certain level of confidence. The results obtained from the correlation show that CPT/SMR bulk conductivity can help identify soil types as well as water types. Such capability is important since CPT/SMR soundings also provide high-resolution and vertically continuous data in real time.

The relation between the median CPT/SMR bulk conductivity in the sand and the average groundwater conductivity measured in the wells supports the interpretation of CPT/SMR data in terms of fluid conductivity (Figure 4.8a). By deriving the fluid conductivity from the CPT/SMR data with a correction applied to the sand, full vertical profiles of fluid conductivity could be obtained, thus providing the variability of the fluid conductivity through the entire aquifer thickness (Figure 4.8b). These data could also guide the installation of discrete wells in the most contaminated layers.

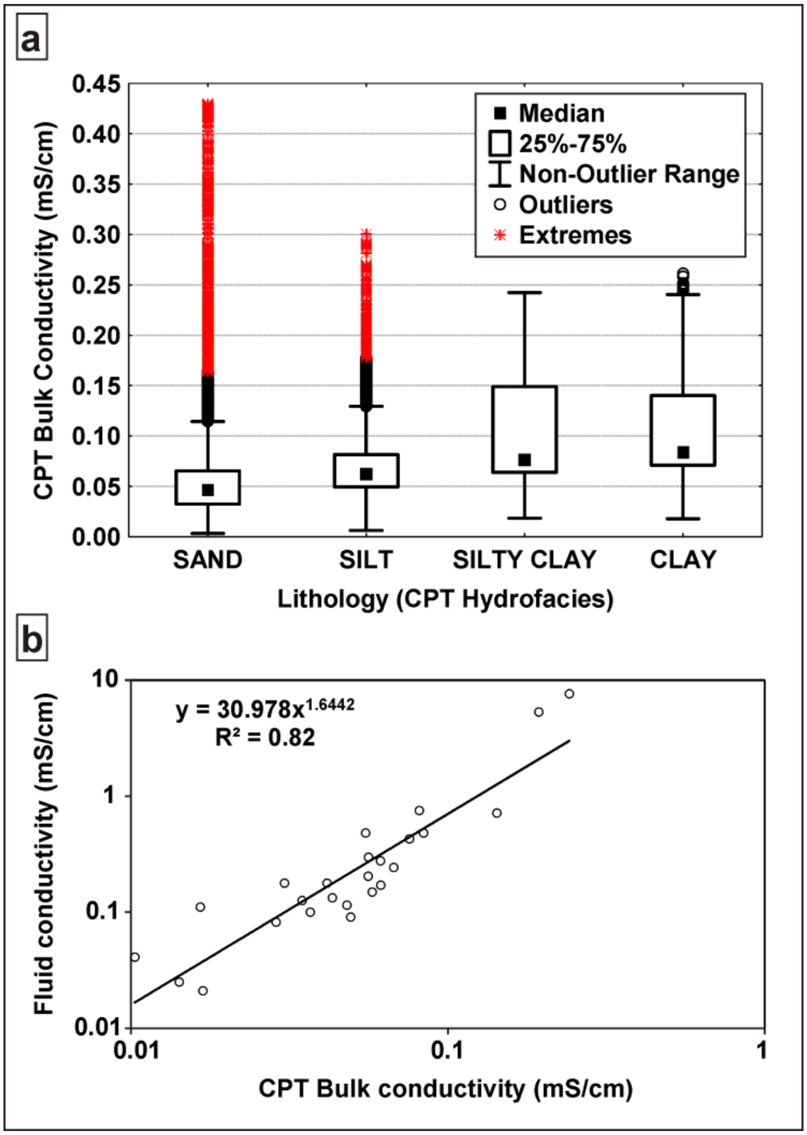

Figure 4.8 : CPT/SMR bulk electrical conductivities related to material types
Recognition of leachate from CPT/SMR bulk electrical conductivities. a) Box plots of CPT/SMR conductivities by material types (from Fellenius and Eslami (2000) CPT profiling chart shown on Figure 4.4). b) Correlation of CPT/SMR electrical conductivities (only in CPT sand hydrofacies) to groundwater electrical conductivities obtained from sampling of observation wells where CPT/SMR soundings were also run. Extreme values of CPT/SMR conductivities in 4.8a are inferred to indicate the presence of leachate, which appears to migrate only in the more permeable sand and silt hydrofacies. In 4.8a, the lower box value (LBV) is the 25th percentile and the upper box value (UBV) is the 75th percentile. Outliers are points with values exceeding UBV+1.5*[UBV-LBV]. Extremes are points with values exceeding UBV+3*[UBV-LBV]

Moreover, another relation can be derived from the measured CPT/SMR bulk conductivity to provide total dissolved solid (TDS) in mg/L, as TDS is proportional to fluid conductivity in micromhos (TDS = (0.54-0.96)*groundwater conductivity (uS/cm); Hounslow 1995), a linear regression can be defined for TDS calculated from the geochemistry of major ions analyses (160 samples) plotted against the measured conductivity of groundwater in each wells (not shown). TDS values can thus be estimated for all the CPT/SMR profiles to a very high vertical resolution. Since there are water quality standards for TDS, for which the maximum concentration limit for potable water is 500 mg/L (WHO 2011), CPT/SMR data can further assess and guide the investigation of zones at risk of contamination. Once the data set is cleared of irrelevant values, using CPT/SMR bulk conductivity (continuous vertically) to derive fluid conductivity values and TDS provides information that complements geochemical data at a contaminated site where the plume has high dissolved solids.

For the purpose of defining hydrogeological conditions, data collected from borehole operations can provide insight on the different contexts of the study area. Classification of three different hydrogeological contexts (recharge, intermediate and discharge) can be achieved using the following parameters (Table 4.2): 1) TDS estimates obtained from CPT/SMR bulk conductivity

Tableau 4.2 : Summary of indicators used to infer hydrogeological contexts
(R: recharge zones. I: intermediate zones. D: discharge zones. L: presence of leachate)

Context	Groundwater temperature (min-max °C)	Depth of temperature variability (m)	Specific conductivity (mS/cm)	pH	DO %	Relative topography	Vertical flow direction	TDS from CPT/SMR (mg/L)
R	4.5-12	8	0.01-0.09	acidic	50-90	High	downward	0-200 increases with depth
I	4.5-12	4	0.1-0.2	neutral	10-50	Medium	0-5 m downward 5-10 m upward	100-200, increases with depth
D	6-8.5	3.5	0.2-0.3	alkaline	0-10	Low	upward	constant along profile at 250
L	5-12	n/a	>0.4	neutral	0-10	High-low	n/a	Above 300

data (Figure 4.9); 2) temperature and physicochemical profiling at different times of the year (Figure 4.10) and; 3) high-resolution profiles of hydraulic conductivity and head between packers (Figure 4.10).

The groundwater within the recharge group showed larger temperatures fluctuations over different seasons and the temperature recorded varied down to a depth of about 8 m below ground surface. At these wells locations, the aquifer was recognized to have coarser sediments and vertical hydraulic head gradients indicating downward flow. Groundwater at these well locations also had higher concentration of dissolved oxygen and lower pH indicative of younger groundwater for the area. This group (named recharge area) was subdivided in two subgroups (high and moderate). The high recharge area subgroup has lower vertical hydraulic head

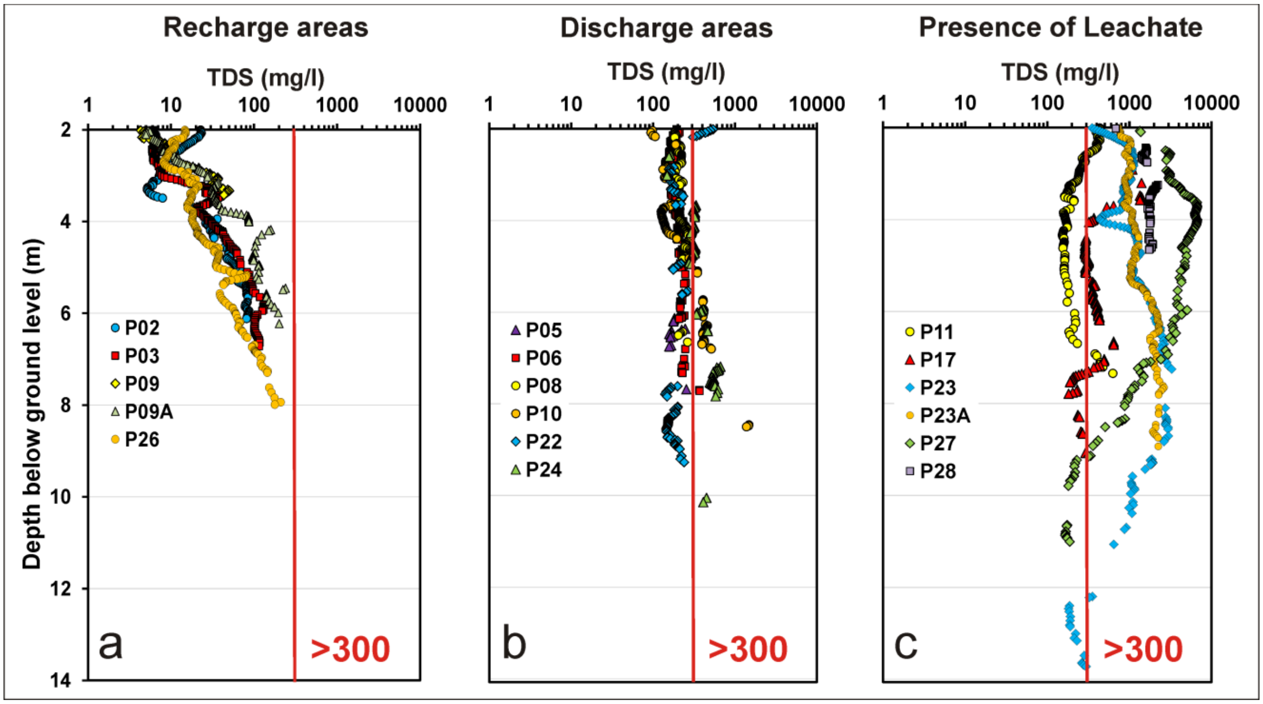

Figure 4.9 : TDS profiles inferred from CPT/SMR bulk resistivity

Profiles of estimated TDS (mg/L) in Fellenius and Eslami (2000) sand hydrofacies (Figure 4.4) derived from CPT/SMR bulk resistivity measurements (using Figure 4.8b correlation). TDS profiles are grouped according to hydrogeological contexts (recharge and discharge areas) based on TDS values and trends with depth. The inferred presence of leachate is based on an empirical local threshold of about 300 mg/L (note the TDS log scale) gradients (near zero m over 11 m depth) but had greater *K* in sands along the profile (10^{-4} m/sconfirmed by flowmeter data and slug tests) and TDS values derived from CPT/SMR measured resistivity were lower. The second subgroup (moderate recharge areas) had stronger vertical hydraulic head gradients (0.5 m over 13 m depth) but sediments were interlayered with finer material compared to the high recharge group which lowered *K* to 10^{-5}-10^{-6} m/s. The second group was defined as intermediate because the recorded vertical hydraulic head gradients indicated downward flow throughout the first upper section of the well (0 to 4 m) but upward flow in the deeper section (4-10 m). This intermediate group had the same range of temperature measured at the surface as the first group, although temperature variability was not noticeable below 4 m depth. Finally, the last group identified as discharge zones was defined based on the following conditions: vertical hydraulic head gradients indicating upward flow, alkaline pH, and estimated TDS around 250 mg/L showing no drastic fluctuations along the profile (Figure 4.9 and 4.10). Temperature and physicochemical profiles acquired over a 4-year period at different times of the year, combined with the CPT/SMR and hydraulic measurement in the wells, thus allowed the definition of three distinctive groups: recharge, intermediate and discharge.

The context of each group and their respective definition (Table 4.2) can be assigned at each CPT/SMR location throughout the study area (Figure 4.11). Spatial distribution indicates a great deal of variability locally; areas defined as high potential for recharge are located on local topographic highs and cover a small percentage of the total areas investigated (Figure 4.11). This preliminary mapping of hydrogeological contexts can guide a study on spatial distribution of recharge rates and orient the types and location of further field surveys required to quantify the recharge processes. Milosevic et al. (2012) stated that contaminant mass discharge into both ground and surface water is a key parameter in the risk assessment of old landfills. Understanding the processes and dynamics of groundwater and surface water interaction is a prerequisite to the prediction of a groundwater contamination plume migration. Many novel integrated approaches and software have been developed to undertake this new research challenge (Gilfedder et al. 2012; Milosevic et al. 2012; Weill et al. 2011; Fleckenstein et al. 2010; Stadler et al. 2010; Kollet and Maxwell 2006). Locating the discharge and recharge areas can lead to the understanding of groundwater and surface water interactions, which is a first indicator of groundwater flow paths.

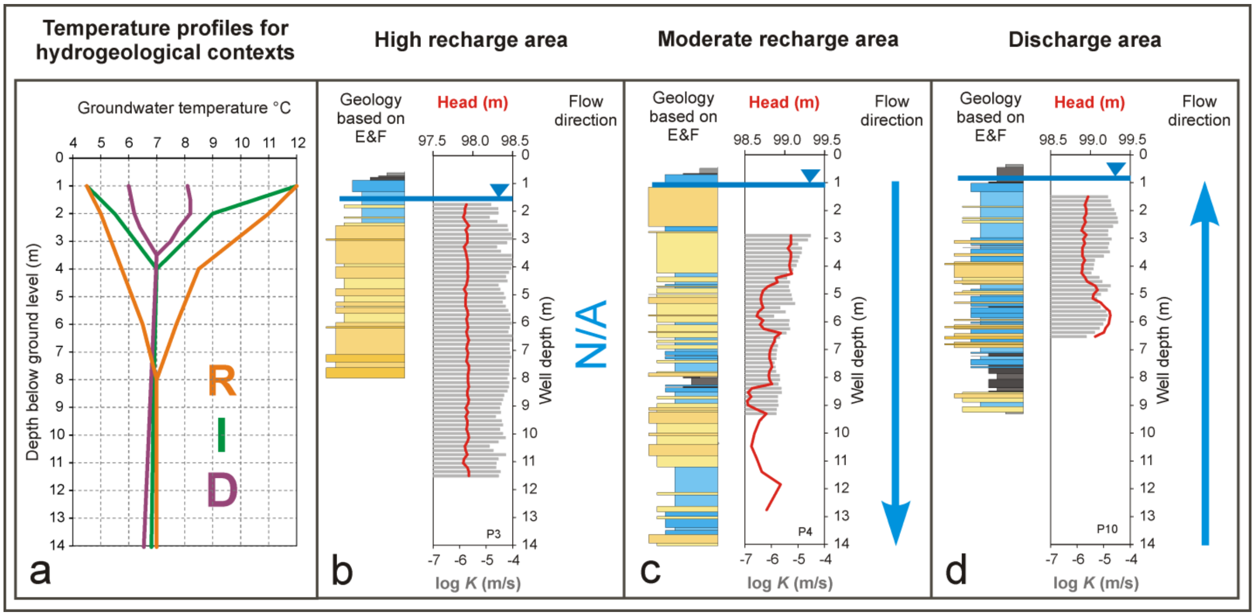

Figure 4.10 : Hydrogeological contexts

Classification of hydrogeological contexts on the basis of groundwater temperature profiles in boreholes (4.10a) and vertical hydraulic gradients (4.10 b, c and d) (other features considered for the classification are summarized in Table 4.2). The temperature profiles on the left are envelopes of typical annual groundwater temperature variations by hydrogeological context. *R* stands for recharge, *I* for intermediate and *D* for discharge. The three hydraulic head vertical profiles (red lines) are typical of hydrogeological contexts. Heads are shown along with CPT hydrofacies (Figure 4.4) and horizontal hydraulic conductivity profiles were measured along with heads within packers in fully-screened wells

4.4.2.5 Hydrogeological conceptual model

Based on the spatial distribution of data illustrated in Figure 4.11, a conceptual cross-section was drawn, integrating aquifer-aquitard boundaries and material types (Figure 4.12). To develop the section, soil samples combined with CPT/SMR data provided the definition of hydrostratigraphy and the vertical correlation of the data locally. Surface geophysical surveys (GPR and ERT) combined with CPT/SMR surveys allowed mapping of the lateral subsurface heterogeneity and provided complementary information on various hydrofacies in the study area, and on important lateral variations in aquifer thickness. At the same time, data derived from the borehole operations complemented the data set by refining the selected hydrofacies and inferring hydraulic conductivities related to each unit. Finally, the qualitative data integration of all surveys allowed the conception of a geological model representative of the aquifer heterogeneity and the delineation of the leachate plume. The cross-section also illustrates the variability in the stratigraphy.

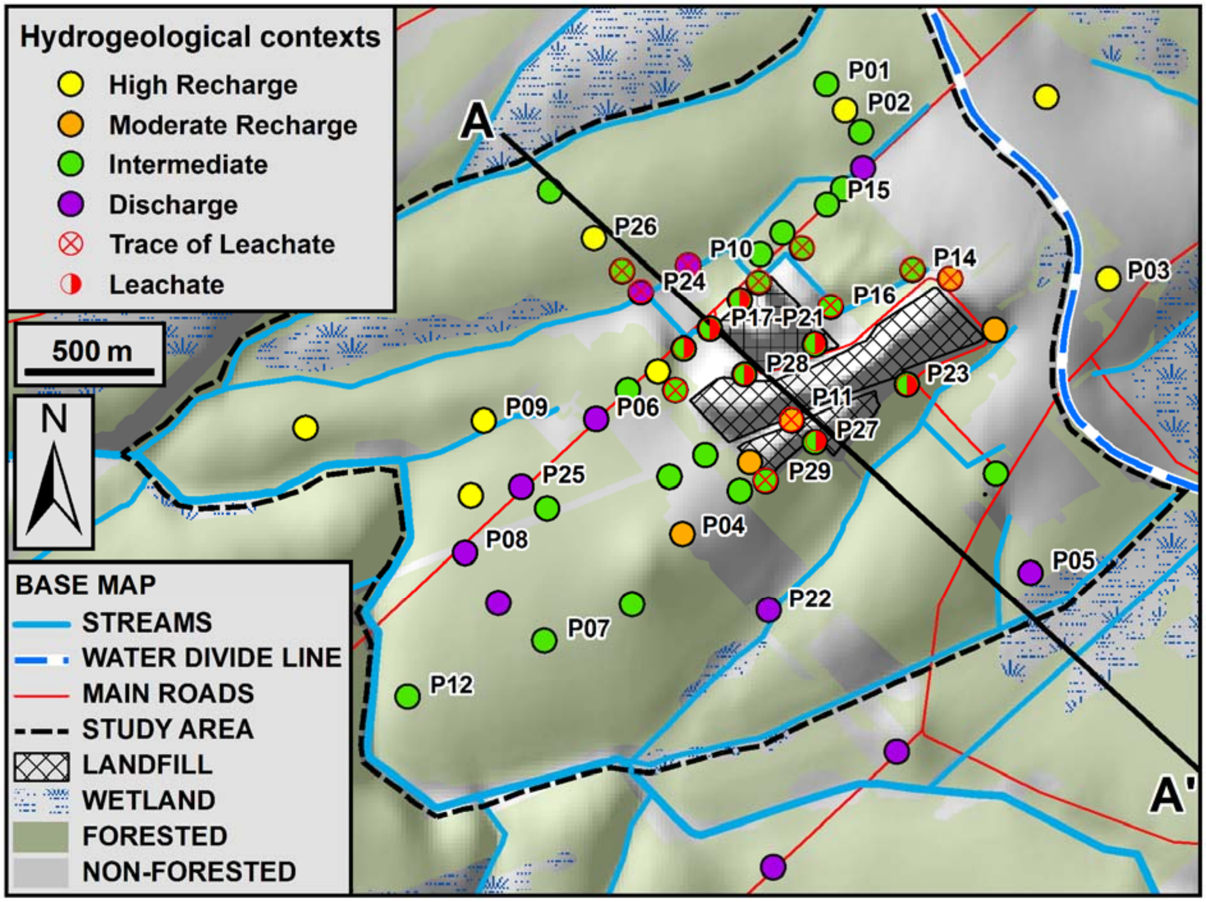

Figure 4.11 : Spatial distribution of hydrogeological contexts

Spatial distribution of hydrogeological contexts at borehole and CPT/SMR sounding locations (see Figure 4.1) based on indicators summarized in Table 4.2. The A-A' line is the location of the section shown in Figure 4.12

Geological layers within the granular aquifer were mostly found to be discontinuous at the subwatershed scale. K_h values of the granular aquifer range from slightly over 10^{-4} m/s, for a small proportion of coarser material, to below 10^{-6} m/s for the finer sediments. Coarser sands are located along the groundwater divide line or at local topographic highs. As depth increases, grain size generally becomes finer. Some sediment layers have bimodal grain-size distributions which render the sediment very compact and dense. In the center of the study area, right below the decommissioned landfill, at an elevation of 115 m asl, such a layer consisting of 2 m of dense sediments limits the infiltration of water. Finally, at an average elevation of 105 m asl and below, the base of the granular aquifer reaches the surface of a discontinuous aquitard composed of glaciomarine silt overlying a till unit (shown on Figure 4.12).

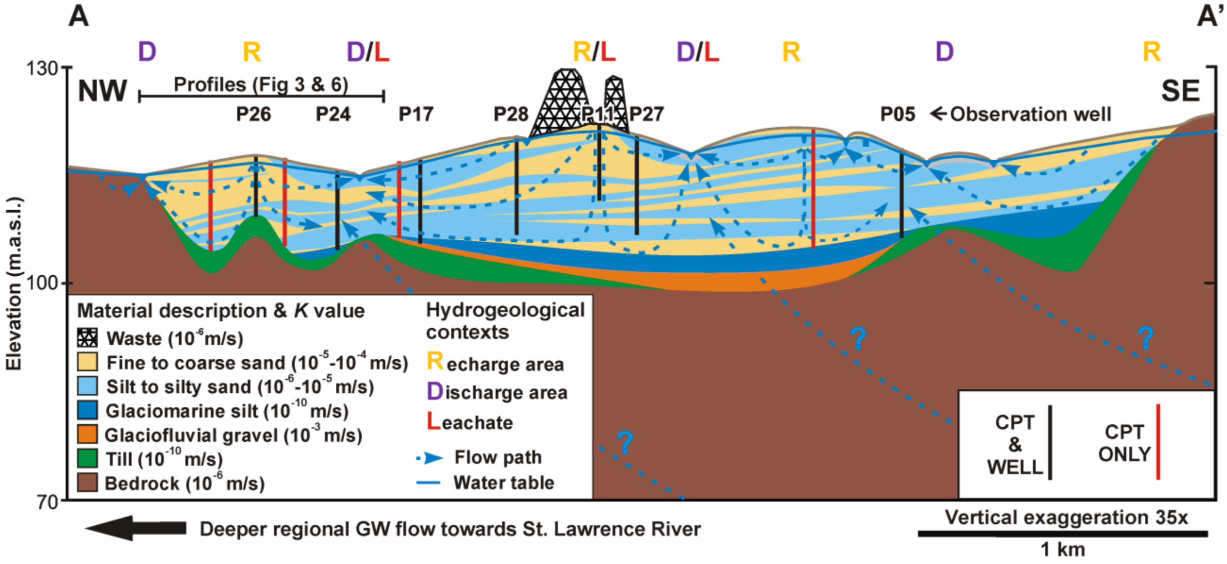

Figure 4.12 : Conceptual model

Conceptual model (cross-section location shown on Figure 4.11). This figure shows the spatial distribution of geological materials (inferred from CPT data interpreted from the chart of Figure 4.4) and groundwater flow based on the data collected in phase 1 of the integrated characterization workflow. Letters refer to hydrogeological contexts defined on the basis of multiple indicators (geochemical composition, physicochemical parameters, temperature and hydraulic gradients; Table 4.2 and Figures 4.8, 4.9, 4.10 and 4.11). Numbers refer to observation wells and the black lines represent wells, whereas red lines represent only CPT/SMR soundings. Geological information for various lithofacies below the reach of CPT data was completed with numerous well logs and grain size analyses available from private consultant reports and registered private wells (not shown)

The glaciomarine silt, deposited shortly after the last glacier retreat, fills depressions of the till unit surface and contains abundant ice-rafted debris. The 0.1 to 3 m thick glaciomarine silt aquitard is overlying either a discontinuous glaciofluvial gravel layer (0.1 to 2 m) or the glacial till (0.1 to 5 m) (Figure 4.12). As for the till aquitard, it is also discontinuous and overlies a sedimentary fractured rock aquifer. The till found in the study area is pink, has a very compact calcareous coarse sand to silty sand matrix and contains fragments of 1 to 5 cm of various rock types. The hydraulic conductivities of the glaciomarine silt and the till recorded by Gauthier et al. (1993) is 10^{-10} m/s for both units. Although the *K* value of the till aquitard is usually very low, Gerber et al. (2001) has demonstrated an anomalously high vertical groundwater flow velocity (>1 m/y) through a thick sandy silt aquitard previously assumed to be of very low permeability. The rapid recharge was attributed to fractures and sedimentary heterogeneities within the till (Gerber et al., 2001), which shows that exchange between the bedrock aquifer and the granular aquifer could exist locally via the till aquitard despite its larger thickness in some locations. No hydraulic data are available for the glaciofluvial gravel, which is only found locally.

As mentioned previously, the data integration also allowed the identification of recharge and discharge zones indicated at the top of Figure 4.12. Furthermore, it is possible to infer the presence of two types of groundwater flow regimes: 1) a rapid local superficial flow system that quickly discharges in the surrounding creeks and 2) a slower deeper groundwater flow system that starts from the landfill high water level mound and flows initially directly downward and finally towards the identified main discharge zones. The TDS profiles derived from measured CPT/SMR resistivity data support the inferred flow patterns. For example, the TDS profiles on Figure 4.9 illustrate 1) a zone of high recharge, characterized by well-sorted medium to fine sand and TDS shows an increase in ionic content as groundwater infiltrates deeper into the aquifer; 2) a discharge area characterized by low variability in TDS along the profiles (except for wells P10 and P24 at depths where leachate migration from the landfill could occur and/or groundwater from the bedrock aquifer emerges into the granular aquifer). At these locations the vertical hydraulic gradients are inverted as shown on Figure 4.10 for P10 (similar to well P24), the sediments are composed of finer material and the temperature has practically no variability along the profile. Finally, groundwater is cooler and varies only near the ground surface. For contaminated areas, TDS values reach 500 to 6000 mg/L and spans a large depth interval, showing that contamination covers several meters.

In summary, the conditions found in the aquifer were qualitatively integrated to develop a conceptual model of the aquifer system. This conceptual model was used as a basis to develop a numerical model of groundwater flow and mass transport. This numerical model was calibrated against hydraulic heads and verified with the use of geochemical tracers and groundwater ages. As illustrated in Figure 4.5, there were mutual contributions, through the cyclic development of the conceptual model, the understanding of geochemical processes and the numerical modeling. These developments led to a coherent representation of the aquifer system and allowed the assessment of the natural attenuation of the leachate plume and the risk to receptors.

4.5 Discussion

4.5.1 Field data: advantages and collection challenges

Data collection during aquifer characterization is site-specific and is the key initial step. For multidisciplinary teams imlicated in integrated characterization involving a wide variety of data types, documentation of data acquisition and processing protocols is necessary, as many users need to utilize the same data set, and thus also need to be aware of the validity and limitation of the data. The present section discusses some of the lessons learned about the acquisition and information value of various data types for the purpose of developing a conceptual model an aquifer system.

4.5.1.1 Surface geophysics

GPR profiles were acquired at the onset of the project and were used to guide the positioning of ERT surveys as well as CPT/SMR soundings and observation wells. GPR facilitated the choice of other surveys at locations of interest by providing a “context” for further data acquisition and “questions” to be answered by complementary data, especially about the nature of materials and fluids. ERT surveys were carried out to investigate attenuated zones appearing on the GPR profiles and to confirm the electrical responses and 2D lateral extent of materials shown by the 1D CPT/SMR soundings (high and low resistivity vertical profiles). Effective interpretation of ERT results required the availability of other field data (CPT/SMR conductivity data and material types; groundwater geochemical analyses). The nature of the soil at the surface of the study area made ERT acquisition difficult, especially following periods of rain, because puddles would form on the roads and remained present for weeks. Moreover, multiple zones around the landfill were inaccessible. Therefore the density of ERT surveys was not sufficient to define the plume concentration as it migrates away from the landfill, as initially planned.

4.5.1.2 Direct push operations

In the context of the study area, CPT/SMR soundings proved to be an excellent tool to define the hydrostratigraphy of the site; it provides reproducible measurements, and required few soil samples to identify the various types of materials present and their general spatial distribution, thus reducing the cost of the site investigation while increasing the level of detailed information. However, CPT/SMR soundings could not always reach the base of the aquifer. As shown by soil samples, a dense layer of well-sorted fine sand and silt restricted penetration severely at depths in the range of 6 to 12 m, despite generally good rig anchoring. In some cases, Rotary Percussion Sounding System (RPSS) operations were conducted only to drill through this compact layer and then it was possible to proceed again with the CPT/SMR survey to the base of the aquifer (top of the underlying till layer). In other instances, the base of the aquifer was identified using only RPSS or direct-push soil sampling. The profiling chart only gives an approximation and first indication of soil type and it must be adjusted to actual local site conditions by correlating CPT/SMR profiles to soil samples and grain size data. Paradis et al. (2011b) thus developed a learning machine approach to provide a more representative classification and recognition of hydrofacies and to predict their hydraulic conductivity using CPT/SMR data.

The CPT/SMR operations reduce disturbance to the formation (thus provide accurate conductivity data at a precise depth), and are very efficient and low cost (Bianchi et al. 2001; Lunne et al. 1997). In a shallow granular aquifer, when considering short travel distances between each survey location (such as around a landfill), up to 5 surveys of an average 10 m per day can be carried out. The high heterogeneity of the material and the small thickness of each lithofacies would have complicated the selection of depth intervals for conventional or multi-level observation well installations. Instead, fully-screened wells over the entire aquifer thickness were installed by direct push to be sampled at discrete intervals using packers. With this type of installation procedure, sediments are maintained in direct contact with the screen, there is minimal disturbance to the surrounding aquifer, and the use of gravel packs is avoided. However, wells must be thoroughly developed to obtain representative information on hydraulic properties and geochemical conditions of the aquifer (Paradis et al. 2011a; Butler et al. 1998).

4.5.1.3 Borehole operations

Well surging was strenuous at the study site; the fine material of the aquifer made the operations last many days for each well. During pumping, the bottom of the wells could fill up with 2 to 6 m of fine sediments. The wells bottom were cleaned by pressing the surge block directly on the sediments and lowering the block as the sediment was removed. Considerable attention was paid to well installation and development to maximize the quality of hydraulic conductivity and geochemical measurements obtained from observation wells. According to Mayo (2010), numerous factors can affect water quality of samples obtained from a heterogeneous aquifer and, since some aquifers have spatial chemical heterogeneity, special attention needed to be paid to well design. Long screened wells can induce cross-aquifer contamination in multilayered aquifers as groundwater from the horizon with the greatest hydraulic head flows into the wellbore and displaces water from horizons with lower hydraulic head. Water can move up or down the wellbore, depending on the direction of vertical head gradients. While pumping can remove the mixed groundwater from the wells and contaminated horizon, the required volume of water purged from the well to help ensure a representative sample of the aquifer is unknown since the true extent of cross contamination is difficult to quantify. Long screen wells dilute water drawn from contaminated horizons with water from non-contaminated horizons, while short screen wells and discrete sampling may bias the sample by either missing contaminated or non-contaminated horizons (Hofmann et al. 2010; Mayo 2010; Appelo and Postma 2005). A study on direct push profiling (Hofmann et al. 2010), demonstrated that undisturbed groundwater samples at high vertical resolution are essential for the understanding of groundwater flow and transport systems. In this study, wells screened over the full thickness of the aquifer were selected over direct-push sampling, since direct-push data such as CPT/SMR bulk conductivity can complement and validate the geochemical sampling performed in the wells screened over the entire thickness of the aquifer (up to 20 m). Fully-screened wells were preferred since sampling could be easily repeated at a preferred depth and the 5 cm (2 inch) diameter wells allow the measurement of multiple parameters with a variety of instruments. Multiple wells with discrete screen intervals were also installed throughout the study area at specific depths to validate the geochemical data obtained previously in long screens. As wells were left open for a few years before geochemical sampling occurred, mixing in the wells with strong vertical hydraulic gradient took place. To minimize the impact of cross-contamination of geochemical parameters with a long screened borehole, wells should be temporary sealed between sampling events.

4.5.2 General applicability of the characterization approach

The characterization approach was based on a combination of complementary field techniques with the emphasis on the acquisition of detailed 1D and 2D indirect hydrogeophysical data that were integrated with direct hydraulic and geochemical data. Besides the choice of methods, the field work sequence was an essential aspect to improve the synergy of the methods used. The characterization program was designed to follow a “coarse to fine” approach to minimize the number of investigated sites while maximizing the quality, extent and significance of the acquired data. This approach is intended to be time and cost efficient as well as adaptable to any type of hydrogeological environment and scale of study. For studies located in a different hydrogeological context, fractured rock aquifers for instance, geophysical methods better adapted to the type of geological environment could be used, keeping the same fundamental objectives of identifying key zones, map the subsurface and the aquifer base and optimize well locations and borehole surveys. For example, a very similar approach was used at the regional scale to define hydrogeological conditions of a 9 000 km^2 fractured rock aquifer system (Lefebvre et al. 2011; Carrier et al. 2013). For this regional study, high-resolution seismic surveys (Pugin et al. 2009) were used to delineate sediment structures and thickness (instead of GPR), Time Domain Electromagnetic (TDEM) surveys helped identify material types (instead of ERT) and CPT/SMR soundings provided deep profiles of material types of up to 50 m thickness in this setting having thick accumulations of marine clay.

4.5.3 General data integration approach, advantages of an integrated study and challenges

Integrated analysis and interpretation of data provided many benefits compared to the independent analysis of individual measurements. Whereas a single measurement generally provides only part of the answer, the combination of multiple measurements provides a better assessment of conditions or a verification of hypotheses, which was the case for many aquifer conditions in the study area: aquifer thickness, groundwater flowpaths, exchanges between the rock and granular aquifers, spatial extent and concentration of the leachate plume, location of recharge and discharge zones, links between the granular aquifer and streams, and locations of leachate discharge in creeks. The sequence of data collection, especially the combination of GPR and CPT/SMR, provided early indications of the general hydrogeological conditions at the site as well as a rough delineation of the leachate plume, with very little data processing. The results of these simple and fast data integration and interpretation steps allowed the improved planning of forthcoming field surveys. After completion of the characterization, the integrated data served as a basis for the development of a representative conceptual model and of a detailed numerical groundwater flow model. The integrated characterization approach made possible the definition of the geometry, internal structure, heterogeneous material distribution and groundwater composition of a granular aquifer in the sub-watershed surrounding a former unlined landfill.

Integration of hydrogeological methods is a subject in itself. Quantitative hydro-geophysical data integration and interpolation is usually performed via theoretical and empirical petrophysical relations or by the application of statistical methods. Each step of the characterization and data integration workflow also involved the integration of various types of data or previously estimated aquifer conditions (Figure 4.5). Although it is not the topic of this paper, “integration” of all members of the multidisciplinary team was as much a challenge as the integration of the different sources of data. Much more work was involved in data integration as part of the other components of the study mentioned in Figure 4.5.

4.6 Conclusions

This paper presented a workflow for field characterization and data integration, adapted to the definition of aquifer heterogeneity at the subwatershed scale, and providing relevant information for contaminant studies and solute transport modeling. The focus of this paper is on the initial data integration steps of the various independent data leading to the development of a hydrogeological conceptual model considering all components of an aquifer system and its heterogeneities.

The multidisciplinary characterization program was carried out on a shallow semi-confined aquifer located in a 12 km^2 sub-watershed enclosing a decommissioned landfill. The proposed methodology involves the integration of multiple proven field methods that can be used efficiently at relatively low operational cost. The characterization emphasis was put on the acquisition of indirect continuous (laterally and vertically) hydrogeophysical data and limited high-resolution direct data that were used to constrain and correlate the indirect data. Data collection was done in a logical sequence to maximize the level of data acquisition while minimizing the number of field sites. Three types of field methods were combined: 1) surface geophysics; 2) direct-push methods with a geotechnical drilling rig; and 3) borehole operations, including high-resolution hydraulic tests and geochemical sampling.

The combination and sequence of field methods was found to be important. In the study area, ground penetrating radar (GPR) very effectively delineated sedimentary structures and aquifer thickness. Electrical resistivity tomography (ERT) complemented GPR data through the delineation of zones of different electrical conductivity that could be related to material or fluid changes. Direct push soundings with cone penetration tests (CPT) and soil moisture resistivity (SMR) allowed the detailed 1D vertical profile definition of material types and also the identification of the presence of leachate. Fully-screened observation wells installed by direct push allowed the measurement of hydraulic properties over the entire thickness of the aquifer using packer slug tests, flowmeter profiles under pumping, new vertical interference slug tests providing vertical hydraulic conductivity measurements, and tomographic slug tests. These wells also allowed the definition of geochemical conditions.

The simultaneous use of GPR, ERT and CPT/SMR data provided unequivocal conclusions about the aquifer geometry, hydrostratigraphy and the presence of natural groundwater or leachate. Aquifer limits, materials and conditions, as well as the extent of the leachate plume could be defined on the basis of direct data as well as indirect indicators (groundwater temperature, conductivity, pH and dissolved oxygen; the relative topography and vertical hydraulic gradients; fluid conductivity and TDS derived from CPT/SMR). The integration of multiple data sources revealed in great details: 1) the aquifer structure (aquifer boundaries, stratigraphy and hydrofacies); 2) the leachate plume extent; 3) the spatial distribution of the main hydrogeological contexts as well as groundwater flow patterns (recharge, intermediate, discharge areas) and; 4) groundwater geochemical conditions found throughout the aquifer. These simple integration steps are a prerequisite for quantitative data integration steps such as: development of a learning machine workflow to recognize hydrofacies and predict *K*, Multiple Points Geostatistical simulations to define a heterogeneous *K* field, and studies of geochemical processes and numerical modeling to assess natural attenuation. Moreover, the initial conceptual model developed possesses the required information to develop a groundwater flow and mass transport numerical model.

Relevant lessons were learned from the study. Such multi-disciplinary studies greatly benefit from the strict documentation of data acquisition and processing protocols to allow multiple users to appraise the data applicability to different purposes. Combined hydrogeophysical methods provide the possibility to fully interpret aquifer conditions and estimate hydraulic conductivity (*K*), even though relations between geophysical responses and *K* are complex. Such relations can be established if hydrogeophysical parameters and *K* values are both available over the full range of materials found in the aquifer. This was achieved in the study by a dense spatial distribution of hydrogeophysical measurements, the use of continuous 2D and 1D hydrogeophysical surveys, but also by fully-screened observation wells collocated with CPT/SMR soundings that provided high-resolution *K* values at the same vertical resolution as CPT/SMR soundings.

Although numerous studies have addressed aquifer heterogeneity and its impact on contaminant transport, the present study involved novel aspects. First, in terms of scale, studies of aquifer heterogeneity, using either hydraulic or hydrogeophysical approaches, have mostly focussed thus far on sites having quite restricted scales (often less than 50 m x 50 m). The present study was carried out at a "regional" scale (a few km^2) to encompass the contamination source zones as well as receptors. Such a scale is needed for numerical models supporting contaminated site management. There is thus a need for characterization and data integration approaches allowing the definition of aquifer heterogeneity at regional scales. Second, numerous studies of aquifer heterogeneity are based on synthetic data, often only in 2D vertical sections. This study aimed to produce the actual data needed to represent aquifer heterogeneity for a real site at a regional scale. This had important implications on the characterization approach followed to achieve the study objectives, especially the focus on indirect hydrogeophysical characterization methods. Third, as other studies have done before, the approach of this study involved multiple hydrogeophysical methods, as well as other data types (geological, hydraulic, geochemical). However, an original aspect of the study is that data from these methods were not combined after completion of the characterization; instead a role for each method and a sequence of hydrogeophysical data acquisition was planned in order to fully benefit from the complementary information provided by the different surveys. Fourth, the use of fully-screened boreholes allowed original hydraulic tests to be carried out, which provided direct measurements of *K* over all aquifer material types. The hydraulic methods used included packer slug tests, flowmeter profiles, newly developed vertical interference tests and tomographic slug tests. Furthermore, the colocation of fully-screened boreholes with CPT/SMR soundings allowed the integration of these indirect data with direct hydraulic measurements of *K*, which facilitates the development of hydrofacies and *K* prediction schemes using CPT/SMR data.

The data collection carried out as part of the study allows the understanding of a complex groundwater flow system, which guides further studies on groundwater hydraulics and natural attenuation of the leachate plume. This characterization approach was found to be time and cost efficient and could be applied to other types of hydrogeological environments and geographical scales of study using field methods adapted to local conditions. This study demonstrates how an integrated aquifer characterization can provide the basis to understand the complexity of contaminated sites and ensure sound environmental management for the protection of the groundwater resource.

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Chapitre 5

Direct and Indirect Indicators of the Spatial Distribution of Recharge and Discharge in a Shallow Granular Aquifer Affected by a Leachate Plume

TITRE FRANÇAIS :

Indicateurs directs et indirects de la distribution spatiale de la recharge et de l'émergence dans un aquifère granulaire superficiel affecté par un panache de lixiviat.

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Laurie Tremblay a fait l'acquisition, le traitement et l'interprétation des données, ainsi que l'écriture de la version initiale de l'article et la réalisation des figures et des tableaux.

Patrick Brunet a acquis et traité les données de profils de charge hydraulique de haute résolution dans les puits et les données d'hydrogrammes de puits 2010-2011 et a mis à jour le bilan hydrologique pour 2010-2011.

René Lefebvre a supervisé les travaux et a participé aux traitements et à l'interprétation des résultats ainsi qu'à la révision de la version initiale de l'article.

Vincent Cloutier a encadré l'étudiante par rapport aux aspects géochimiques et a révisé la version préliminaire de l'article.

REVUE CONSIDÉRÉE POUR LA PUBLICATION :

L’article sera soumis après sa révision qui tiendra compte des commentaires du jury de la thèse doctorale. Les revues suivantes sont considérées pour la soumission de l’article : Journal of Hydrology, Hydrological Processes et Journal of Environmental Quality.

RÉSUMÉ

La compréhension des processus de recharge et d’émergence est essentielle pour la gestion environnementale et l’évaluation des risques de pollution de l’eau souterraine ainsi que de l’eau de surface reliée hydrauliquement à un aquifère. Cette étude démontre l’utilité de mesurer plusieurs indicateurs superficiels afin de valider un modèle hydrogéologique conceptuel et pour clarifier les conditions limites d’un aquifère, particulièrement la recharge. L’étude porte sur un sous-bassin versant de 12 km^2 où un ancien site d’enfouissement repose sur un aquifère granulaire hétérogène peu profond. Les indicateurs considérés sont la topographie, de multiples propriétés physiques et géochimiques des sols, la géochimie de l’eau souterraine à faible profondeur et celle de l’eau de surface, les types de sédiments sous les cours d’eau, les charges hydrauliques mesurées dans des puits superficiels en bordure des cours d’eau et des mesures de débit dans les cours d’eau à l’étiage. Ces indicateurs sont combinés à des données isotopiques de l’eau souterraine (^{18}O, ^{2}H, ^{13}C, ^{14}C, ^{3}H/^{3}He), des profils physicochimiques dans les puits et des données hydrauliques de haute résolution dans les puits d’observation pour établir les taux de recharge, l’écoulement de l’eau souterraine et les zone d’émergence, ainsi que leur impact sur la migration et l’atténuation naturelle d’un panache de lixiviat. Les résultats démontrent une grande variabilité des patrons de recharge sur la petite zone investiguée (de près de 0 à 420 mm/an). Cette variabilité est surtout influencée par la topographie, l’hétérogénéité de l’aquifère, la nature des matériaux présents en profondeur et les directions verticales d’écoulement de l’eau souterraine. Des zones locales de résurgence de lixiviat dans les cours d’eau ont été identifiées et le flux de masse a été évalué à partir de multiples traceurs (chlorure, sodium, potassium, ammonium, carbone organique dissous, etc.). Ces résultats ont été intégrés dans le modèle conceptuel du système aquifère et ils fournissent des conditions représentatives qui pourront servir de base au développement d’un modèle numérique de l’écoulement et de transport de masse, permettant ainsi de guider des recommandations par rapport à l’utilisation du territoire et à la gestion du risque environnemental relié à l’ancien site d’enfouissement sanitaire.

MOTS CLÉS : validation d’un modèle conceptuel, estimation de la recharge, aquifère granulaire, hétérogénéité, site d’enfouissement.

ORIGINALITÉ ET PERTINENCE:

Plusieurs études ont abordé l’évaluation de la recharge dans une perspective de compréhension de l’écoulement et de la vulnérabilité des aquifères. Quelques études ont aussi abordé l’importance de définir les zones locales d’émergence d’un panache de contamination dans les cours d’eau et d’en évaluer les flux de concentration afin de minimiser l’impact de la contamination sur les milieux récepteurs. Enfin, d’autres études ont proposé l’utilisation de traceurs environnementaux et de la datation de l’eau souterraine afin de mieux comprendre les problématiques mentionnées ainsi que les temps de résidence de l’eau souterraine. Par contre, aucune étude, à la connaissance des auteurs, n’a combiné l’étude de la recharge et de l’émergence de l’eau souterraine et de ses impacts sur la migration et l’atténuation d’un panache de lixiviat géré par atténuation naturelle, à l’aide d’indicateurs mesurés dans la partie supérieure d’un aquifère. Selon les auteurs, les approches décrites dans l’article ont les aspects novateurs et originaux suivants par rapport aux études antérieures réalisées sur ce sujet:

Échelle d’étude

Les études sur l’évaluation de la recharge ont mis l’emphase sur des sites régionaux en relation avec l’évaluation de la ressource en eau et la vulnérabilité des aquifères sur de grandes surfaces géographiques (plus de 50 km x 50 km). L’étude documentée dans cet article englobe une plus petite zone d’étude (de quelques km^2) requise pour considérer les zones sources de contamination et les milieux récepteurs. À ce niveau d’étude, la variabilité de la recharge peut grandement influencer l’écoulement de l’eau souterraine. Cette échelle permet de considérer les mécanismes "fins" qui contrôlent la recharge et qui ont été peu étudiés. Cette échelle d’étude est aussi requise pour la gestion des sites contaminés et pour offrir la possibilité de développer des modèles numériques nécessaires pour supporter les décisions reliées à cette gestion.

Utilisation originale d’indicateurs superficiels

L’originalité principale de cet article repose sur l’utilisation et l’intégration d’indicateurs, mesurés à faible profondeur à travers la zone d’étude, qui permettent de mieux définir certaines conditions du système d’écoulement. Ces indicateurs sont basés sur des mesures directes et indirectes, quantitatives et qualitatives, et incluent la topographie, plusieurs mesures physiques et chimiques sur les sols, la géochimie de l'eau souterraine à la surface de la nappe, des mesures de débitmètre dans les cours d'eau à l'étiage, des mesures dans des piézomètres installés en bordure des cours d'eau, etc. Les travaux réalisés dans le cadre du projet doctoral montrent la complémentarité de ces indicateurs par rapport aux données hydrogéologiques conventionnelles ne fournissant que des informations limitées sur certains aspects importants du système d'écoulement. Ces indicateurs sont aussi importants car ils fournissent des mesures indépendantes permettant de vérifier la représentativité et de compléter un modèle conceptuel initial du système aquifère de la région d'étude.

Intégration originale des divers indicateurs pour la compréhension d'un système d'écoulement

Les indicateurs sont combinés avec des données isotopiques de l'eau souterraine, des profils physico-chimiques et des mesures hydrauliques de haute résolution faites dans des puits d'observation. Les données collectées suite aux mesures effectuées dans les puits permettent de quantifier les temps de résidence de l'eau ainsi que les directions et les vitesses d'écoulement. Ensemble, ces données permettent de préciser le modèle conceptuel d'un système aquifère granulaire peu profond déjà développé sur la base d'une caractérisation hydrogéologique intégrée. Ces indicateurs contribuent aussi à définir la distribution spatiale des zones de recharge et d'émergence de l'eau souterraine, ainsi que de comprendre le lien entre les eaux de surface et l'eau souterraine, de saisir l'importance de la recharge à l'échelle très locale, ainsi que son influence sur les temps de résidence de l'eau souterraine. Ces mesures, faites sur le terrain et en laboratoire, sont simples, efficaces et peu coûteuses.

Validation du modèle conceptuel défini sur la base de la caractérisation intégrée

Le modèle conceptuel ainsi raffiné explique les conditions d'écoulement et les voies de migration d'un panache de lixiviat émis par un ancien site d'enfouissement. Les eaux de surface sur le site étudié sont le principal milieu récepteur potentiel des contaminants migrant dans le panache de lixiviat. Les indicateurs et traceurs géochimiques utilisés permettent de mieux définir les liens entre l'aquifère et les cours d'eau ainsi que de distinguer les apports de contaminants provenant de l'eau souterraine de ceux provenant des eaux de lixiviation traitées et rejetées directement dans les cours d’eau. Les indicateurs et traceurs donnent ainsi une évaluation plus précise des zones à risque dans les cours d’eau ainsi que de la provenance souterraine ou de surface des produits dégradant la qualité des eaux de surface. Le portrait décrit par ces indicateurs permettra de soutenir des recommandations faites au gestionnaire de l’ancien site d’enfouissement par rapport à l’utilisation du territoire pour la gestion des risques associés à la migration et à l’atténuation naturelle du panache de lixiviat.

Estimation des taux de recharge en combinant 4 méthodes adaptées à la zone d’étude

Quatre méthodes ont été utilisées pour estimer la recharge : 1) hydrogrammes de puits (niveaux d’eau); 2) bilan hydrique basé sur les données météorologiques, 3) mesures isotopiques de l’âge de l’eau souterraine, et 4) mesures de débits à l’étiage dans les cours d’eau. Ces méthodes ont été sélectionnées suite à l’analyse qualitative et semi-quantitative des sols et des hydrogrammes de puits. En combinant ces méthodes adaptées à la région d’étude, il a été possible un portrait représentatif de la distribution spatiale du taux de recharge et des facteurs qui la contrôlent.

Pertinence de ces données pour des études ultérieures pour la prédiction du devenir du panache de lixiviat

Les données acquises ainsi que leur utilisation pour valider le modèle conceptuel permettent d’assurer la protection de l’environnement à long terme, grâce à la compréhension du système d’écoulement. Elles ont aussi permis d’identifier des traceurs géochimiques clés pour distinguer les eaux provenant du rejet dans les eaux de surface du lixiviat traité par rapport aux eaux de lixiviation qui émergent dans les cours d’eau suite à leur migration dans l’eau souterraine. La compréhension des processus de recharge dans la zone d’étude démontre la grande variabilité des taux de recharge, et leur impact sur l’écoulement de l’eau souterraine et sur les conditions géochimiques qui influencent l’atténuation naturelle du lixiviat. Il est donc essentiel de considérer la variabilité de la distribution spatiale de la recharge dans le modèle numérique d’écoulement représentant la région d’étude. De plus, ces indicateurs pourront être utilisés pour prédire les effets des changements climatiques sur la recharge et sur la migration et l’atténuation du panache de lixiviat, ou encore les impacts de changements dans l’utilisation du territoire dans la région d’étude.

ABSTRACT

The understanding of recharge and discharge processes is needed to manage water resources and evaluate pollution risk to groundwater as well as surface water connected to aquifers. This study demonstrates the relevance of measuring multiple simple superficial indicators to validate a hydrogeological conceptual model and clarify aquifer boundary conditions, especially groundwater recharge. The study focuses on a 12 km^2 sub-watershed within which a decommissioned landfill is located over a shallow semi-confined heterogeneous granular aquifer. The indicators considered include topography, multiple physical and chemical soil properties, groundwater and surface water chemistry, stream bed sediments, hydraulic heads measured in shallow wells on stream banks, and flowmeter measurements in streams at baseflow. These shallow indicators are combined with groundwater isotopic data (^{18}O, ^{2}H, ^{13}C/^{14}C, ^{3}H/^{3}He) and high-resolution hydraulic heads measured in fully screened observation wells to assess recharge rates, groundwater flow directions, discharge rates to streams as well as their impacts on the migration and natural attenuation of a leachate plume. Results show a high variability of recharge rates over the small area investigated (from near 0 to 420 mm/y). The variability is mostly influenced by topography, aquifer heterogeneity, subsurface material types and vertical groundwater flow paths. Local leachate discharge zones into streams were also identified and mass discharge was estimated with multiple tracers (chloride, sodium, potassium, ammonium, dissolved organic carbon, etc.). Results are integrated into the aquifer conceptual model, and provide firm indication of representative conditions needed for the development of a numerical model of groundwater flow and mass transport and will guide recommendations for land use and risk management of the decommissioned landfill.

KEYWORDS: conceptual model validation, recharge estimation, granular aquifer, heterogeneity, landfill leachate

5.1 Introduction

Before the development of engineered landfills, abandoned gravel pits, frequently part of unconfined granular aquifers, often served as waste disposal sites (Roy 1994). These sites, located on permeable material, did not have any liner to prevent precipitation infiltration into the waste, providing leachate which rapidly reached the water table. Therefore, numerous decommissioned landfills throughout the world rely on natural attenuation processes to reduce the mass of contaminants to less harmful forms and concentrations within aquifers through dilution, dispersion, sorption and microbial degradation. Since certain chemical compounds are not subject to some of the natural attenuation processes or their concentrations might not decreased to acceptable levels, contaminant mass emissions from unlined waste disposal sites can pose a threat to ground and surface water over decades to centuries (Milosevic et al. 2012; Bjerg et al. 2011, 2003; Cozzarelli et al. 2011; Lorah et al. 2009; Park et al. 2008; Kjeldsen et al. 2002; Christensen et al. 2001, 1994).

The responsible management of groundwater and surface water requires knowledge of the shallow subsurface, groundwater occurrence, flow dynamics, aquifer connectivity to surface water, contamination sources from land surface, solute transport and the representation of these processes in detailed conceptual and numerical models (Milosevic et al. 2012; Nordstrom 2012; Schmelzbach et al. 2011; Konikow 2010; Janković et al. 2009). While it is well accepted by the scientific community that the spatial variability in observed contaminant concentrations results from geological and geochemical heterogeneities (Anderson and McCray 2011; Allen-King et al. 2010; USEPA 2008, 2007; de Marsily et al. 2005), groundwater recharge and discharge characteristics must also be considered since they exert dominant controls on flow and groundwater chemistry (Stephens 2009; Illman 2008; USEPA 2008, 2007; Bredehoeft 2007; Bredehoeft 2005; Healy and Cook, 2002; Kim 2002; Arnold and Allen 1999). Furthermore, fresh oxidizing water infiltrating through soil can contribute to leachate plume attenuation processes (Scholl et al. 2006; Christensen et al. 2001, 1994). It is thus important to obtain detailed information on existing conditions in aquifer systems to support the development of conceptual models that are further constrained by independent data and tested with numerical simulations.

In any assessment of groundwater supply or aquifer vulnerability, groundwater recharge is one of the most important factors to be considered. While representative recharge values are also a key parameter in any groundwater flow numerical model (Sanford 2002), groundwater recharge is still one of the most difficult aspects of hydrogeological systems to assess (Healy 2010). Recharge is defined as the proportion of precipitation transiting through the unsaturated zone that reaches the water table and contributes to groundwater flow (Freeze and Cherry 1979). Recharge varies widely in space and time, making it difficult to measure directly (Healy 2010; Healy and

Cook 2002; de Vries and Simmers 2001). In general, the interaction of climate, geology (aquifer heterogeneity), geomorphology (topography, soil condition, vegetation) and land use controls recharge processes (Atchley and Maxwell 2011; Healy 2010; Scanlon et al. 2002; Arnold 2000; Arnold and Allen 1999).

At the local scale, recharge and runoff generation can be induced by minor topographic variations (Frei 2010). Areas of high recharge are often associated with areas of high aquifer vulnerability to contamination and, in general, the likelihood for contaminant movement to the water table increases as the rate of recharge increases. Methods used to assess aquifer vulnerability often use recharge as an important indicator (Aller et al. 1987). Moreover, at the local scale, focused recharge and preferential flow can lead to fast contaminant migration though the unsaturated zone to underlying aquifers. In that sense, unconfined and shallow granular aquifers will be more vulnerable to contamination, and the migration paths of that contamination will be related to recharge patterns and rates, as they will influence groundwater flow and water table configurations. Since spatial variability is important for assessing aquifer vulnerability to contamination, methods that provide point estimates of recharge have to be considered (Healy 2010). Moreover, it is also necessary to identify the main recharge and discharge areas as well as contaminant discharge zones in unconfined aquifers. According to Milosevic et al. (2012), a systematic approach for the identification of hot spots (areas with high concentrations of leachate indicators), contaminant transport paths and discharge zones has to be part of the characterization of landfill sites in heterogeneous geological settings. To evaluate the pollution risk to the aquifer and connected surface water, both recharge patterns and groundwater-surface water interactions need to be considered within the assessment of groundwater flow dynamics and contaminant migration (Molosevic et al. 2012; Fleckenstein et al. 2010).

Several studies on recharge estimation have demonstrated the usefulness of integrating various data types to develop representative conceptual models of recharge processes and assess the uncertainty of recharge estimates. Recharge estimation approaches include the water table fluctuation method (WTM), water balance methods, baseflow discharge and many more (Healy 2010; Coes 2007; Delin et al. 2007; Scanlon et al. 2002; de Vries et al. 2002; Arnold et al. 2000). Moreover, many studies, carried out in different hydrogeological contexts, have demonstrated the usefulness of geochemical tracers to define recharge mechanisms and rates when combined with conceptual models based on field data (Chen 2011; Choi et al. 2010; Healy 2010; Stadler et al. 2010; Blasch 2007; Delin et al. 2007; Scholl et al. 2006; Macpherson and Sophocleous 2004; Böhlke 2002; Scanlon et al. 2002; Solomon et al. 1993). However, many of these studies were limited to the regional scale (> 50 km^2) and focus mainly on arid or semi-arid regions (Healy 2010; Stadler et al. 2010; Houston 2002). Studies available on regions with temperate climates were also mostly done at a regional scale (> 50 km^2), and related to either aquifer sustainability or vulnerability (Chen et al. 2011; Choi 2010; Coes et al. 2007; Croteau 2006).

Few studies have evaluated the impact of recharge patterns on contaminant plume migration and attenuation (Scholl et al. 2006) or have evaluated mass discharge from landfill leachate into local streams (Milosevic 2012; Troldborg 2012). There is thus a need in contaminant hydrogeology for the elaboration of systematic approaches leading to the development of representative conceptual models, based on a wide range of characterization data, acquired at a scale encompassing source zones and receptors, to facilitate an understanding of the hydrogeological system and propose solutions to land use and aquifer protection. The conceptual model should be developed at the beginning of a study as it can be revised and adjusted as additional data and analyses provide constraints on the hydrogeological system (Bredehoeft and Konikow 2012; Healy 2010; Bredehoeft 2005; Zheng and Bennett 2002). Scientists from diverse hydrogeological research specialties (recharge quantification, groundwater quality, wellhead protection areas, modeling of solute transport, etc.) advocate for the development of a conceptual model based on actual field data prior to efforts aiming to resolve issues related to groundwater management and protection (Nordstrom 2012; Sousa et al. 2012; Sudicky and Illman 2011; Clement 2010; Healy 2010; Crumbling et al. 2003). This paper aims at making a contribution to such developments, by investigating multiple superficial indicators that can be used to constrain a conceptual model that would support contaminated site management and risk assessment to receptors. The investigation addresses the following contaminated site characterization issues: identification of discharge zones and hydraulic connectivity of streams (as the main receptor for leachate) with the granular aquifer, assessment of indicators defining recharge or discharge conditions within the aquifer, delineation of patterns of groundwater flow and leachate migration, and definition of physico-chemical conditions related to soil and shallow groundwater. All of these indicators help define what controls the emission of contaminants, groundwater flow paths controlling the plume migration, the relation between surface and groundwater and the risk of stream water quality degradation due to leachate discharge. These processes are needed to understand the system dynamics and validate an existing conceptual model developed on the basis of an extensive integrated field characterization (Tremblay et al. 2013).

The present study thus aimed to develop an approach using shallow indicators that could relate surface processes (infiltration, runoff, hypodermic flow) to subsurface processes that are controlling leachate migration and its discharge into surface water. This study assesses the relevance of measuring, in the field, multiple simple superficial indicators, to validate a hydrogeological conceptual model and clarify boundary conditions that need to be defined for groundwater numerical flow models, especially the spatial distribution of recharge. Data collection is very site specific and is one of the most important steps of groundwater assessment. The work described in this paper represents the second phase of a multidisciplinary integrated field characterization program and was derived from the first phase of field work (Tremblay et al. 2013). Results from such a study can offer guidance to contaminated site managers in relation to land use and risk management of leachate plumes.

5.2 Background

5.2.1 Study context and location

The study area covers 12 km^2 and is located 35 km south of Quebec City, Canada (Figure 5.1). In the center of the study area, two municipal landfills, one unlined and one engineered, are located on a 1 km^2 property managed by a public organization ("Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière", RIGDCC). The engineered landfill is in operation and meets current waste management standards. The unlined landfill is decommissioned which was in operation between 1974 and 1997, during which 9 million tonnes of waste from varied sources were disposed directly over permeable sands. From 1993 to 1997, the waste piles were reshaped and sealed with an impermeable cover (RIGDCC 2013). Prior to the decommissioned landfill closure, landfill leachate was detected in the surrounding creeks (Gauthier 1993a). Since its closure, the unlined landfill is solely managed by natural attenuation, although no studies have thus far verified the effectiveness of this management method.

In 2006, a study was initiated to investigate the effectiveness of the natural attenuation of the leachate plume emitted by the decommissioned landfill. In 2007, an integrated multidisciplinary field characterization program was carried out, at a scale encompassing source zones and receptors and specific to the hydrogeological setting in place (Tremblay et al. 2013). Characterization methods included: 1) surface geophysics including a) ground penetrating radar (GPR) and b) electrical resistivity tomography (ERT); 2) direct-push methods including a) cone penetration tests (CPT) with soil moisture resistivity (SMR), b) soil sampling and c) installation of fully-screened observation wells over the entire thickness of the granular aquifer; 3) borehole operations including a) flowmeter measurements while pumping, b) high vertical resolution (15 cm) packer slug tests, and c) multilevel measurements of geochemical parameters and groundwater sampling. The integration of the field data allowed the definition of aquifer boundaries and thickness, geological heterogeneity of the aquifer material and the extent of the leachate plume.

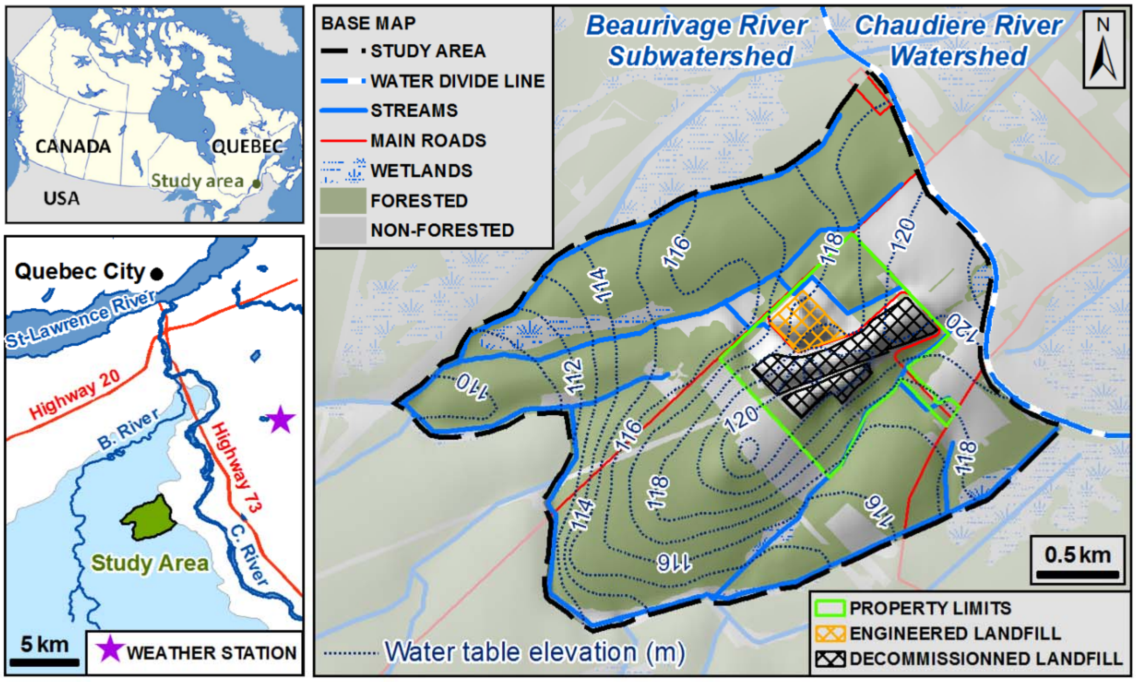


Figure 5.1 : Location and general context of the study area

Lower left map: B. River stands for Beaurivage River and C. River for Chaudière River; blue shaded area illustrates the Beaurivage subwatershed within which the study area is located; the star indicates the location of the weather station used in this study (modified from Tremblay et al. 2013)

On the basis of the integrated characterization data, a preliminary but detailed conceptual model was established (Tremblay et al. 2013). The initial conceptual model illustrated the high degree of heterogeneity of the aquifer material, the great variability in elevation of the aquifer-aquitard boundaries and the lack of continuity of certain geological units. A hot spot location with higher concentration of leachate was also identified (Tremblay et al. 2013). The characterization showed no concentrations of dissolved ions in groundwater exceeding standards (MDDEP 2012) outside the landfill property limits of 1 km^2. Hydrogeological contexts (recharge, discharge) were also defined on the basis of multiple conditions (groundwater temperature, conductivity, pH and dissolved oxygen; the relative topography and vertical hydraulic gradients; fluid conductivity and total dissolved solid (TDS) derived from cone penetration tests (CPTu) with soil moisture resistivity (SMR)). The goal of this second phase of field work was to further investigate the recharge and discharge zones previously defined and identify possible locations of leachate discharge into streams. This study also aimed to quantify recharge rates and groundwater residence time to help predict the fate of the leachate plume and serve as a basis for the development of a representative groundwater flow model. The hydraulic connectivity between the aquifer and the surrounding stream is not known but will be investigated along with the surface water quality.

5.2.2 Physiological and hydrogeological setting

The study area is easily accessible by primary, secondary and forestry roads (Figure 5.1). The subwatershed covering an area of 12 km^2 was defined on its eastern side by a groundwater divide between the Beaurivage River subwatershed and the Chaudière River watershed. Elsewhere, limits were based on the physiology of the surrounding creeks and rivers (Figure 5.1). The study area is mostly forested but there is also some peatland, agricultural fields and operational gravel pits. The vegetation types covering the area are 60% fir tree, 20% red maple tree, 20 % spruce, and smaller percentages of birch and larch. Zones covered only by coniferous trees have acidic soil leading to the absence of vegetation between trees as well as a shallow root zone (10-30 cm). There are two main agricultural fields within the study area. One on the eastern side of the landfill property grows corn while the one on the western side grows canola. The natural topography of the study area varies from 125 m above sea level (asl) at the groundwater divide line to 109 m asl at the study area surface water outlet (Tremblay et al. 2013). The landfills, reaching 130 m in elevation, changed the local topography. The water table, which follows topography, is quite shallow, and generally found between 0.1 to 2 m below ground surface. Within the subwatershed, groundwater flows from higher elevations found under the former landfill towards local creeks in the southern and northern direction (Figure 5.1). In this relatively flat region, the surface soil is mostly composed of poorly drained podzols of marine origin and sparse zones of organic material and well decomposed black peat (Laplante 1962). As a consequence of flat topography and soil type, an important forestry and agricultural drainage network is in place and, in combination with natural streams, local hydrology strongly influences groundwater flow (Tremblay 2013). The Beauséjour meteorological station located 10 km northeast of the study area (Figure 5.1) provides a full weather database between year 1978 and present and shows that the area received a yearly average of 1200 mm of precipitation.

The aquifer is composed of 10 to 20 m of highly heterogeneous sand interbedded with silt and clayey silt lenses. The aquifer overlies discontinuous layers of glaciomarine silt and till forming the relatively impermeable base of the aquifer. The till deposited by the last glaciation overlies the regional bedrock aquifer composed of fractured shale and sandstone. The Quaternary fluvial, littoral and deltaic sediments making up the aquifer were deposited after the last glaciation (Bolduc 2003). The unconsolidated material originated from sea level changes during the regression of the Champlain Sea at the former Chaudière River outlet. At that time, the system was controlled by sea tides and currents, followed by fluvial erosion and organic sedimentation. This littoral depositional environment is responsible for the high heterogeneity of the aquifer material (Parent and Occhietti 1988) as well as naturally occurring anoxic and oxidizing zones that influence the natural geochemical composition of groundwater (Tremblay et al. 2008). Added to this complex environment, is the presence of a leachate plume emitted by the decommissioned landfill which undergoes important geochemical transformations along its path (Tremblay et al. 2008).

5.3 Methodology

5.3.1 General approach for data collection

The study focuses on a 12 km^2 subwatershed within which a decommissioned landfill is located over a shallow heterogeneous granular aquifer. Multiple indicators were investigated to understand recharge patterns, surface and subsurface interaction in relation to groundwater flow, plume migration and attenuation as well as leachate discharge zones into local streams. Multiple techniques considering climatic data, surface water, the saturated and unsaturated zones and groundwater tracers were combined to evaluate recharge patterns over the study area. The indicators considered included topography, multiple physical and chemical soil properties, groundwater and surface water chemistry, stream bed sediments, hydraulic heads and gradients in shallow wells on stream banks, flowmeter measurements in streams at baseflow, and other indicators. These indicators were combined with groundwater isotopic data (^{18}O, ^{2}H, ^{13}C/^{14}C, ^{3}H/^{3}He) and high-resolution hydraulic head measurements in fully screened wells to estimate recharge rates, groundwater flow directions and discharge rates as well as their impacts on the migration and attenuation of a leachate plume. As a first step, a qualitative data analysis was done for certain parameters fitting these two conditions: 1) a quantitative estimate was difficult to obtain through field measurements and 2) typical values were available in the literature. Secondly, based on the qualitative interpretation, logical locations were selected for quantitative measurements with adapted techniques. This methodology had the objective to minimize the number of samples and survey locations requiring lengthy and more costly analyses and to maximize the spatial distribution of reliable data. This methodology was efficient to get a good understanding of the mechanisms controlling the hydraulics of the aquifer system and allowed to focus on the most important processes.

5.3.2 Field investigation and laboratory analyses of surficial sediments

First, a Quaternary geology map was completed in detail at an appropriate scale of 1:5 000 for the study area based on aerial photographs and field investigations. Field investigations involved descriptions of sediments collected with a hand auger over a 0 to 3 m depth interval. These surveys included numerous points along stream beds (Figure 5.2a), identification of rock and till outcrops at multiple locations and topographic surveys (Tremblay et al. 2013). Drive point piezometers were installed in different stream bed contexts along stream banks to monitor hydraulic heads and infer aquifer hydraulic connectivity to streams.

Next, the Quaternary map was combined with a soil map at a scale of 1: 20 000 (MAPAQ 2008; Laplante 1962) to plan soil sampling and field investigations of the unsaturated zone at locations where the surficial materials were different. The saturated hydraulic conductivity was measured in the field using a Guelph Permeameter (Soilmoisture Equipment Corp. 2005). Figure 5.2a illustrates the location for saturated conductivity measurements at 30 cm depth. Following the interpretation of permeameter tests and the identification of soil types, organic soil was sampled at 9 locations representing different soil profiles to determine the geochemical composition and the possible impact of soils on shallow groundwater (Figure 5.2b). Organic samples were lyophilized, digested and analysed for major and minor elements through ICP-MS by the INRS laboratory in Quebec City. Carbon, sulphur and nitrogen were analyzed directly with a LECO analyser in the lab.

5.3.3 Surface water flow and water chemistry

Stream sections were selected based on the longest straight reach found along each stream and where the channels were free of disturbances. Measurements were also made before and after an incoming stream. At each location, depths in relation to water height were measured every 30 cm along a stream section and flow rates were measured at three strategic water heights. A Flow-Mate 2000 flowmeter (Marsh-McBirmey Inc. 1994) was used for these measurements. All measurements for recharge estimation were done in September 2009, at the lowest baseflow recorded for that year. For mass discharge estimation, flow measurements were made throughout the month of August 2010 while water sampling was carried out (Figure 5.2c). To calculate flow, the trapezoidal method was used for each 30 cm subsection measured along the stream width and the circular conduit method was used where conduits were located (Marsh-McBirmey Inc. 1994).

5.3.4 Well operations

5.3.4.1 Physical measurements in wells

Flowmeter measurement were done in eighteen (18) fully-screened observation wells and packer slug test vertical profiles were carried out at 15-cm intervals into eight (8) fully-screened observation wells installed by direct push (Paradis et al. 2011a). Multilevel hydraulic head measurements were also made to help define the connectivity between hydrofacies identified along profiles. Pressure and temperature loggers were installed in nine (9) wells throughout the study area as well as barometric pressure loggers to correct well hydrographs (Figure 5.2d). Physicochemical profiles were measured in the wells as a first indicator of groundwater geochemical conditions found in the aquifer. The physicochemical parameters (temperature, redox potential (ORP), dissolved oxygen (DO), pH, and conductivity) were recorded with a YSI 556 probe (YSI 2013). The probe was lowered in the well and measurements were taken at every meter. The probe was left stationary from 10 to 30 minutes at the same depth to allow parameters to stabilize before recording measurements. Profiling was carried out during three different seasons over a period of 4 years and results are illustrated in Tremblay et al. (2013). Finally, groundwater geochemical and isotopic analyses were carried out (Figure 5.2d).

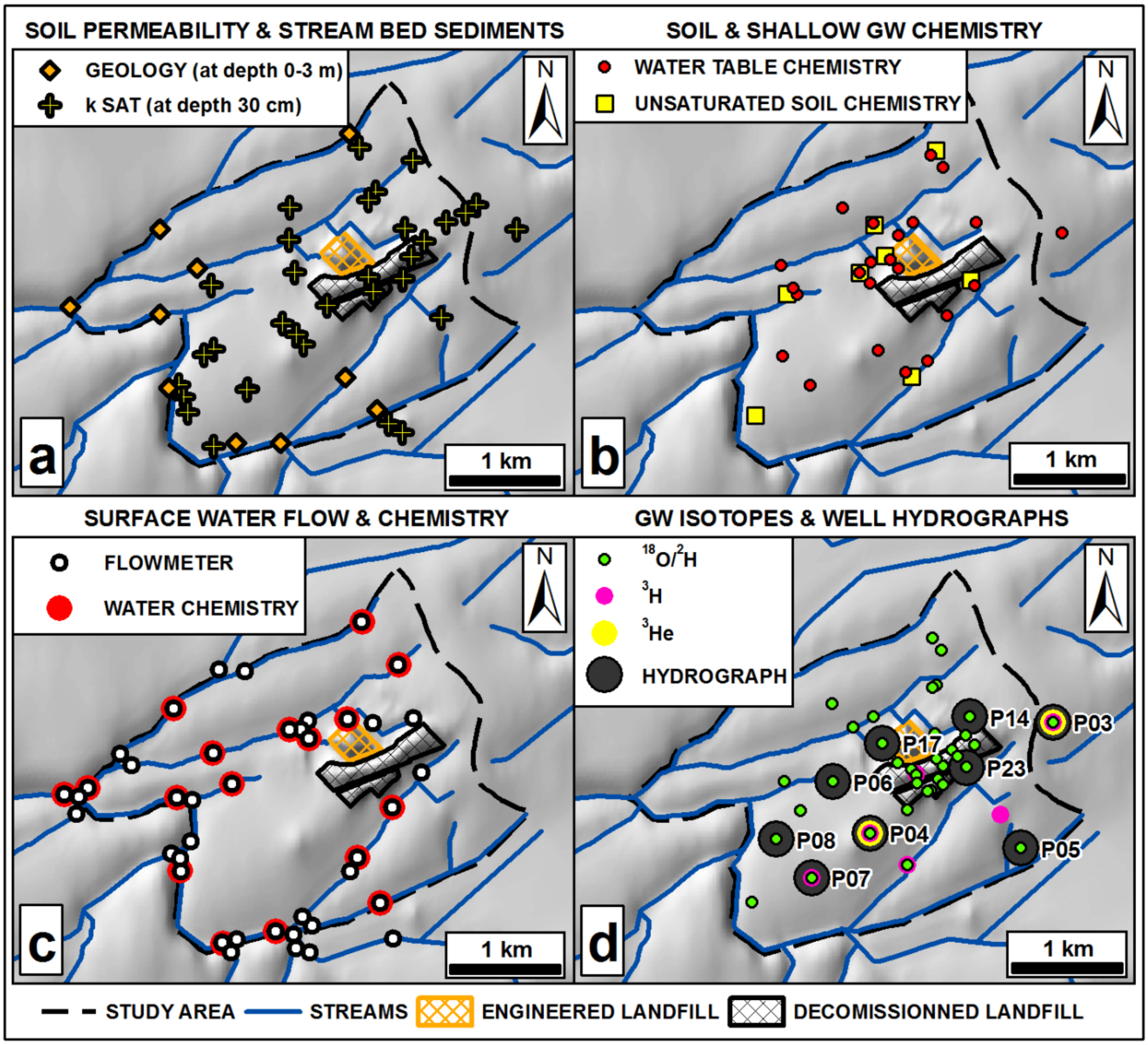


Figure 5.2 : Field work location

Spatial distribution of field measurements of direct and indirect indicators: a) soil physical properties; b) chemistry of soil and shallow groundwater; c) surface water flow and chemistry; and d) well hydrographs and isotopic signature of groundwater (GW)

5.3.4.2 Groundwater and surface water sampling and analyses

Groundwater and surface water sampling

Sixteen (16) water samples were collected in streams (Figure 5.2c) and 120 groundwater samples were collected throughout the study area between 2010 and 2011. Groundwater samples were collected: 1) between packers at shallow depth (2-5 m) in 19 wells screened over the full thickness of the aquifer; and 2) in 10 discrete shallow piezometers (1.5 to 2 m below ground surface). Wells were purged more than three times their total volume before introducing packers and the intervals isolated by packers were also purged over three times the total volume of the water in the packer and riser tubing assembly. Low-flow groundwater sampling was done with a peristaltic pump to preserve sensitive geochemical conditions. Temperature, electrical conductivity, dissolved oxygen (DO), redox potential (ORP) and pH were recorded on site with an YSI 556-MPS multi-parameter probe (YSI 2013). Groundwater was collected when the physicochemical parameters had stabilized (10 min to 30 min), through 45 µm high capacity HDEP filters fitted directly at the outlet of the MFLEX FDA Viton #16 25' tubes. Water was analyzed for 31 soluble metals, fluoride, ammonium, total sulphur, anions (Br, Cl, SO_4, NO_2, NO_3) dissolved organic carbon (DOC), and dissolved inorganic carbon (DIC), chemical oxygen demand and biological oxygen demand on 5 days (BOD_5) as well as for stable (^{18}O, ^{2}H, ^{13}C, ^{15}N, $^{3}He/^{4}He$, ^{20}Ne, ^{21}Ne, ^{22}Ne) and radiogenic (^{14}C, ^{3}H) isotopes. Alkalinity was determined on site by titration with the total alkalinity method (AWWA 1995a). Fe^{2+} and S^{2-} were dosed directly in the field but readings were taken with a spectrophotometer Thermo Spectronic Genesys 10 µv in 1 cm^3 cells in the lab at the end of each day (Voillier et al. 2000, AWWA 1995b). All samples were stored on ice after collection and during transport to the laboratory. Teflon pipe 1/4-1/16 mur and Teflon union straight connectors were cleaned between each sample with oxalic acid 0.4 M and HCl 1%. The pipes were thoroughly rinsed with demineralised water between the uses of each of the products mentioned and, finally, analyses were performed on the demineralised water to confirm the absence of trace elements in the pipes from the cleaning operation. Figure 5.2b shows the location of the shallow groundwater samples considered for the study and Figure 5.2c shows surface water sampling locations. Finally, Figure 5.2d shows the location for isotopic data.

The tritium-helium (^{3}H/^{3}He) groundwater dating method was performed outside the leachate plume extent, since leachate from landfills is known to have elevated concentration of tritium (Bandeira 2008; see also Figures 5.9d and 5.9e). Eight (8) discrete 2.5 cm (1 inch) diameter wells were installed at multiple intervals of 1.5 m within two known recharge zones. These zones were defined on the basis of well hydrographs and vertical flow zones based on vertical hydraulic gradients. Gases for ^{3}H-^{3}He dating of groundwater (He and Ne) were collected using two different methods. The first method used diffusion samplers, a method involving in-situ passive sampling of dissolved gases (Manning et al. 2003; Sanford et al. 1996). Two pieces of approximately 10 cm annealed copper tubing with 0.6 cm outer diameters were attached to silica tubing (also about 10 cm long) using stainless steel wires, to create a seal preventing water from entering the initially air-filled tubing; the outer ends of the copper tube are sealed (Murphy 2011; Murphy et al. 2009). Gases can diffuse across the semi-permeable membrane of the silica tube. Therefore, the partial pressures of gas components dissolved in the groundwater equilibrate with the gas components inside the diffusion sampler. The diffusion samplers were lowered to specific depths into the discrete wells while attached to a rigid ¼ inch diameter tube and were left in the wells for two weeks. Water was pumped from the well every two days to ensure the replenishment of representative groundwater within the interval. Gas samples were collected by removing the diffusion sampler from the water column and immediately pinching off the copper tubes with a customized pinch-off tool, which creates a cold-weld seal that prevents escape of gases (Murphy 2009).The second method involved the direct collection of water in a copper tubing completely sealed from the ambient air while sampling. The copper tube was fixed vertically outside the well and water was flushed for a couple of minutes before the copper tubes were pinched off on both sides (Aeschbach-Hertig 2012).

Laboratory analysis of environmental tracers and isotopes in groundwater

Major and minor elements were analysed at an accredited laboratory named MAXXAM Analytic. Evaluation of ^{2}H/H and ^{18}O/^{16}O ratios in water was made using an on-line gas equilibration system (Gas Bench II) linked via a universal interface (Conflo IV) to an isotope ratio mass spectrometer (DELTA V PLUS from THERMO FINNIGAN MAT) at the Delta Lab of Natural Resources Canada in Quebec City. ^{14}C and ^{13}C analyses were prepared through the Environmental Isotope Laboratory at the University of Waterloo on 1 L groundwater samples and analyses were carried out at the National Isotope Center of the Institute of Geological and Nuclear Sciences Ltd in New-Zealand. Enriched tritium analysis was carried out at the Environmental Isotope Laboratory at the University of Waterloo on 1 L groundwater samples. Water samples were first deionized on a mixed bed ion exchange resin, and underwent electrolytic enrichment followed by liquid scintillation 69 counting (Heemskerk and Johnson, 1998). The average analytical precision is ±1.1 tritium units (T.U.). ^{3}He/^{4}He ratios were measured at two laboratories: 1) Helium isotope analysis was carried out at the Noble Gas Laboratory of the Department of Earth Sciences at the University of Ottawa; neon isotopes were also analyzed, since they are required to quantify non-tritiogenic components of ^{3}He (Schlosser et al. 1989). Samples were analyzed using a Mass Analyzer Products (MAP) 215-50 mass spectrometer connected to a stainless steel vacuum line for cryogenic processing and separation. ^{3}He was analyzed on a channel electron multiplier, and ^{4}He, ^{20}Ne, ^{21}Ne and ^{22}Ne were analyzed on a Faraday Cup. The average analytical precision for Ne and He concentrations is ±10%; 2) At the AORI - The University of Tokyo. Gases were extracted from water offline and then purified in a stainless-steel line following procedures described in Sano et al. (2010). Helium concentration and isotopic ratios were determined using a HELIX SFT mass spectrometer calibrated against the HESJ standard (Matsuda et al. 2002), with a standard reproducibility of 0.03% (Sano et al. 2010).

5.4 Results

5.4.1 Physical properties of soil

The study area is located within the St. Lawrence lowlands where the topography is frequently interrupted by sand or gravel bands, bedrock outcrops and numerous depressions covered by peat. These features create micro-topographic environments that restrict water flow and drainage (Laplante 1962). The term soil is defined as the portion of the regolith that is biochemically weathered under climate influence. It contains mineral matter and a higher proportion of organic material which constitutes a natural environment for plant growth (Brady 1990). In this paper, the term soil will refer to the material composing the unsaturated zone varying in thickness from 0.1 m to 1 m. The nature of soil material and its thickness within the unsaturated zone could affect groundwater recharge by restricting or preventing water from reaching the water table, and could influence groundwater chemistry and flow at the same time. At their study site in

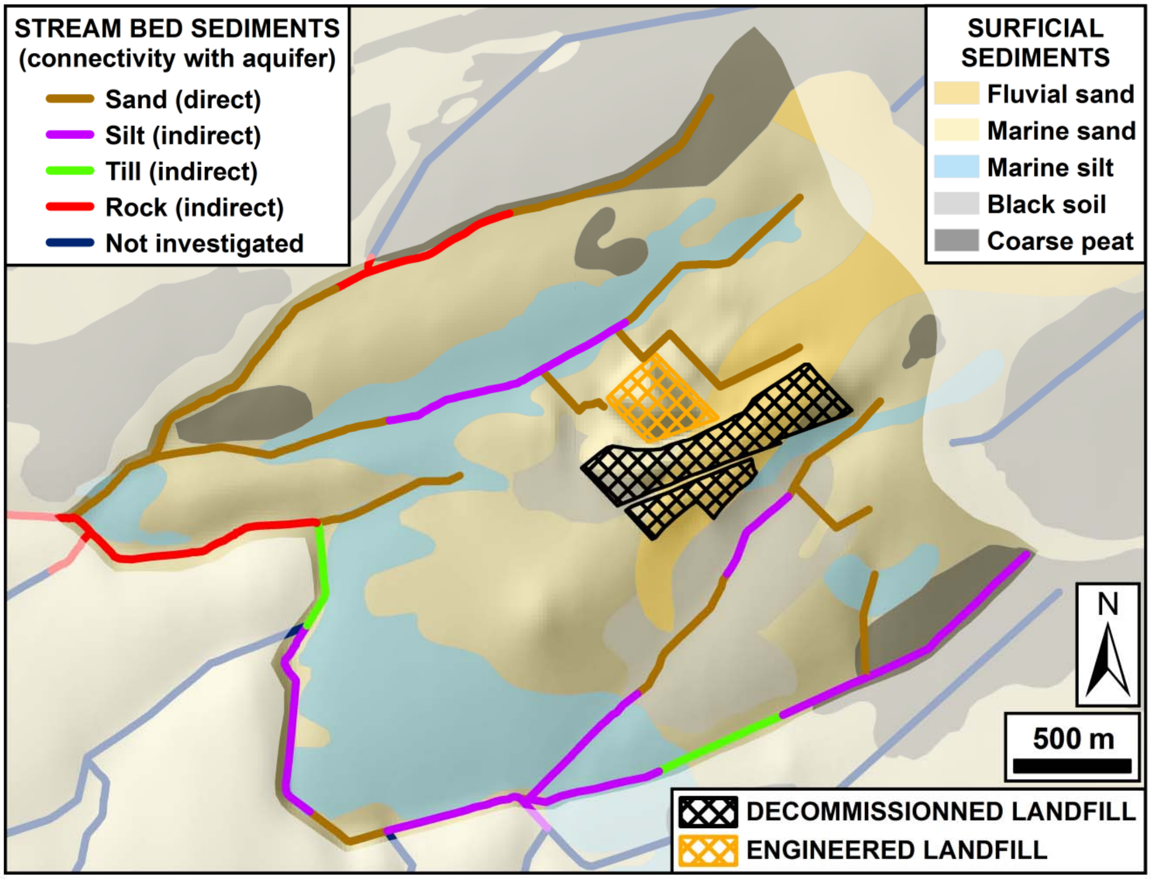


Figure 5.3 : Surficial sediment distribution

Surficial sediment distribution and inferred link between the granular aquifer and streams based on sediment types found in stream beds. The surficial material distribution is based on soil and Quaternary sediments maps, soil sampling and field investigations. The location of field work is shown on Figure 5.2a and Table 5.1 provides details on the various soil contexts

Minnesota, Delin et al. (2007) demonstrated that small-scale variability in recharge was related to variability in soil properties and land-surface topography. Coes et al. (2007) stated that recharge correlated significantly with the surficial soil properties of percent sand and percent silt. Moreover, Gee et al. (1992) demonstrated that coarse-texture soil without plant cover yield the most recharge while fine texture soils with or without plant cover yield the least. For this study, five main categories of soils were defined on the basis of Quaternary and pedology maps, soil permeability values and field verification (Figure 5.3). These categories refer to as pedological units described along with physical measurements in Table 5.1. The main soil types (sand and silty sand), which cover 67% of the study area, are both poorly drained podzols of marine origin, with high to moderate saturated hydraulic conductivity values ranging from 10^{-4} and 10^{-5} m/s. Despite the potential ease for water to infiltrate these soil types, they are known to retain water due to their high porosity, elevated proportion of fine material and compactness. The area drawn in blue on the map, under the name marine silt, delineates a portion of the territory where clayey sand is located in the shallow subsurface (0.3 to 1 m depth) and keeps the water table under semi-confined conditions where its thickness reaches more than about 0.3 m. Moreover, this soil type limits water infiltration. Three other soil types cover, respectively, 11% of the study area (fluvial sand, coarse peat and black soil). The fluvial sand is permeable and allows a better drainage. Unfortunately, this soil type is located immediatly below the location of the former unlined landfill, which means that leachate can easily infiltrate the ground and reach the water table. Coarse peat is present at a few locations and was found to have an elevated saturated hydraulic conductivity (10^{-3} m/s). Water infiltrated easily, however due to the elevated porosity, the coarse peat has a tremendous capacity to retain water. On the contrary, well decomposed black soil has a low saturated hydraulic conductivity which will either lead to water puddles at the surface of the soil or will promote shallow hypoderdermic flow (0-10cm) toward streams where the slope is around 1.5% or more and where areas are deforested. The identification of soil variability within the study area helped understand how the physical properties of the surficial sediments can affect recharge processes and narrowed down soil sampling sites for chemical analyses.

Hydraulic links between the aquifer and surface water within the study area was deduced from stream bed sediment types and hydraulic head measured in drive point piezometers installed along the stream edges (Figure 5.4). Stream bed sediments were collected up to 3 m depth on stream edges or directly in the stream where the water level was less than 10 cm. The material was either loose sand, compact sand, loose silt, compact silt, till or bedrock. In general the streams are gaining streams. In the northern section of the study area, it was obvious that the aquifer drained directly into the streams as sand was very loose and hard to collect and hydraulic heads measured on the stream banks were above the water level in the streams. Drive point piezometers installed in silt and till had water levels slightly higher than the water level measured in the stream and water took a few days to equilibrate to static levels when water was pumped from the piezometer. In this situation the link between the aquifer and the stream was qualified as “indirect” and it was inferred that the stream drained mostly the superficial portion of the aquifer in these areas as the flow coming from the deeper part of the aquifer is slowed down by the thickness of the compact silt or till material in the stream beds. The direction of flow for the hydraulic link between bedrock outcrops and stream was not investigated. However, it is suspected that the direction of the vertical hydraulic gradient might vary between the fractured bedrock aquifers in relation to the water table level within the granular aquifer. This investigation was useful to understand how groundwater flow was linked to local streams within the study area and to select baseflow measurement locations for recharge evaluation.

Tableau 5.1 : Physical properties for the main surficial soil types
Description and basic physical properties for the main soil types found within the study area. Measured physical values attributed to the 5 soil contexts (Figure 5.3) and theoretical values related to each category of soil found within the study area

Soil Type	Pedological Units (Laplante 1962)	Sediment Types	Area* (%)	Estimated Total Porosity (Croteau 2006)	Measured Sat. *K* (m/s)	Unsaturated Zone Thickness (cm)	Drainage Capacity (Laplante 1962)
Fluvial sand	Beaurivage Loamy sand (podzol)	Coarse to loamy fine sand	11	0.4	10^{-3}	>100	Readily drained
Marine sand	Saint-Jude loamy sand (50%), Saint-Samuel loamy sand (30%), coarse peat (20%) (podzol)	Coarse to fine loamy sand	26	0.43	10^{-4}	10-150	Imperfectly drained
Marine silt	Saint-Jude loamy sand (50%), Saint-Samuel loamy sand (30%), coarse peat (20%) (podzol)	Fine loamy sand. Presence of a silt layer (0.1-3 m) confining the water table where its thickness exceed 0.5 m	41	0.45	10^{-5}	<100	Imperfectly drained
Black soil	Well decomposed black soil	Organic (humic)	11	0.85	10^{-6}	10-50	Not well drained
Coarse peat	Coarse peat	Organic (sphagnum)	11	0.9	10^{-3}	10-100	Not well drained

*: the total study area covers 12 km^2.

5.4.2 Shallow groundwater chemistry in relation to leachate, recharge rates and soil properties

Aquifers need to have an adequate supply of electron acceptors to naturally remediate contaminants from groundwater (Cozzarelli et al. 2011; Scholl et al 2006; van Breukelen et al. 2003; van Breukelen 2003). Recharge processes and associated changes in electron acceptor availability in the unsaturated soil interface may provide a fresh supply of electron acceptors for plumes attenuation (Scholl et al. 2006). Therefore, natural soil types and recharge rates will control, in part, the natural attenuation processes of the upper portion of the aquifer. In this study, shallow groundwater chemistry was investigated to infer natural background concentrations of inorganic elements for shallow groundwater in relation to leachate groundwater, recharge rates, and soil types. Figure 5.4 illustrates the geochemistry for shallow groundwater samples in relation to soil context. While no clear trend can be observed between soil types and shallow groundwater chemistry, certain generalizations can still be made. Leachate groundwaters were distinguished by their pH above 7 and elevated concentrations of total dissolved solids (TDS), hydrogen carbonate, sodium, potassium, chloride, iron and sulfate. Groundwater samples showing an elevated TDS value above 300 mg/L (TDS = sum of ions + SiO_2 mg/l - HCO_3^- * 0.5082; Hounslow 1995) were either related to the leachate plume emitted by the decommissioned landfill or to the combined effect of a hypodermic flow, in deforested portions of the territory, and where the slope was over 1.5%. Where shallow groundwater is affected by significant hypodermic flow, the groundwater chemistry was also characterized by elevated concentrations of strontium, calcium and barium above background values and a pH above 6. Moreover, natural shallow groundwater contains chloride concentrations below 1 mg/L in general, except in large deforested zones where concentrations could reach 5 mg/L. A Ca-HCO_3 water type where TDS is less than 50 mg/L is related to locations where recharge is suspected to be the most elevated for the study area. At these locations, the subsurface material is known to be homogeneous and coarser when compared to other locations. Moreover, the shallow groundwater will remain acidic (pH 4-5) at these locations. The water type Ca-HCO_3-SO_4 is typical for wells located near a stream, where recharge groundwater mixes with upflowing groundwater. Also, at these locations fresh water oxidized S^{2-} found in organic soil and where Fe remains in the oxidized form. However, the most common water type found in the superficial part of the saturated zone is Ca-HCO_3-Fe without regard to the type of soil present at the surface.

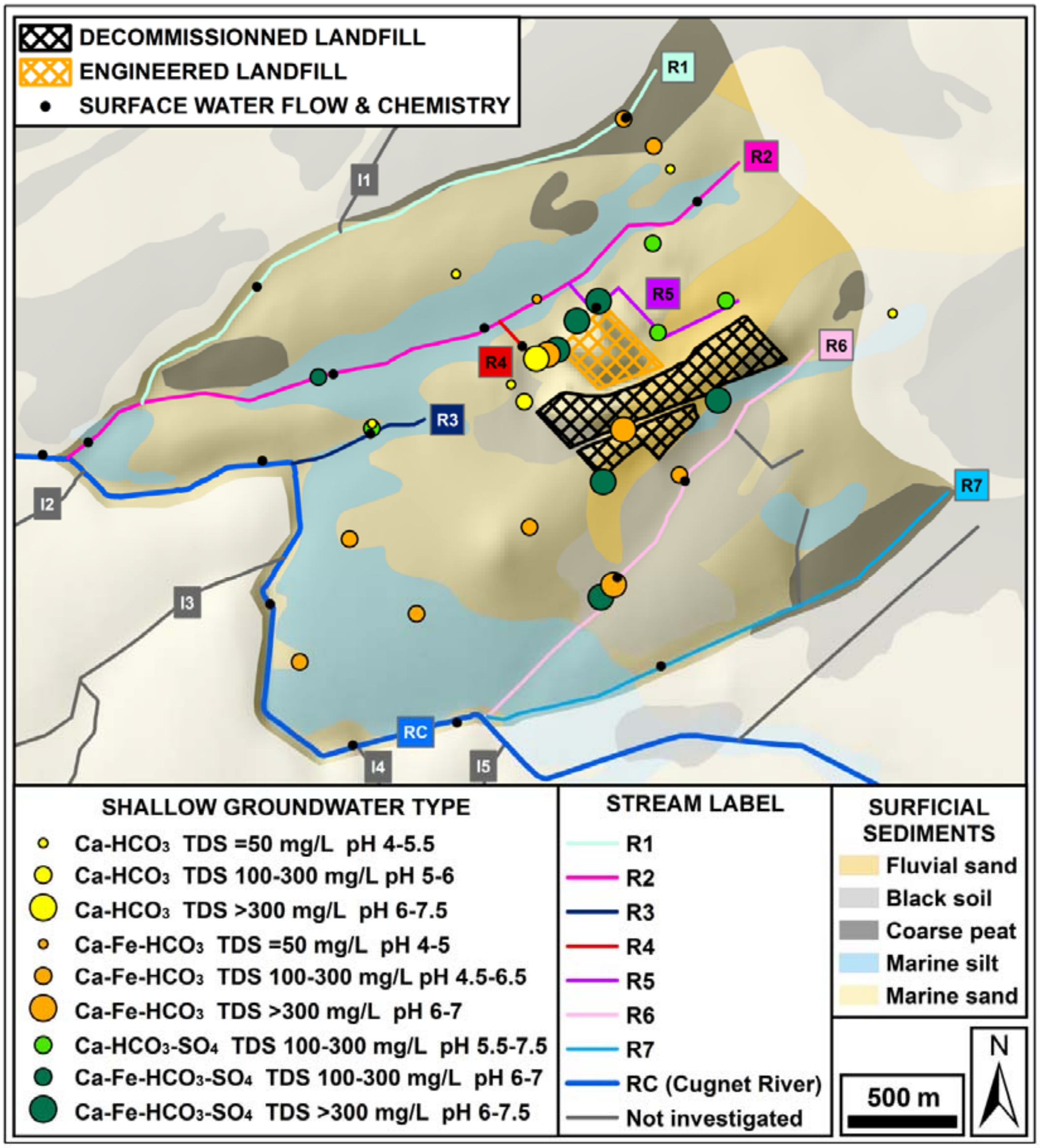


Figure 5.4 : Groundwater types in relation to surficial sediments
Labels assigned to specific stream stretches are illustrated. (I#) stands for incoming stream.
Locations for flux and water chemistry measure point (MP) in stream are also indicated

For most of the study area, soils are ferruginous and clayey, thus causing the shallow subsurface to be under anoxic conditions. Moreover, the soil is mostly acidic and contains a relative abundance of iron and manganese ions. Added to these soil conditions, the aquifer also has naturally reducing conditions characterized by elevated concentrations of iron, manganese and sulphur, which complicated plume delineation and identification of natural attenuation processes. Soil chemistry was investigated for selected samples representing various soil contexts found within the study area. The goal was to link shallow groundwater geochemical signatures to soil context. At afield-scale of 800 m^2, Derby et al. (2013) established a link between surface topography, groundwater quality and groundwater electrical conductivity, which demonstrated the importance of investigating the relation between land surface and shallow groundwater quality to understand where the shallow groundwater is the most susceptible to contamination. In this study, not enough samples were collected and analysed for chemical concentrations to be able to establish clear relationships between the parameters mentioned. Results from the chemical analyses of soil showed up to 40% of organic carbon for peat soil and less than 5% for the sandy loam. Also, the organic material had ten times the concentration of sulphur as well as higher concentrations of calcium, barium, sodium and trace of arsenic. Higher concentrations of dissolved sulphur and iron were also found in the shallow groundwater below 30-90 cm of peat that kept shallow groundwater under anoxic condition. In general it could be concluded that the longer the water is retarded by soil during infiltration (i.e. peat, black soil) the stronger the geochemical signal in relation to the soil type in place. Water found in recharge zones (i.e. higher topography, more permeable subsurface material) infiltrates more readily through the unsaturated zone, thus preventing the shallow groundwater from acquiring a geochemical signal related to the soil in place. Without being able to make a clear assumption about the link between soil properties and groundwater chemistry, it seems that the shallow groundwater chemistry is mostly influenced by water table fluctuations, related topographic location, land use, soil pH and redox conditions as well as the permeability of the subsurface material.

Historical groundwater geochemistry data (2007-2012), for seven wells located within the landfill property limits, reveal seasonal fluctuations of dissolved iron and sulfate concentrations for the upper portion of the aquifer (1-5 m) in relation to water table fluctuation. Cozzarelli et al. (2011) observed redox reactions in the upper boundary of a leachate plume responding to recharge events and seasonal water table fluctuation. Scholl et al. (2006) concluded that recharge and seasonal water table fluctuations drove sulfide oxidation and reduction in the upper part of the aquifer and suggested that similar cycles of ammonium oxidation and denitrification may occur under some conditions. While Scholl et al. (2006) found that seasonal patterns of sulfate concentration were the lowest in the summer, when transpiration decreased recharge and groundwater levels were at a minimum, and highest in the fall and early winter after recharge occurred, no straight-forward trend could be established at the present study site between water table levels and iron and sulfate concentrations. Scholl et al. (2006) documented many studies that have tested, in the laboratory and in the field, sulfate reduction and oxidation patterns. Sulfate concentrations in groundwater can be increased by oxidation of sediment-associated sulfides, or can be decreased by sulfide mineral precipitation during microbial sulfate reduction (Knöller and Schubert 2010; Scholl et al. 2006). In anoxic environments, bacterial sulfate reduction produces sulfides, which may react with iron to form sulfide minerals (Knöller and Schubert 2010). When redox conditions change due to changes in groundwater level or influx of recharge, these sulfides may be oxidized to sulfate (Knöller and Schubert 2010; Scholl et al. 2006). The shallow groundwater chemistry data presented in Figure 5.4 were collected during the summer under low water table conditions. At the study site, it was observed that water table fluctuation and recharge rates can affect these water types in relation to iron and sulfate concentrations. Under natural conditions, one sample located in the marine sand had an iron concentration ten times lower after the spring recharge than at the end of summer. Scholl et al (2006) demonstrated that sulfate reduction was an active process in both contaminated and uncontaminated aquifers and that recharge-driven sulfate reduction could be an important process in contaminant remediation. Iron and sulfate reduction patterns in relation to the water table fluctuation were observed at the study site under natural and contaminated conditions. Since the natural capacity of leachate attenuation will be based on the available ions for reactions, these reactions can be crucial in understanding contaminant mitigation. Therefore, based on this investigation, a detailed isotopic investigation of sulfate and iron in relation to temporal water level fluctuation would be required to confirm the rates and the processes involved. The investigation of shallow groundwater chemistry in relation to leachate, recharge rates and soil properties allowed the identification of key processes at the study site and the delineation of the leachate plume in the upper portion of the aquifer. Natural conditions were found to be either oxidizing or anoxic. Anoxic or sub-oxic zones were characterized by elevated concentrations of dissolved iron and the presence of sulfur. By understanding the natural conditions better, leachate groundwater could be identified. And while no clear link can be established between soil and shallow groundwater based on the available data, multiple variables were also identified as being involved in the shallow groundwater chemical signature. To further understand the leachate extent and the potential points of discharge of leachate in streams, surface water quality was also investigated.

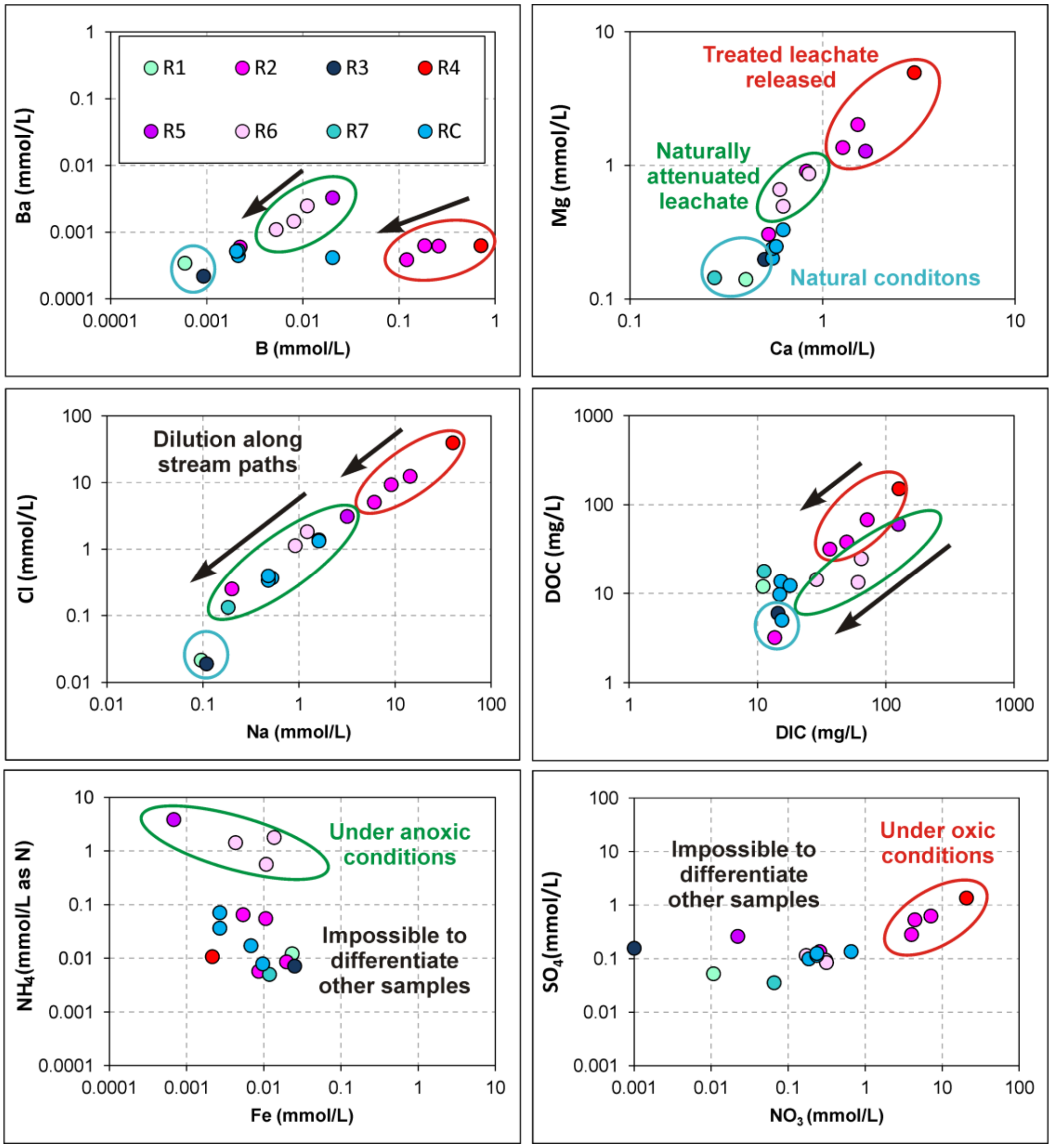


Figure 5.5 : Surface water quality

Discharge of leachate in surface water and dilution of inorganic elements along stream stretches illustrated in Figure 5.4. Treated leachate released in R4 can be traced all the way to the outlet in RC through R2. Discharging leachate in groundwater that has undergone natural attenuation is observed in R5 and R6 and can also be traced from R6 all the way to RC. The red circles indicate samples taken along the path where treated leachate is released. The green circles indicate samples where leachate has discharged from groundwater into a stream stretch, after which dilution is observed along the stream path. Blue circles indicate samples representing the natural condition of the stream. Table 5.3 gives details on the tracers of interest in relation to the stream context

Tableau 5.2 : Surface water quality

Geochemical tracers for stream stretches receiving leachate from different sources and natural background conditions found within the study area. DL stands for detection limits and – for value below the detection limit.

Geochemical concentrations (mg/l) found in streams under different conditions									
Parameter		**Treated leachate is released**			**Untreated leachate migrates**			**Natural stream conditions**	
	DL	MIN	MAX	**Tracer**	MIN	MAX	**Tracer**	MIN	MAX
Ca	0.02	25	120	**x**	22	67	**x**	11	21
Mg	0.01	8	120	**x**	4.9	31	**x**	3.4	7.4
Na	0.1	37	920	**x**	11	73	**x**	2.2	4.6
K	0.1	14	440	**x**	3	50	**x**	0.7	2.8
N-NH_4	0.02	0.11	0.91		0.24	54	**x**	0.07	0.17
Alc. $CaHCO_3$	1	86	532	**x**	65	550	**x**	54	65
Cl	0.05	47	1400	**x**	12	110	**x**	0.67	8.9
SO_4	0.1	13	130	**x**	8.1	25		3.4	15
NO_3	0.01	9.2	290	**x**	0.31	4.4		-	-
NO_2	0.01	-	1.2	**x**	0.02	0.39		-	-
Sr	0.002	0.23	0.74	**x**	0.19	0.77	**x**	0.095	0.16
B	0.005	0.22	7.7	**x**	0.022	0.22	**x**	0.0064	0.024
Ba	0.002	0.053	0.086		0.061	0.45	**x**	0.012	0.082
Ni	0.001	0.0072	0.14	**x**	0.0029	0.034	**x**	-	0.02
Li	0.001	-	0.11	**x**	-	-		-	0
Sb	0.001	0.0017	0.006	**x**	-	-		-	0
As	0.001	0.0011	0.016	**x**	-	0.0057	**x**	-	0.0027
Cr	0.0005	-	0.027	**x**	-	0.0011		-	0.001
Cu	0.0005	0.00078	0.012	**x**	-	0.00092		-	0.048
Co	0.0005	0.00057	0.011	**x**	-	0.0085		-	0.0032
Mo	0.0005	0.00059	0.0058	**x**	-	0.0013		-	0
COD	10	28	340	**x**	11	91	**x**	-	36
BOD_5	4	-	-		-	17		-	4
C-DIC	0.1	18.0	126.7	**x**	14.9	125.4	**x**	11.1	14.4
C-DOC	0.1	12.3	151.0	**x**	5.0	60.4	**x**	3.2	17.9
Estimated TDS	-	409	6188	**x**	243	1658	**x**	106	232
Conductivity (m*S*/cm^3)	-	0.431	8.026	**x**	0.22	1.432	**x**	0.03	0.205

5.4.3 Surface water flow and quality

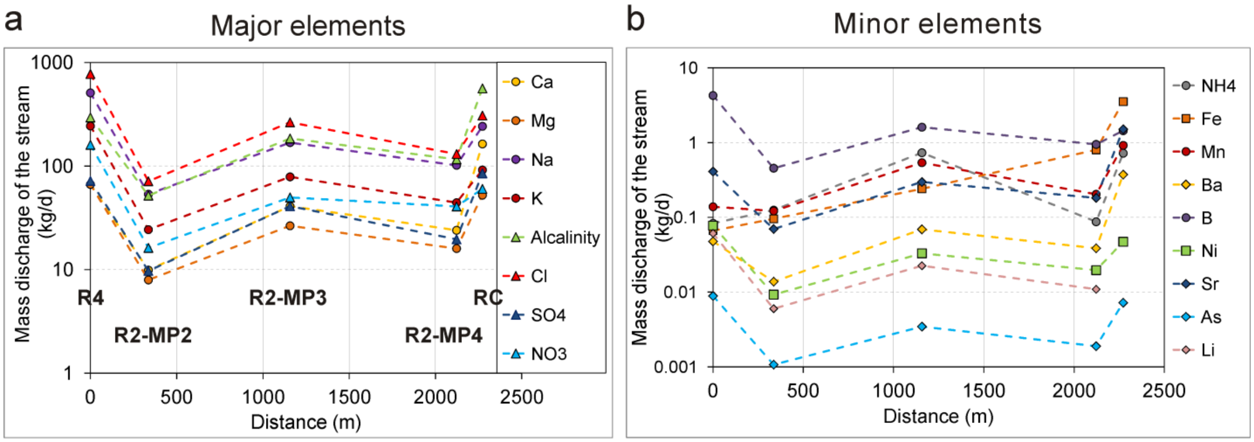
Surface water quality was assessed to gain knowledge on the natural chemistry and to identify possible discharge zones of leachate into the surrounding streams. Surface water flow and chemistry were investigated at each measuring point (MP) indicated in Figure 5.4. Leachate collected from the engineered landfill is released in stream R4, illustrated on Figure 5.4, after aerating in a large basin and decanting into peat beds. Discharging leachate from groundwater that has undergone natural attenuation is observed in R5 and R6 (Figure 5.4). Figure 5.5 illustrates the relation between a few inorganic elements found in surface water that show the difference in concentration between three sources: 1) natural stream chemistry; 2) treated leachate release to stream and; 3) naturally attenuated leachate discharging into streams via groundwater flow. Dilution is observed along all stream paths of leachate emissions.

Tableau 5.3 : Description of surface water flow and chemistry at Measure Point (MP)

Surface water flow and chemistry at Measure Point (MP) are described in terms of Flux evolution explaining variability in mass flux on Figure 5.6. Locations of the MP are indicated on Figures 5.4 and 5.2c.

Treated Leachate – surface release into stream		
Measure Point (MP)	**Measure Point (MP) description**	**Flux evolution**
R4	Drainage ditch located on the landfill property, the treated leachate is released directly 50 m upstream	Large initial flux ranging from 50-250 m^3/j except in winter
R2-MP2	Entry of R4 (drainage ditches) in R2	Mass loss to aquifer
R2-MP3	MP further along R2	Gain from sandy aquifer connected directly to stream
R2-MP4	Last sampling point on R2 after merging of R1	Gain from stream R1
RC-MP5	Last MP on the Cugnet River at the outlet of the study area	Gain from Cugnet River
Untreated leachate – subsurface discharge into streams		
R6-MP1	First MP along R6 located south of the former landfill	Most of the leachate discharge into this stream occurs upstream of this point
R6-MP2	Second MP along R6	The concentration increases slightly at this location, gain from aquifer and hypodermic flow
RC-MP1	First MP along the Cugnet River after the merging of R7 and R6	Gain from R7 and RC
RC-MP2	Second MP along the Cugnet River after incoming stream I4	Gain from I4
RC-MP3	Third MP along the Cugnet River	Gain from the aquifer
RC-MP4	Fourth MP along the Cugnet River	Gain from the aquifer and incoming stream I3

Treated leachate released in R4



Untreated leachate naturally discharging in R6

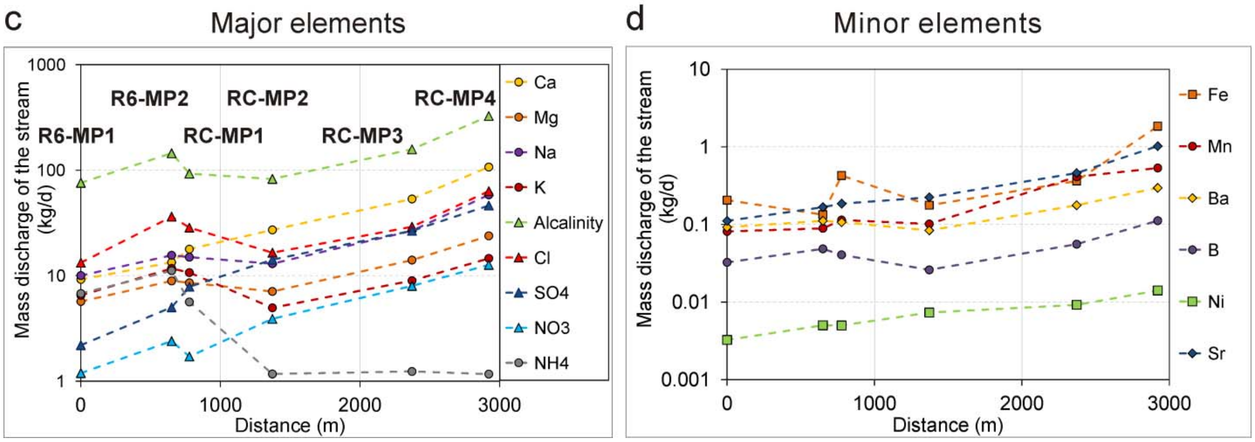


Figure 5.6 : Mass flux calculated for selected leachate indicators

Mass flux calculated for selected leachate indicators (kg/d) into stream stretches of interest. Table 5.3 gives details on the location context at each measure point (MP). Upper figures a and b illustrate mass discharge for the treated leachate released in R4 from the collection basin of the engineered landfill at a rate of 250 m^3/d. Lower figures c and d indicate mass discharge for the leachate discharging from groundwater in R6. Distance 0 represents the location for the first sampling station along the stream (Figure 5.4 and Figure 5.2c). Missing elements from Figure d but found in b were mostly found below detection limits along the southern streams

Figure 5.6 shows mass flux (kg/j) for major and minor elements along respective stream paths for the two sources of leachate. Table 5.3 describes the location of each measuring point in relation to stream context and flux evolution. Treated leachate released in R4 travels to stream R2 and finally reaches the main Cugnet River (RC) at the North West outlet of the study area.

The treated leachate shows higher concentrations for many elements and undergoes dilutionalong its path in streams (Figure 5.5). Since the treated leachate was aerated, its composition reveals elevated concentration of nitrates and sulfate compared to water samples from natural conditions. Untreated leachate discharging into stream R6 shows elevated concentrations of ammonium and iron in contrast to the aerated leachate. Moreover, Figure 5.6 demonstrates that it takes over 1000 m and the dilution effect of the Cugnet River before ammonium concentration returns below the detection limits in the surface water. Most elements on Figure 5.6 show the same patterns for minor and major elements at the exception of iron, manganese and sulfate that are also controlled by natural redox and pH conditions. In that case, iron, manganese and sulfate can only be used as indicators of leachate if they are found in elevated concentrations, but cannot be used as tracers along stream paths.

Based on the water chemistry and mass flux interpretation, natural concentrations and a list of parameters to be monitored under the two different conditions of leachate emission can be inferred (Table 5.2). Furthermore, this analysis successfully identified discharge zones of leachate into the surrounding streams R5 and R6. Unfortunately, the release of treated leachate in the northern stream masks the possible discharge of leachate originating from groundwater in R2.

5.4.4 Recharge evaluation

5.4.4.1 Well hydrographs

Nine (9) well hydrographs, distributed within the study area (Figure 5.2d), were monitored throughout year 2011 and were investigated for their potential to evaluate recharge with the water table fluctuation method. The water table fluctuation method is based on the premise that rises in groundwater levels in unconfined aquifers are due to recharge water reaching the water table (Healy 2010). Since groundwater level fluctuations are not always indicative of groundwater recharge or discharge (Healy 2010; Healy and Cook, 2002), not every well can be used to derive recharge from changes in water level.

Figure 5.7 illustrates the well hydrographs measured in the study area, corrected for atmospheric barometric pressure, in relation to precipitation and mean air temperature. Small recharge events are visible in well hydrographs below 121 m asl as soon as temperature rises close to zero

Celcius in winter. The first recorded recharge peak visible in all hydrographs occurs in late March as the temperature rises above zero. The main recharge event recorded for 2011 occurs in the months of April and May and is only clearly visible on well hydrographs P03 and P04 which are located on local topographic highs. Throughout the summer, small recharge events, seemingly linked directly to precipitation, are visible in all wells. However, many small short-term fluctuations recorded in wells located below 120 m asl seem to be induced by other mechanisms than recharge. Some possible causes for short-term water fluctuations recorded at the water table at the study site are discussed below.

At the site, the water table is very shallow (0.1-2 m) and can exhibit diurnal fluctuations in response to higher evapotranspiration during the day (Healy 2010). Wells P14, P07, P17, P08 are located next to a stream or to drainage ditchs that can prevent the water table from rising at these locations. Moreover, many wells are located at local topographic lows (P23, P05, P17, P06, P08) and are prone to groundwater ridging. The increased groundwater discharge during runoff events is related to a rapid rise in hydraulic head along the perimeter of transient and perennial discharge areas. This groundwater ridging phenomenon arises from the almost instantaneous conversion of the near-surface tension-saturated capillary fringe into phreatic water (Sklash and Farvolden 1979). Since the water table is less than 30 cm deep at many locations, only a small quantity of water is required to saturate the ground. Also, soil in the low topographic locations is often composed of fine clayey soil preventing water from infiltrating readily. In the described conditions, the combined effect of more than one mechanism might induce false rises and falls of the water table. Since most wells can be under the influence of many of these mechanisms, P03 and P04 were the only hydrographs selected to evaluate representative recharge values with the water table fluctuation method. These two wells are located on topographic highs, they both show two main peaks clearly associated with recharge, there is no significant groundwater pumping in the area, the water table is deeper, the surface soil sandy and coarse (weak capillary zone in function of evapotranspiration), and there are no nearby streams. P03 and PO4 demonstrate that in the absence of recharge, water levels recede due to the flow of groundwater to the streams connected to the aquifer, which steadily decreases the volume (and level) of water in the aquifer. During these periods without recharge, water levels in observation wells follow recession curves, which are nearly linear. Recharge occurs mostly during spring snow melt and in the fall. During winter, there is minimal recharge as precipitation accumulates as snow on the soil surface. During summer, evapotranspiration (ET) consumes all the infiltration that does not runoff.

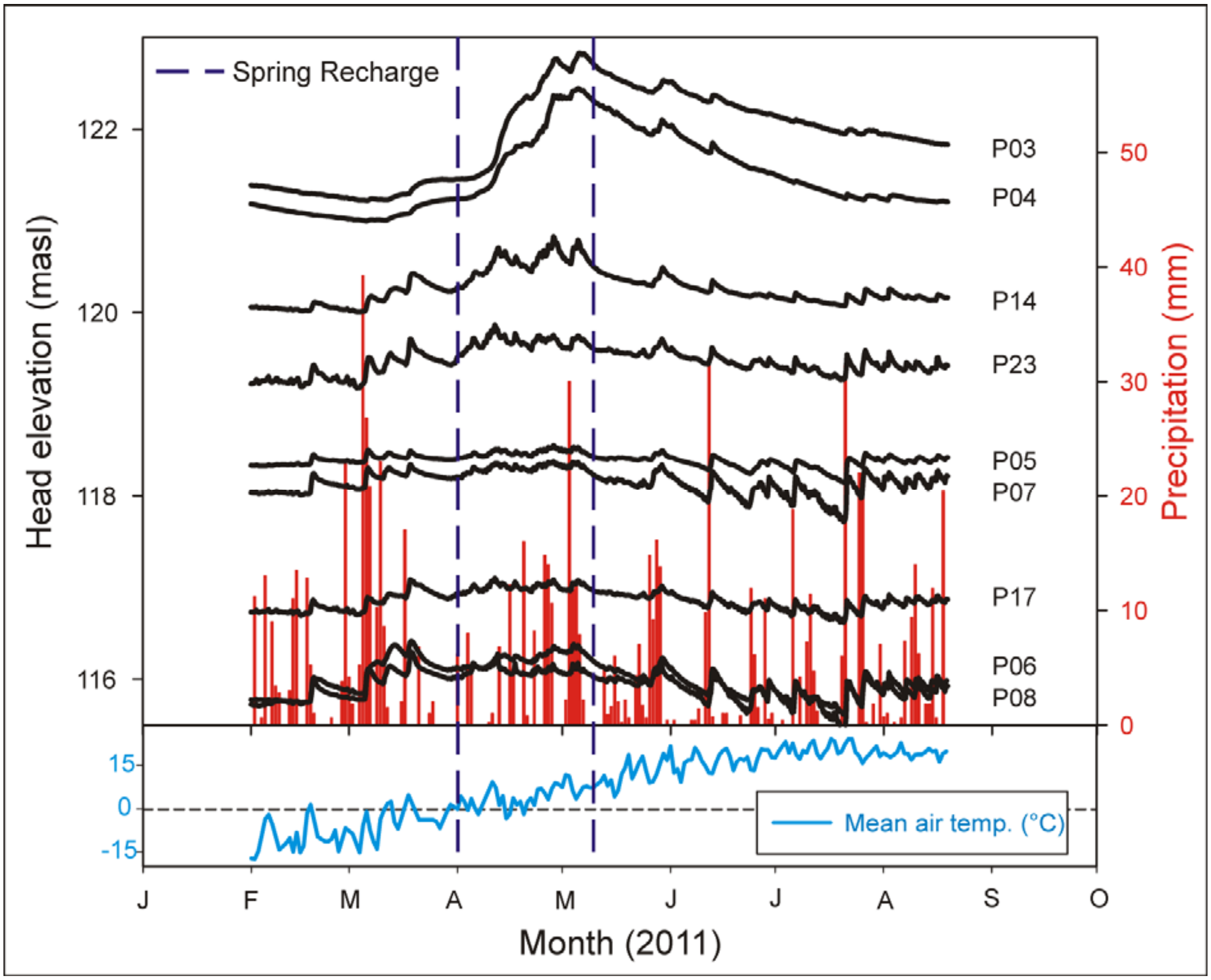


Figure 5.7 : Well hydrographs

Well hydrographs distributed in the study area related to mean daily air temperature and precipitation (locations shown in Figure 5.2b). The patterns of hydrographs depend on the hydrogeological contexts (discharge, recharge, etc.) and on their geographical locations. The main recharge event occurs in the spring during snowmelt

The changes in water level that are caused by an infiltration event can be obtained from a well hydrograph by the distance between the peak in water level and the extrapolated level of the recession curve under the peak. The interpretation of well hydrographs to derive recharge is thus relatively straightforward. However, small or diffuse recharge events are hard to assess from hydrographs, as the competing effects of level recession and small infiltration do not lead to clear peaks in water level. There is no long-time series available for the study area. Additionally, the estimation of S_y can have a major effect on the recharge estimated with this method. Recharge was calculated as follows:

$$I = S_y \cdot \Delta h \text{ [1]}$$

where, I is infiltration (recharge, mm/y) and S_y is specific yield (-). Values were estimated at 536 mm/y for P03 with a S_y of 0.2. Since, S_y is often unknown and represents the main difficulty with this method, the value of 0.2 was selected based on theorical values reported in the literature for fine sands (Healy 2010) and based on the adjustement of a set of parameters described in the next section. Using the same specific yield value, 0.2, a recharge estimate comparable to well hydrograph P03 can be obtained for well hydrograph P04. However, no recharge estimate was derived from well hydrograph P04 with the WTM because it was impossible to infer a unique value for the specific yield du to the high heterogeneity of the sediments at this location.

5.4.4.2 Daily soil moisture balance adjusted to well hydrographs

A recharge assessment combining daily soil moisture balance (SMB) and well hydrographs (Baalousha 2005), adapted to unconfined aquifers in temperate regions (Lefebvre et al. 2011), was used in the study area. The principle of the method is to 1) estimate recharge from daily SMB derived from weather data and 2) to calculate the change in groundwater level related to that daily recharge using the specific yield (S_y) of the unconfined aquifer in order to compare synthetic hydrographs derived from daily SMB to observed water levels, thus allowing the calibration of hydrological parameters used in the SMB. The method also accounts for the recession of groundwater levels during periods without recharge. Equation 2 summarizes the principle of the method as adapted by Lefebvre et al. (2011):

$$\Delta h = \frac{I}{S_y} - \frac{Q_{out}}{S_y \cdot A} = \frac{P - R - ETR}{S_y} - \frac{\partial h}{\partial t} \cdot \Delta t \text{ [2]}$$

The daily change in water level Δh is related to 1) the change in water level induced by recharge infiltration I, considering the specific yield S_y of the aquifer, and 2) the decrease in water level related to the loss of water from the aquifer Q_{out} over the outflow area A. The infiltration is calculated from a water balance, as follows:

$$I = P - R - ETR \text{ [3]}$$

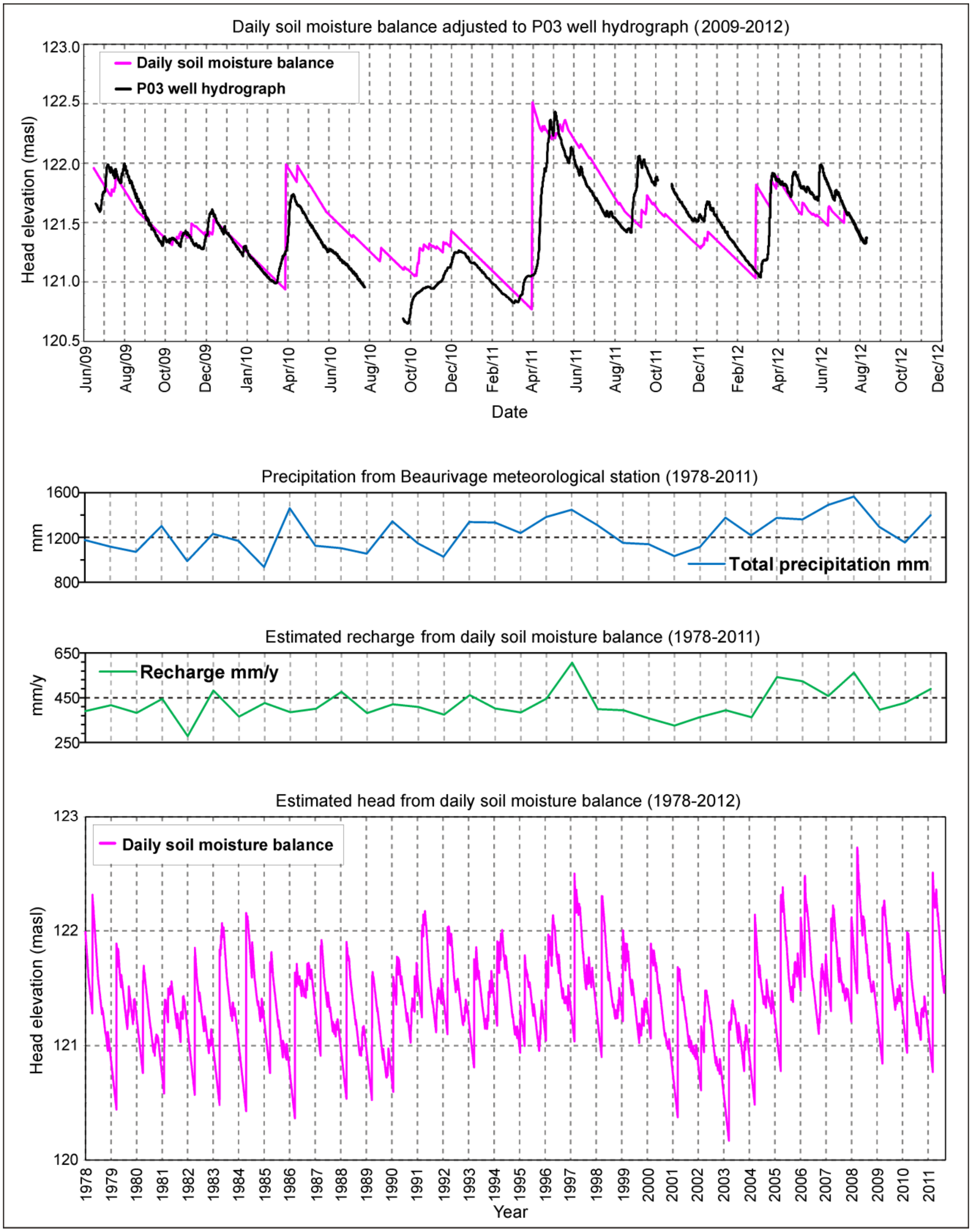
Where P is precipitation, R is runoff and ETR is the real evapotranspiration. R was obtained from the product of the runoff coefficient CR by precipitation ($R = P \cdot C_R$). Potential ET (ETP) was calculated using the Thornthwaite method (de Marsily 1986), whereas ETR was obtained from the maximum value of ETP compared to the sum of infiltration I and the Readily Available Supply RAS of water in soils calculated after the previous time step ($ETR = \mathrm{MAX}(ETP; I + RAS)$. Water accumulation as snow was considered and snowmelt was set to occur above a certain temperature (T threshold). This threshold actually had to be set higher than 0 °C in order not to calculate recharge events for short duration warm winter periods near 0 °C where water refreezing into the snowpack prevents infiltration to occur. The water level recession rate (dh/dt) in periods of no recharge was directly applied as observed on hydrographs.

Daily soil moisture balance calculations based on precipitation-temperature time series (1978-2011) collected from the Beaurivage meteorological station (Figure 5.1) were used to calibrate the well hydrograph P03 (2009-2011). Modifications of hydrological parameters allowed the match of well hydrographs with calculated changes in water level, after considering the delay related to the infiltration time after a recharge event (infiltration through the unsaturated zone). Table 5.4 lists hydrologic parameters used to match synthetic hydrographs to measured ones, as well as the range of values that could still provide an acceptable match. The top graph of Figure 5.8 illustrates the synthetic water levels obtained from daily soil moisture balance adjusted to well hydrograph P03 (data collection from well P03 was interrupted throughout the 2010 groundwater sampling campaign). It shows that observed water levels are satisfactorily reproduced by the daily SMB.

Tableau 5.4 : Parameters of the calibrated soil moisture balance

Parameters of the calibrated soil moisture balance (shown on Figure 5.8). RAS stands for readily available supply.

Parameter	Calibrated value	Range of values
Runoff coefficient C_R	0.3	0.25-0.35
RAS max (mm)	20	10-30
Specific yield S_y	0.2	0.18-0.22
Recession rate dh/dt (m/y)	0.3	0.2-0.6
Snow melt temperature (°C)	3	1-5
Initial water elevation (m)	122	121-123
Recharge delay (d)	15	10-30


Daily soil moisture balance adjusted to P03 well hydrograph (2009-2012)

Precipitation from Beaurivage meteorological station (1978-2011)

Estimated recharge from daily soil moisture balance (1978-2011)

Estimated head from daily soil moisture balance (1978-2012)

Figure 5.8 : Soil moisture balance compared to well hydrograph

Calculated water levels based on daily soil moisture balance compared to observation well P03 hydrograph (top graph) and for longer-term available weather data (bottom graph). Daily soil moisture balance provides an average recharge of 420 mm/year from 1978 to 2012

Recharge and water levels were also both estimated from the daily SMB from 1978 to 2011 (lower graph of Figure 5.8). For the central regions P03 and P04, an average of 420 mm/y (values vary between 279-606 mm/y), was estimated with a runoff coefficient of 0.3 and a low value of RAS of 20. These long-term results show that precipitation (and other climate conditions not shown) significantly changes from year to year, thus leading to quite variable values of recharge in any given year. It is thus important to apply daily SMB calculations to long weather data sets to get representative average recharge values. Also, Lefebvre et al. (2011) mention that synthetic daily SMB water levels are "cumulative" curves, so that if hydrological parameters used in calculations are non-representative, the calculated water levels will systematically shift either above or below observed values. The absence of systematic departures in estimated water levels in the lower graph of Figure 5.8 thus indicates that the set of hydrological parameters is coherent, thus further validating the hydrological parameters used in SMB calculations, besides the match with observed levels.

For the wells located in low topographic areas, an attempt was made to evaluate recharge with the daily soil moisture balance adjusted to well hydrographs. A good match was achieved by using a high run off coefficient of 0.6-0.7 and a specific yield of 0.1 and a very low RAS since the water table is almost at the ground level. Yearly recharge for the wells located in the regions peripheral was estimated at 100-150 mm. Since the wells used for this last analysis have a water table nearly at the ground level and due to other mechanisms causing noise in the wells hydrographs, this value only represents a crude approximation.

5.4.4.3 Groundwater isotopic signature

Figure 5.9 shows isotopic results for stable isotopes δ^2H and $\delta^{18}O$ and the radiogenic isotope ^3H. Isotopic measurement locations are indicated in Figure 5.2d. Figure 5.9a shows the relationship between δ^2H and $\delta^{18}O$ in groundwater for the study area in relation with the Ottawa (black) and the Chaudière River watershed meteoric water lines based on analyses of precipitation (Benoit et al. 2011). The groundwater types were derived from multivariate analysis discussed in Tremblay et al. (2013). Stable isotope values from precipitation are mostly a function of prevailing atmospheric conditions during rainfall and snowfall (Blash and Bryson 2007; Clark and Fritz 1997). The stable isotopes collected in the Chaudière River watershed, where the study is located; show enriched values for the summer-fall precipitation compared to lighter values for the snow and spring samples (many points plotted outside the frame of the graph with much lighter values). The same pattern is observed for a few samples collected in the spring from the acidic water group plotting lower on the meteoric line, and for a few samples collected in late fall that plot higher up. Precipitation and groundwater data correspond well and it can be concluded that aquifer recharge is controlled mostly by spring snow melt and that the shallow system is replenished with young water. While most groundwater samples collected in late summer plot along the local meteoric line, many samples plotted slightly above the line. This phenomenon can be explained by mineralization through recharge, which is supported by the correlation between conductivity and $\delta^{18}O$ (Figure 5.9b). Also, because heterogeneity of soil and aquifer material induces variable redox conditions in the study area, there are other explanations for the observed slight enrichment: precipitation or adsorption of iron causing fractionation of the stable isotopes involved in the reactions (Oba and Poulson 2009; Teutsch et al. 2005) or sulfate redox processes (Cozzarelli et al. 2011; Knöller and Schubert 2010; Scholl et al. 2006; Sueker 2003). Figure 5.9c shows that leachate water is clearly enriched in $\delta^{2}H$ due to extensive methanogenes. Figure 5.9b shows that $\delta^{18}O$ is also enriched for some of the leachate samples. However, this strong fractionation is only observed in leachate groundwater collected from wells located below the waste pile or within 25 m of the former waste piles. This mean that strong methanogenes only occurs in the vicinity of the decommissioned landfill and that deuterium cannot be used to trace the leachate plume extent since a strong isotopic fractionation is necessary to observe this phenomenon (Clark 1997). Bandeira et al. (2008) investigated tritium as a tracer for leachate contaminated water, as many sources of tritium are found in landfills (Bandeira 2007; Verhagen et al. 1999; Robinson and Gronow 1995). Tritium concentrations in leachate are often greatly above those observed in global precipitation; concentrations up to 105 TU were recorded by Verhagen et al. (1998). In this study, tritium concentrations range from 100-600 TU in leachate groundwater. However, tritium concentrations for groundwater containing attenuated leachate were similar to natural groundwater (Figure 5.9d and 5.9e). Tritium concentrations in groundwater within recharge zones are around 10 TU, while discharge areas have values below the detection limits of 0.8 TU. Therefore, tritium cannot be used to trace the extent of the leachate plume either. However, tritium can be used as a good tracer in the natural system to investigate recharge and discharge zones.

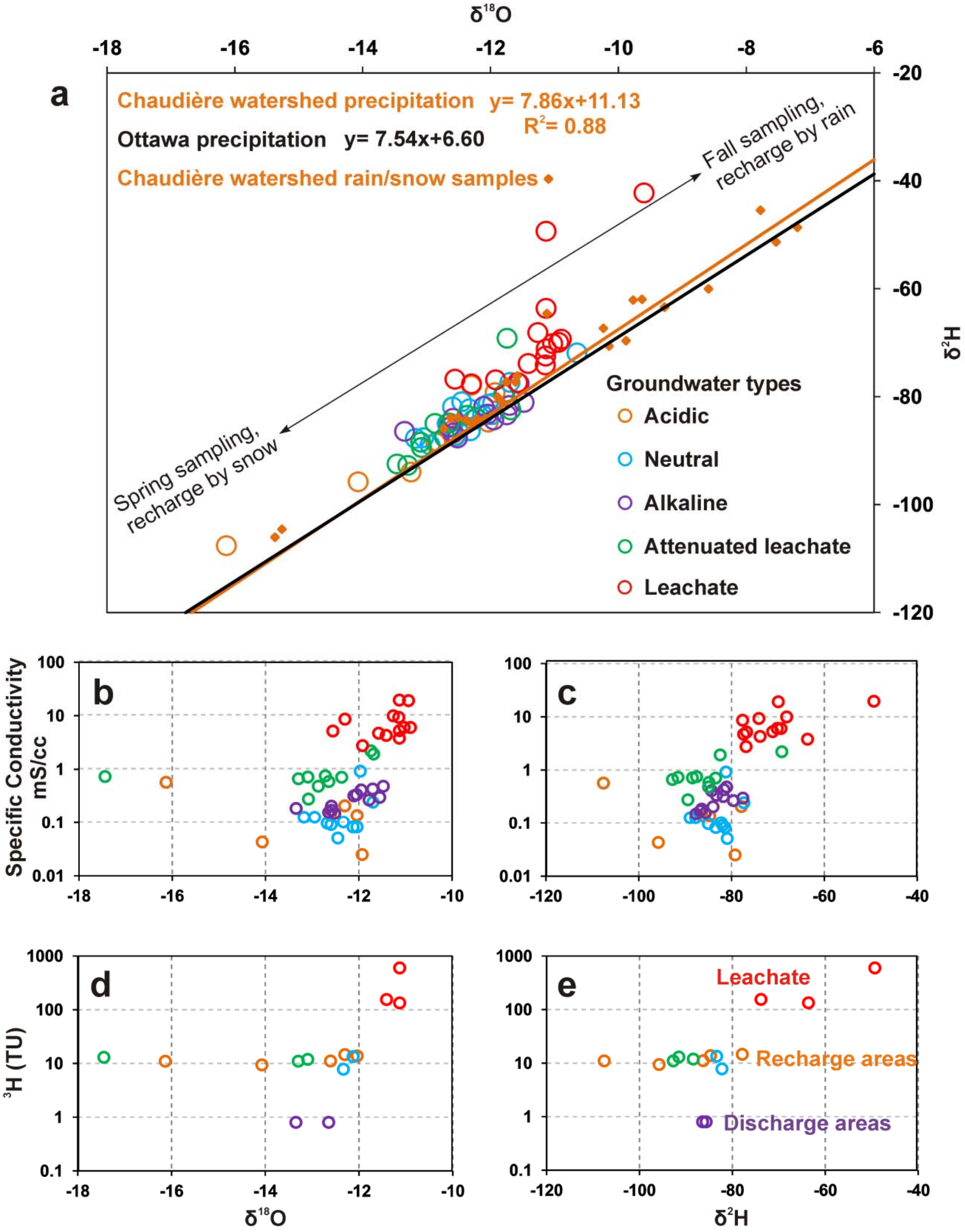


Figure 5.9 : Groundwater isotopes

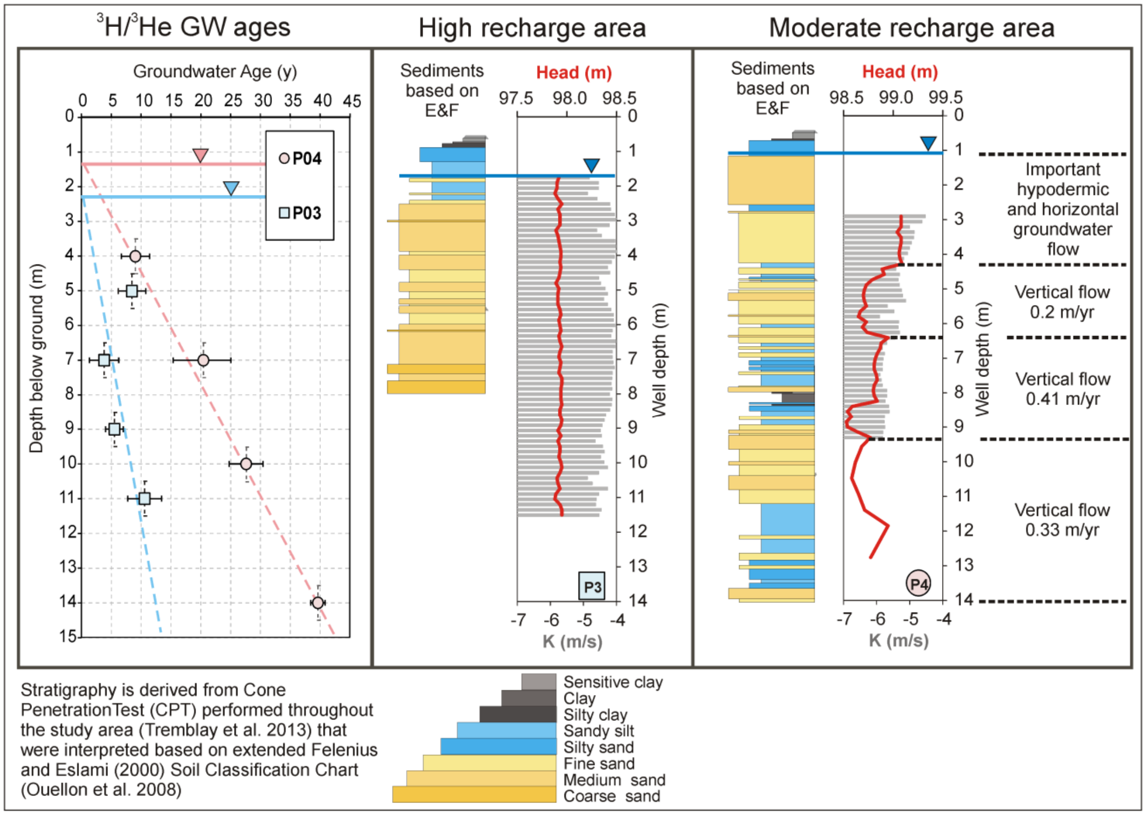
a) Groundwater δ^2H and δ^{18}O for the study area compared to meteoric water lines (orange dots); b) relation between groundwater conductivity and δ^{18}O; c) relation between groundwater conductivity and δ^2H; d) relation between ^{3}H and δ^{18}O and; e) relation between ^{3}H and δ^2H

The tritium-helium ratio method can be used to date relatively young groundwaters (Murphy et al. 2011; Solomon et al. 1993, 1992; Solomon and Sudicky 1991; Schlosser et al. 1989), so it is applicable in shallow granular aquifers with limited residence time. Such conditions are found in the study area in the main recharge zones at higher elevation . Therefore, $^3H/^3He$ were used for groundwater dating, outside the landfill property limits, at the two main recharge location identified with well hydrographs (PO4 and PO3; Figure 5.7). Depth for $^3H/^3He$ measurements at P04 and P03 were selected based on the stratigraphy inferred from Cone Penetration Tests (CPT) and high-resolution data from packer slug tests (hydraulic conductivity profiles and hydraulic heads) (Figure 5.10). Hydraulic measurements were made in fully-screened wells whereas $^3H/^3He$ measurements were made at 4 different depths in nests of discrete-screen (1 m) observation wells for both locations.

Figure 5.10 shows that despite their similar hydrographs (Figure 5.7), water does not infiltrate at the same rate at the two locations. Infiltration of water is slower at P04 partly due to the heterogeneous stratified nature of the material at this location, which results in low vertical hydraulic conductivity (shown by head losses). Also, during the main recharge events in May following snow melt, regressing slopes are steeper for well hydrograph P04 compared to P03 (Figure 5.7). This and many other factors indicate the presence of important hypodermic flow occurring in the 0-4 m depth interval below ground at P04. However, well hydrographs for P03 and P04 show similar head elevations through time. While the same quantity of water reaches the water table, the same quantity does not contribute to groundwater flow deeper within the aquifer. Groundwater recharge was calculated based on the method suggested by Solomon and Sudicky (1991):

$$R = V_n \cdot \theta. \quad [4]$$

where, R is the recharge rate [L/T], θ is the effective porosity [L^3/L^3], and V_n is a component of the average linear velocity normal to the water table [L/T]. For a unique porosity value of 0.3, recharge at P04 was estimated at 105 mm/y, and 412 mm/y at P03 based on the deepest ages measured. These values are also coherent with the vertical hydraulic gradients measured in the wells, as the same gradients can be recalculate with velocity inferred from age in relation to depth. These values revealed that less water participate to the deep subsurface flow at P04 than at P03.



Stratigraphy is derived from Cone PenetrationTest (CPT) performed throughout the study area (Tremblay et al. 2013) that were interpreted based on extended Felenius and Eslami (2000) Soil Classification Chart (Ouellon et al. 2008)

Figure 5.10 : Recharge conditions at two distinct locations

Comparison of recharge conditions with isotopic data ($^3H/^3He$) in the main recharge areas (bars on the isotopic data indicate the precision of age derived from the data and the sampling interval). Despite their similar hydrographs (Figure 5.7), water does not infiltrate at the same rate at the two locations. Infiltration of water is slower at P04 partly due to the heterogeneous nature of the material at this location, which results in low vertical hydraulic conductivity (shown by head losses)

Ages measured 2 m below the water table at P03 and P04 are suspected to be erroneous. The gas samples for these depths were supersaturated with respect to atmospherically derived gases such as Ne. In addition, other evidences demonstrate that the water at those depths is much younger. At P04, seasonal variabilities of dissolved iron in relation to water table fluctuation are recorded at 4 m depth below ground surface. Moreover, a land use change that occurred in 2007 (a whole area was completely deforested), is traceable in the $\delta^{13}C$ signal and obvious from the elevated dissolved organic carbon (DIC) concentration measured in 2011 (Figure 5.11), meaning that the water has to be less than 4 year old as opposed to the 8 years calculated from the $^3H/^3He$ method.

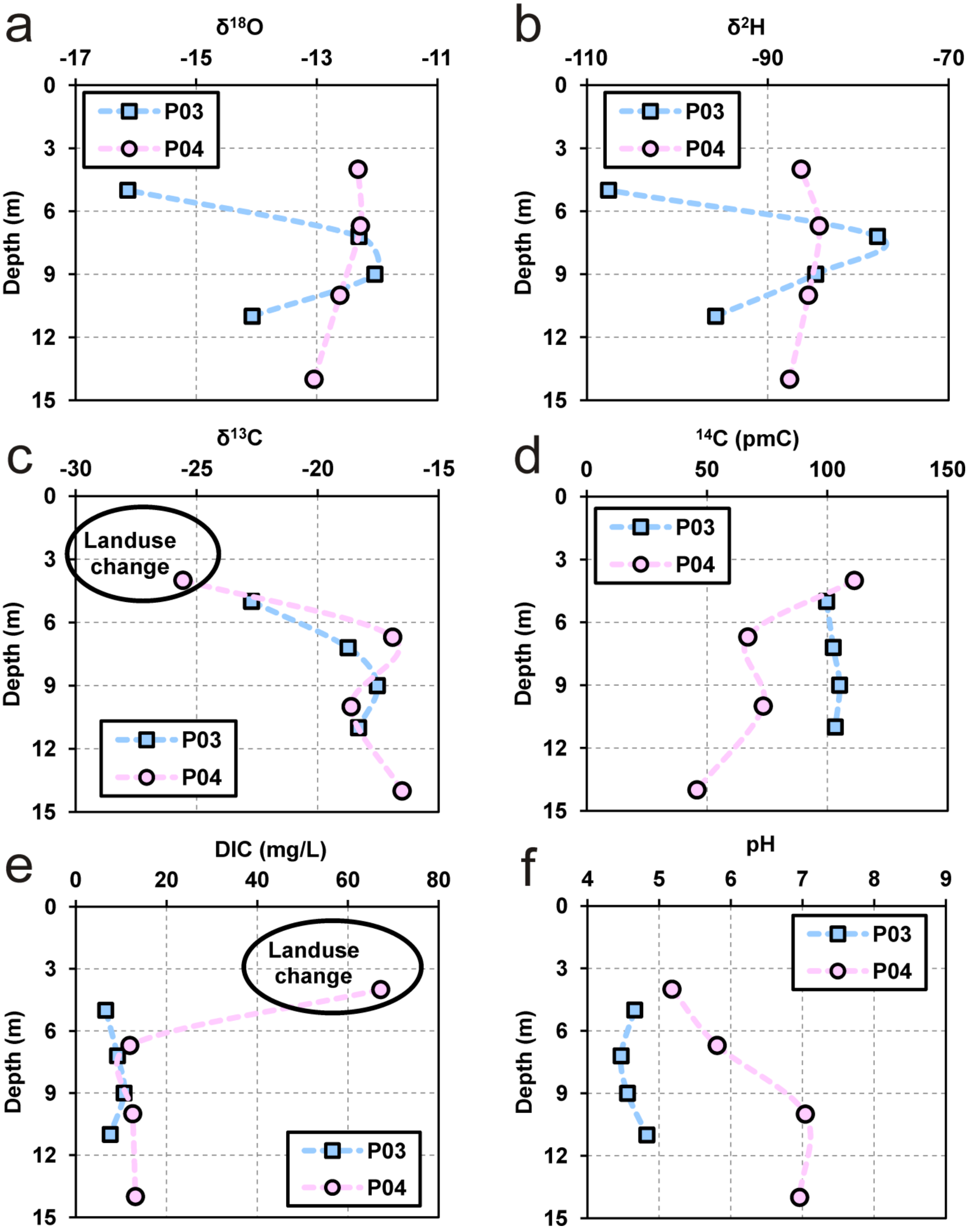


Figure 5.11 : Vertical profiles for different geochemical tracers at P03 and P04

Other studies have documented a lower reliability of $^3H/^3He$ age method for dating very young water (Healy 2010).

Other differences between P03 and P04 can be observed in Figure 5.11 over depth profiles in relation to diverse geochemical measurements made in early spring. Groundwater δ^2H and $\delta^{18}O$ sampled in late spring, after the main recharge event shows a lighter signal at the top of both profiles, 2 m below the water table, because spring recharge is characterized by a lighter isotopic signal. At both locations the signal is enriched at around 7 m depth and becomes lighter with depth afterward. Stronger fractionation effects are visible at P03 where recharge rates are known to be higher and water infiltration through the aquifer faster. Moreover, $\delta^{13}C$ is a good tracer in relation to carbonate dissolution and groundwater evolution in recharge zones, since $\delta^{13}C$ evolves to more enriched values as carbonate is dissolved (Clark and Fritz 1997). Figures 5.11c and 5.11d demonstrate that dissolution occurs under closed conditions at P04 and under open conditions at P03. At P04 $\delta^{13}C$ is diluted by DIC from the carbonate source mineral and the ^{14}C signal is diluted to less modern values (Clark and Fritz 1997). Moreover, DIC values are higher at P04 and pH increases with depth compared to P03. At P03, $\delta^{13}C$ is enriched by about 4‰ and ^{14}C remains modern, as well as DIC and pH. These observations are a first important step in the understanding of groundwater geochemical evolution and residence time in the shallow granular aquifer under study.

5.4.4.4 Baseflow method

In watersheds with gaining streams, groundwater recharge can be estimated from baseflow measurements in streams. At baseflow, it is presumed that the measured flow multiplied by the drained area is equivalent to the aquifer recharge (Healy 2010). However, baseflow discharge does not necessarily directly equate to recharge because of many factors such as: pumping, evapotranspiration and underflow to deeper aquifers that may also be significant (Scanlon et al. 2002). In the study area, the water table intersects the stream channels and all streams are gaining since they are connected to the granular aquifer. Furthermore, recharge is thought to be focused and localized; meaning that most of the recharge takes place in local topographic highs and where subsurface sediments are the most permeable, which implies that recharge is not uniform over the study area. The streams are very small and stream banks are less than 0.5 m high at most locations, except along the Cugnet River where they can reach 2.5 m. Moreover the streams react quickly to rain events. Given such conditions, it is therefore assumed that the baseflow method can provide representative recharge estimates for the study area.

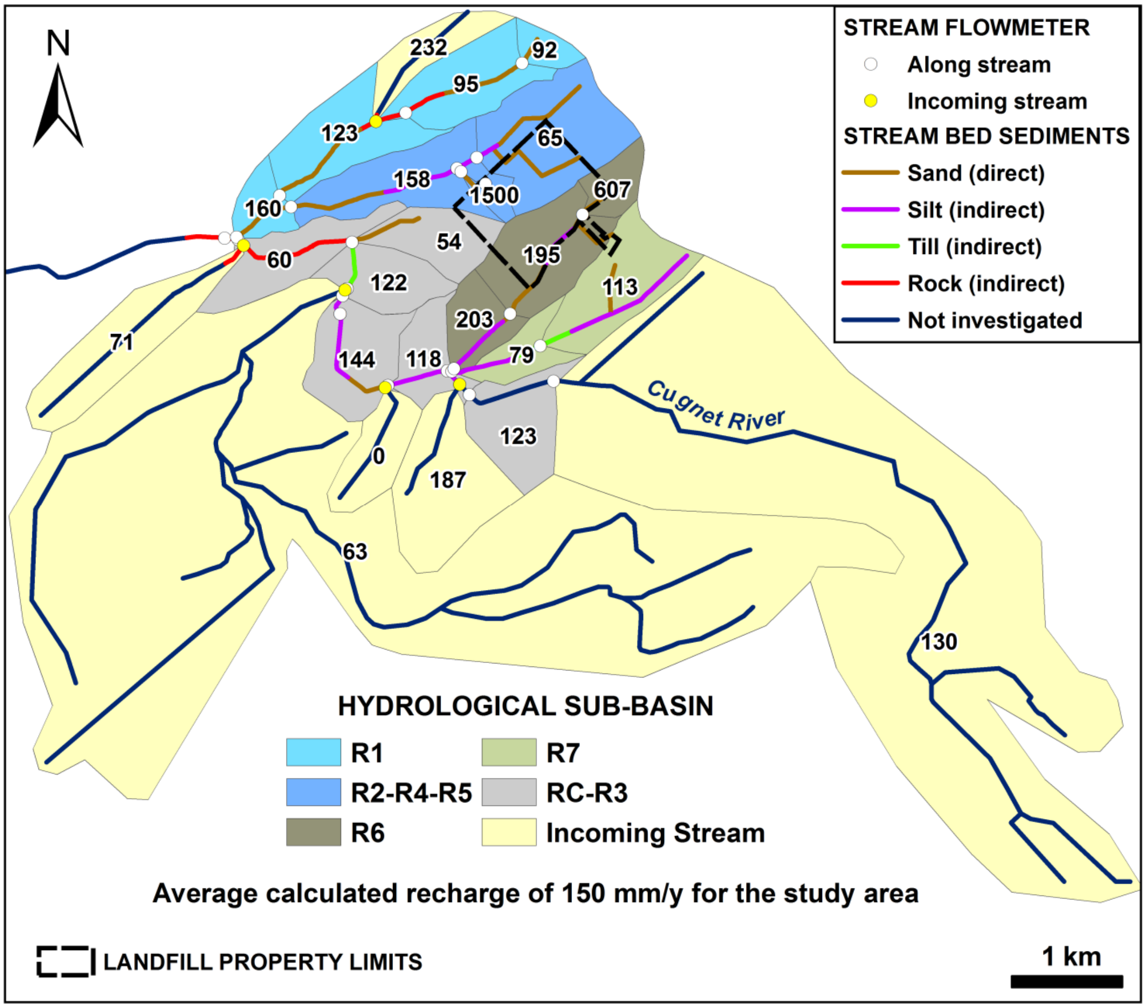


Figure 5.12 : Recharge estimated over the study area from stream baseflow

Recharge estimated over the study area from stream baseflow (flowmeter measurements in September 2010). Sub-watersheds related to streams are distinguished by their color. Numbers correspond to estimated recharge (mm/y) over the sub-watershed area (delineated by grey lines) corresponding to baseflow measurements (white dots are measurements in a stream and yellow dots are flow rates of entering streams). The top graph of Figure 5.8 shows that in September 2009 groundwater levels were at their lowest. The value of 1500 mm/y within the landfill property represents the stream stretch where leachate is released and was not taken into account to estimate the average recharge rate

Baseflow measurements in streams were made at multiple locations (Figure 5.2c) at the lowest of baseflow, as indicated on the well hydrograph for the month of September 2009 (Figure 5.8), and following a one-month period without precipitation. To obtain an estimate of recharge from these baseflow measurements, the topography was first interpolated using ordinary kriging (Davis 2000) in the geostatistical analysis tools of ArcGIS (ESRI 2008). The data points used for the interpolations included continuous indirect data and local direct data. The topographic surface was interpolated at the study area scale of 12 km^2 using high precision (+/- 5 cm) land survey points (point data) and contour lines (continuous data) from a 1:50 000 topographic map sheet previously adjusted with the recorded survey points. A more detailed topographic map of the landfill property was also used to redraw contour lines. The Digital Elevation Model (DEM) was interpolated at a vertical resolution of +/- 0.5 m (when compared with survey data points) (Tremblay et al. 2013). Using the hydrology function of the Spatial Analysis Tools in ArcGIS 9.3, multiple steps were carried out to generate hydrological sub-basins combining: 1) the interpolated DEM; 2) the stream locations and surveyed elevations and; 3) the flowmeter stations along streams (ESRI 2008).

Figure 5.12 shows the recharge estimated for each of the sub-basins considered on the basis of baseflow measurements. It is interesting to note that flow rate decreases in sub-bassins associated with streambed sediments such as till and bedrock. This suggests that at the lowest of baseflow, streams might be losing water to the bedrock aquifers. Also, based on recharge estimates on the map, the stream stretch R6, located south of the former landfill, seems to drain more water than elsewhere. This is in agreement with the surface water analysis of section 5.4.3 demonstrating that a significant leachate discharge occurs into the stream at this location. Overall, the average recharge value for the study area from baseflow measurements is about 150 mm/y at the entire subwatershed scale.

5.4.5 Spatially-distributed recharge rates

Four (4) categories of spatially-distributed recharge rates were inferred from the analysis of all surface and subsurface indicators: high recharge, moderate recharge, low recharge and discharge zones. Figure 5.13 shows the spatial distribution of these zones of recharge rates over the sub-watershed and Table 5.5 lists the characteristics of the four (4) recharge categories.

Tableau 5.5 : Selected indicators and their characteristics relative to recharge conditions

	High Recharge	**Moderate recharge**	**Low recharge**	**Discharge zone**
Superficial indicator				
Topography	Subwatershed highest elevations	Local high	Local intermediate	Local low, near streams or drainage ditches
Unsaturated zone thickness	0.6-2.5 m	0.3-1 m	0.3-1 m	0.1-0.3 m
Shallow groundwater type	Ca-HCO_3 acidic, oxidizing	Ca-Fe-HCO_3 acidic, anoxic	Ca-HCO_3-SO_4 or Ca-HCO_3-Fe, acidic	Ca-Fe-HCO_3 anoxic
Subsurface indicator and well measurements				
TDS inferred from CPT/SMR conductivity data (Tremblay et al. 2013)	0-200 mg/l increases with depth	100-200 mg/l increases slowly with depth	100-200 mg/l increases with depth	constant along profile at ≈250 mg/l
Sediments type and degree of heterogeneity	Homogeneous Coarser sand	Heterogeneous Medium to fine sand to silty sand	Heterogeneous Medium to fine sand to silty sand	Heterogeneous Mostly fine material fewer sand layers
Thermal profiles in wells	Large range (4-12 °C) varies up to 8 m depth	Large range (4.5-12°C) varies up to 8 m depth	4.5-12 °C varies up to 4 m depth	6-8.5 °C varies only at the surface up to 3 m depth
Groundwater pH	acidic	acidic to neutral	neutral	alkaline
Vertical hydraulic conductivity and head gradients	K (10^{-4}-10^{-5} m/s) and head gradient constant along the profiles	K (10^{-5}-10^{-6} m/s) and downward flow	K (10^{-5}-10^{-6} m/s) and downward flow 0-5 m downward 5-10 m upward	K (10^{-5}-10^{-6} m/s) and upward flow
Aquifer thickness	5-20 m	15-20 m	<12 m	<12 m
Well hydrographs	Shows spring recharge and rain events	Shows spring recharge and rain events	Small fluctuations and very noisy	Small fluctuations and very noisy
^{3}H and ^{3}H/^{3}He age	Young water at 11 m depth (8 y)	Older water at 14 m depth (40 y)	Tritium data only show values around 11 TU	Tritium not detected above detection limit of 0.8 TU
Recharge evaluation				
Precipitation reaching the water table	420 mm/y	420 mm/y (surficial) and 105 mm/y at depth	50-250 mm/y	0-50 mm/y
Restriction	None	Compact sediment layer at depth 7 m limits vertical flow and hypodermic flow 0-5 m	Reverse groundwater flow at depth, silt or organic matter restraining infiltration	Hypodermic flow 0-10 cm, water does not reach 1.5 m depth, discharge zones
Average total recharge evaluated to participate to groundwater flow	420 mm/y	260 mm/y	125 mm/y	30 mm/y
Area*	13%	4%	63%	20%
Average	150 mm/y as illustrated by the base flow method shown on Figure 5.12			

*: the total study area covers 12 km^2.

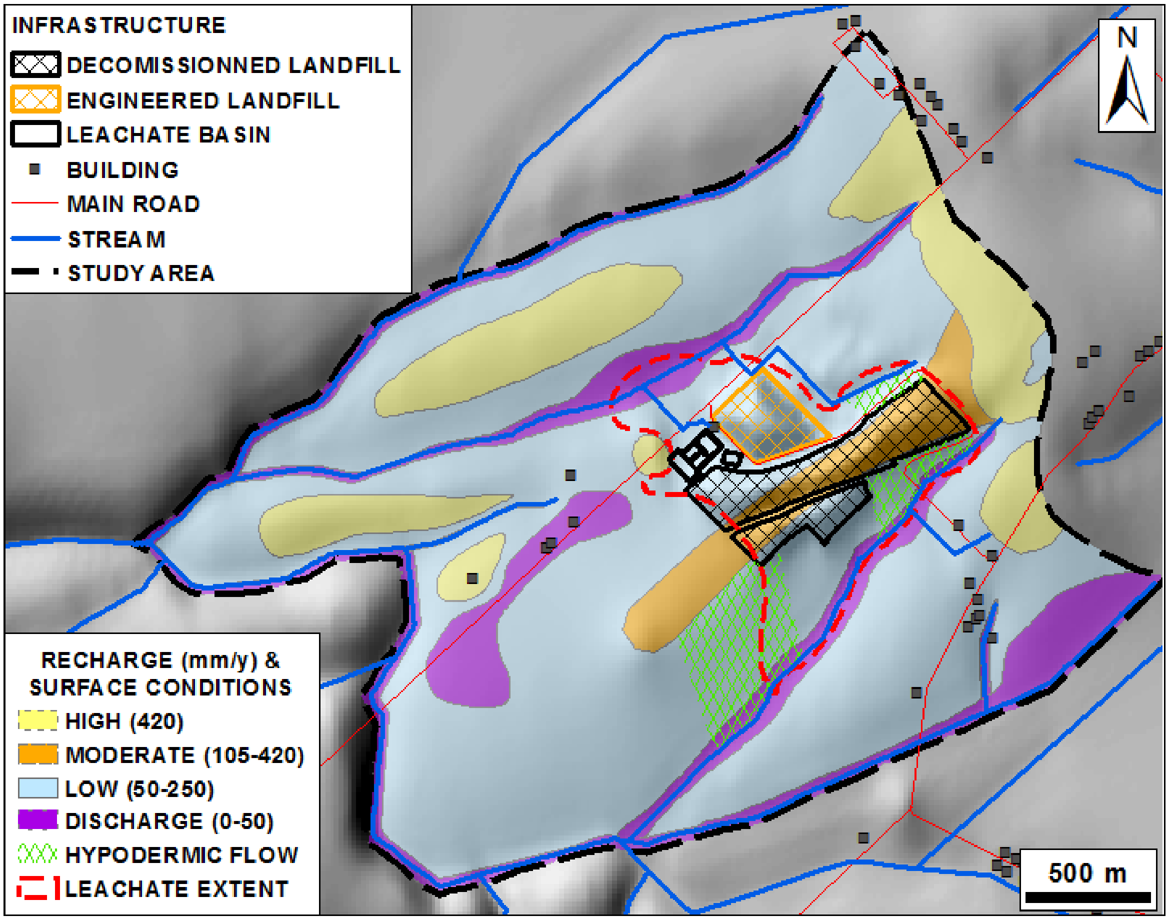


Figure 5.13 : Spatial distribution of groundwater recharge

Spatial distribution of groundwater recharge and location of infrastructure impairing natural recharge. The map also shows the leachate plume extent and areas where there is an important component of hypodermic flow transporting higher concentrations of leachate to streams. The DEM presented in background provides indications of changes in topography

For high recharge zones, a maximum recharge rate of 420 mm/year was estimated where topography is higher and aquifer sediments are coarser and relatively homogeneous, which occurs mostly at the eastern groundwater divide line and in a few sand mounds on the north and northwest parts of the study area. The homogeneous sand found at these locations was either deposited at a former shoreline or later reworked by wind, which produced a relatively coarse and homogeneous sediment accumulation facilitating water infiltration and groundwater flow.

For moderate recharge zones, a rate of 260 mm/y was assigned to zones where infiltration at surface is impeded from reaching deeper in the aquifer due to the heterogeneous (layered) nature of the material in place, especially the presence of a confining layer at about 7 m depth. Observation well P04, whose conditions are shown in Figure 5.10, shows the typical conditions of moderate recharge zones. The infiltrating water in these zones will in part reach streams or drainage ditches through hypodermic flow and will not flow at depth in the aquifer. In the case of P04, although infiltration in the surficial part of the aquifer may correspond to 420 mm/y, based on well hydrograph (Figure 5.8), the net recharge contributing to flow at depth is reduced to 105 mm/y, based on ^{3}H/^{3}He groundwater age (Figure 5.10).

Elsewhere, in low recharge zones, surface soil, topography and heterogeneous sediments control recharge rates which are estimated to vary from 50 to 250 mm/yr. Therefore a representative value of 125 mm/y was selected for these zones. In discharge areas a value of 50 to 0 mm/yr was estimated. Important hypodermic flow and runoff zones were also identified throughout the study area and the leachate extent was inferred from a previous characterization and later confirmed with shallow groundwater chemistry. An average value of 30 mm/y was selected for the discharge zones.

The spatial distribution of recharge zones is coherent with recharge estimates from baseflow values (Figure 5.12). Furthermore, the integration of mean recharge values in all zones gives up to an average of 150 mm/y for the whole study area which is the equivalent value inferred from baseflow estimated recharge for the study area.

5.5 Discussion

5.5.1 Gains from collecting and integrating multiple shallow direct and indirect indicators

Background information on potential controls on recharge is extremely valuable. The climate, geomorphology (including soil and vegetation) and geology of a site control the location and timing of recharge, and therefore they impact the choice of techniques that can be used for estimating recharge (Scanlon et al. 2011). In this study, the collection and integration of multiple shallow direct and indirect indicators were crucial for: 1) understanding the variability of the study area; 2) planning appropriate field surveys at appropriate locations, as well as 3) defining site-specific conditions. The study also benefited from a detail hydrogeological conceptual model, considering aquifer heterogeneity, developed on the basis of multi-source data collected through a previous integrated characterization (Tremblay et al. 2013). The multiple shallow indicators were used to constrain and refine that initial conceptual model, as well as to select the best recharge quantification methods for the study area.

Each indicator contributed to the understanding of recharge and discharge in the study area. Soil mapping showed its variability and infiltrometer measurements of saturated hydraulic conductivity provided the infiltration capacity of the various soil types, and thus their impact on recharge. However, since the water table is very shallow, it was found that the infiltration of water to the subsurface was also controlled by other factors: local topography, unsaturated zone thickness, aquifer heterogeneity and sediment type as well as, evapotranspiration depending upon climate conditions. These conditions were taken into account for the distributed evaluation of recharge in the study zone. The knowledge of material distribution also guided soil sampling to investigate soil chemistry and its effect on shallow groundwater chemistry and to choose appropriate sites for stream bed sediment sampling.

The relation established between soil chemistry and groundwater chemistry was also used to provide indications of recharge locations and magnitude, as well as groundwater flow conditions. The aquifer was found to be under semi-confined conditions in certain locations due to the presence of a silt layer (1-3 m) at the surface in the lower topographic areas. In these local low topographic areas, both the silt layer and organic soil induce anoxic conditions at the top of the water table. Shallow groundwater chemistry provided indications about these processes and the initial conditions at the water table (anaerobic or oxidizing) as well as the residence time and the water types resulting from the water infiltrating under specific soil types.

For the investigation of discharge areas in streams, hydraulic head measured in standpipe piezometers installed along stream edges, field verification of stream bed sediments and stream water types revealed the hydraulic connectivity between aquifer and streams. The identification of recharge and discharge areas as well as the hydraulic connectivity between the aquifer and the surface water is a prerequisite in the evaluation of contaminant mass discharge to surface water (Milosevic et al., 2012). This additional knowledge of surface-subsurface processes is mandatory in the risk assessment of a former landfill managed by natural attenuation. Streams were found to be hydraulically directly connected to the aquifer mostly in the northern portion of the study area where stream beds are composed of sand and indirectly connected in the south-western portion where stream beds are composed of 1-3 m of compact silt or till. Local leachate discharge zones into streams were identified and mass discharge was evaluated with multiple tracers (chloride, sodium, potassium, ammonium, dissolved organic carbon, etc.). Different sources of leachate emission into streams were identified and differentiated.

An important gain from the combination of indicators was the comprehensive knowledge of recharge conditions and processes leading to heterogeneous geochemical conditions of groundwater. This allowed the correct interpretation of natural as well as leachate-impacted groundwater and surface water conditions. Without such understanding, the presence of confounding factors such as upstream contaminants, sources of leachate or leachate dilution by other water sources within a collection system could have lead to confusion in geochemical and isotopic data interpretation (North et al. 2006).

5.5.2 Recharge estimation methods used and spatially distributed recharge values

Because actual recharge rates are never known with certainty at a given location, the use of multiple recharge estimation methods is beneficial. Groundwater recharge to shallow aquifers is a complex phenomenon since it depends on a multitude of climatic and site-specific factors related to land use and aquifer heterogeneity (Arnold and Allen, 1999). Recharge rates are limited in large part by the availability of water at the land surface, which is controlled by climatic factors, such as precipitation and evapotranspiration as well as by surface geomorphic features. To take into account the spatial and temporal variability of the multiple factors controlling recharge rates, four methods were used: 1) water table fluctuation method, 2) soil moisture balance adjusted to well hydrograph P03, 3) baseflow method and 4) groundwater age dating at two known recharge zones put in context with hydraulic and stratigraphy data. However, each method is limited by inherent assumptions and uncertainty, which are not always quantifiable. Moreover, estimates from multiple methods may not quantitatively reduce uncertainty since consistency in results may not be a reliable indicator of accuracy (Healy 2010). So, even though recharge estimates obtained from the different methods corroborate well, an examination of uncertainty in relation to each method, in the context of the study area, is done in the present section.

The water table fluctuation method was directly applicable to only one of the nine well hydrographs recorded in the study area. For the wells located in lower topographic areas, water levels responded to multiple phenomena and were not necessarily indicative of groundwater recharge and discharge. Healy and Cook (2002) state that if the water is transported away from the water table at a rate similar to recharge, then the water fluctuation method is of little value. For wells located next to streams or drainage ditches, changes in water level following precipitation events can be induced by a rise in water level in the stream or in relation to soil water content or heterogeneous hydraulic properties (Sophocleous 2002). The present study also showed that subsurface conditions also need to be considered in the interpretation of recharge from well hydrographs. While well P04 has a similar hydrograph as P03, it was demonstrated with $^3H/^3He$ groundwater dating that net groundwater that participated to the deeper flow within the aquifer was quite different between the two locations. Low permeability material at P04 led to lateral flow and slower infiltration. Finally, the accuracy of recharge estimates from the water table fluctuation method is improved when long records are available. Only two years of hydrograph data were available for the study, which limited the interpretation possible from hydrograph data alone.

The daily soil moisture balance (SMB) method matched to well hydrograph P03 seemed a good recharge estimation method. It is based on readily available weather data and is simple to use. However, results are very sensitive to hydrological input parameters, which are thus also better estimated from daily SMB matched to hydrographs. Ideally, the application of daily SMB requires long-duration hydrographs to get representative hydrological parameters but it also takes into account the natural variability in yearly recharge (Lefebvre et al. 2011). SMB calculations are simple, but there is a relatively large uncertainty for many of the input parameters so that results are uncertain as well. However, estimation of long-term water level trends can be done using weather data, thus verifying that hydrological parameters used to match well hydrographs were coherent (Lefebvre et al. 2011). The daily SMB method could also be improved by using other methods for estimating hydrological parameters, especially potential evapotranspiration (Rushton et al. 2006).

The baseflow method involves multiple flowmeter measurements in streams at baseflow and extensive study on soil and surface water to assess the connectivity of streams to the aquifer.

Although conceptually simple, this method was difficult to implement due to the conditions found in the study area. In the study area, streams were re-dug in the 60's and multiple drainage ditches were excavated. The streams are obstructed by numerous beaver dams and fallen trees. A lot of leaves and organic matter accumulate in the stream beds as stream flow is very low in general. The Cugnet River, which is the main stream, has a maximum width of 10 m wide and depth of 1.3 m. It was thus very difficult to find good locations for flow measurements. The stream cross-section areas were measured as precisely as possible and multiple flow measurements were made for each section. However, at some locations the streams were almost completely disconnected from the aquifer during baseflow conditions. In addition, recharge calculations based on flow measurements involve the evaluation of the area drained. A DEM was used to evaluate these zones, so that the uncertainty in the result is based on the model used. While the precision of the recharge rate obtained for each sub-basin is a somewhat subjective, the average value calculated for the whole area is realistic and corroborates with the other measurements.

Environmental tracers were critical tools for understanding groundwater flow through the subsurface as well as groundwater recharge (Sanford 2011). In the present study, geochemical analyses of shallow groundwater and soils helped confirm that soil redox conditions have an important control on the recharge water chemistry. It also demonstrated that important reactions with regards to sulfate and iron have taken place in relation with the water table fluctuations in the surficial portion of the aquifer and that these mechanisms can contribute to the natural attenuation of the leachate plume. Moreover the shallow chemistry revealed that recharge patterns are associated with a characteristic geochemical groundwater signature. Furthermore, recharge conditions are characterized by distinct temperature profiles and distinct TDS profiles inferred from CPT resistivity values (Tremblay et al. 2013). ^{18}O/^{2}H results were also useful to identify the origin of recharge water and its behavior along vertical depth profiles. ^{3}He data served to identify the different natural context of recharge and discharge as well as confirming the leachate plume signature. The ^{3}H/^{3}He age dating method was also useful to understand further the recharge zones and differentiate groundwater evolution in the different context identified. ^{13}C, ^{14}C, DIC and pH values along the same profiles were useful to understand groundwater geochemical evolution and residence time in the shallow granular aquifer under study. Overall, although used qualitatively (besides ^{3}H/^{3}He age), geochemical tracers were important independent indicators of both the location and magnitude of recharge, besides characterizing the effect of soil types and context on groundwater geochemistry, as discussed in the previous section.

Although significant uncertainty is associated with each recharge assessment method, the methods used and the time frame considered for recharge estimation demonstrated that temporal and spatial variability of recharge in the study area has a major impact on groundwater flow. The comparison of recharge values obtained from the four methods helps understand spatial and temporal recharge patterns and demonstrates that recharge rates were mostly influenced by topography, heterogeneity and nature of the subsurface material and vertical groundwater flow direction.

5.5.3 Implication for conceptual model validation and numerical modeling

A conceptual model has to be reviewed and revised as additional data and analyses provide new insight to the hydrologic system (Zheng and Bennett 2002; Bredehoeft 2005). For the study area, an initial conceptual model defined the aquifer heterogeneity, boundary conditions and the extent of the landfill leachate plume (Tremblay et al. 2013). The use of indicators of recharge and discharge documented in this paper significantly enhances the conceptual model: 1) it provides an understanding of recharge mechanisms, the spatial and temporal distribution of recharge over the study area and its magnitude; 2) it characterizes the effect of soil and context on the geochemistry of shallow groundwater, showing that quite variable geochemical conditions can occur in shallow groundwater, which will influence leachate attenuation; 3) it provides indications of the location of groundwater discharge in streams and of the effect of direct leachate discharge in streams compared to discharge from groundwater. The understanding emerging from the study is thus that recharge and geochemical processes are linked and lead to quite variable conditions over the study area. Such variability has to be known to properly interpret both the natural groundwater geochemistry and the natural attenuation of leachate. This improved conceptual model is also important to assess the environmental risks associated to streams, which are the main receptors in the study area. Therefore, the approach proposed in this paper allowed the validation and the refining of the hydrogeological conceptual model already established: 1) by facilitating the understanding of geochemical processes occurring in the shallow subsurface as well as specific to recharge zones and 2) by mapping distributed recharge rates. The refined conceptual model explains groundwater flow conditions resulting from recharge, discharge and the internal hydrostratigraphy and heterogeneity of the aquifer. Groundwater recharge is a key parameter to any groundwater numerical model as it was demonstrated that recharge variability can induce additional dispersion (Kim et al. 2000). Therefore the data obtained from the present study will be considered in the development of a numerical model and initial conditions of distributed recharge will be applied.

5.5.4 Implications for site management and future work

For practical and economic purposes, public organizations managing waste sites seek to extend the operation life of landfilling at a same location for as long as they can. This could imply expansion by deforestation, larger infrastructure to accommodate larger amounts of waste (i.e. larger leachate collection basin, garage for machinery), etc. To support site management decisions, especially concerning leachate natural attenuation, approaches such as those described in this paper can provide a thorough understanding of recharge and discharge processes, groundwater flow and residence time as well as the connectivity between aquifers to surface water, which are essential conditions to understand and to assess the risk to receptors.

Although the present study provides indications that natural attenuation of the leachate plume is such that groundwater and surface water are not significantly impacted, some issues remain to be resolved. Further work is needed first to investigate geochemical heterogeneity of aquifer sediments and groundwater in more detail, and their implications on flow paths and leachate migration and attenuation. Numerical modeling of flow and mass transport is also needed to quantitatively verify the conclusions about flow and natural attenuation conditions. Results from the present study will put into context the study of geochemical processes and natural attenuation and it will ensure the use of representative conditions in the numerical model.

5.6 Conclusion

The study focused on a 12 km^2 subwatershed within which a decommissioned landfill is located over a shallow heterogeneous aquifer. Multiple indicators were investigated to better understand recharge and discharge processes and thus complement and improve a previously developed initial conceptual model of the aquifer system. The indicators considered in this study include: topography, multiple physical and chemical soil properties, groundwater and surface water chemistry, stream bed sediments, hydraulic heads and gradients measured in shallow wells located on stream banks, and flowmeter measurements in streams at baseflow. These indicators are combined with groundwater isotopic data (^{18}O, ^{2}H, $^{13}C/^{14}C$, $^{3}H/^{3}He$) and high-resolution hydraulic head measurements made in fully screened wells to estimate recharge rates, groundwater flow directions and discharge rates as well as their impacts on the migration and attenuation of a leachate plume. The collection and integration of multiple shallow direct and indirect indicators were crucial for: 1) understanding the variability of the zone under investigation; 2) planning appropriate field surveys at the right location as well as 3) defining the site specific conditions. Recharge rates were evaluated with four methods adapted to the study area. Soils were found to have only a small impact on recharge variability in the area, because most soils are poorly drained and the unsaturated zone is very shallow (0.1-1.5 m). Recharge rates were mostly influenced by local topography, heterogeneity and nature of the subsurface material. The use of indicators of recharge and discharge documented in this paper significantly enhanced the conceptual model: 1) it provides an understanding of recharge mechanisms, the spatial and temporal distribution of recharge over the study area and its magnitude; 2) it characterizes the effect of soil and context on the geochemistry of shallow groundwater, showing that quite variable geochemical conditions can occur in shallow groundwater, which will influence leachate attenuation; 3) is provides indications of the location of groundwater discharge in streams and of the effect of direct leachate discharge in streams compared to discharge from groundwater. This study has implication for the understanding of groundwater flow and boundary conditions. These results will be integrated into a numerical model of groundwater flow and will guide recommendations for land use and risk management of the decommissioned landfill.

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Chapitre 6

Geochemical Heterogeneities of Aquifer Sediments, Groundwater and Landfill Leachate at the Sub-watershed Scale

TITRE FRANÇAIS :

Hétérogénéité géochimique des sédiments d'un aquifère, de l'eau souterraine et du lixiviat d'un site d'enfouissement à l'échelle d'un sous-bassin versant

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CONTRIBUTIONS:

Laurie Tremblay a planifié et coordonné l'acquisition des données de terrain. Était personnellement responsable de l'acquisition, du traitement et de l'interprétation des données. Au niveau de l'article, elle a réalisé les figures et écrit la version préliminaire de l'article.

René Lefebvre a coordonné le projet, encadré les étudiants impliqués et révisé l'article.

Vincent Cloutier a encadré l'étudiante par rapport aux aspects géochimiques et a révisé la version préliminaire de l'article.

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RÉSUMÉ

Cette étude a pour but de décrire les conditions géochimiques qui pourraient influencer la migration et l'atténuation d'un panache de lixiviat dans un sous-basin versant de 12 km^2 à l'intérieur duquel est situé un ancien site d'enfouissement sanitaire sans base imperméable qui émet un panache de lixiviat dans un aquifère granulaire peu profond. L'atténuation naturelle d'un panache de lixiviat est influencée par la géochimie naturelle de l'eau souterraine dans laquelle le lixiviat est émis. Cette géochimie est étroitement liée aux propriétés physiques et chimiques des sédiments qui composent l'aquifère. Cette étude a pour but de décrire les conditions naturelles qui influencent l'évolution géochimique du panache de lixiviat et de délimiter l'étendue de la contamination dans l'eau souterraine. Les travaux sont basés sur l'interprétation des données de géochimie du lixiviat et des eaux souterraines dans la région d'étude. De plus, considérant l'importance du lessivage des sols sur la géochimie de l'eau souterraine, cette étude propose un plan de travail pour caractériser, efficacement et à moindre coûts, les propriétés physiques et géochimiques des sédiments d'un aquifère. Un outil de fluorescence X portable a été utilisé pour analyser la géochimie des sédiments. La géochimie des eaux souterraine a aussi été caractérisée grâce à une gamme étendue de paramètres analytiques. La géochimie des sols et de l'eau souterraine a été interprétée avec les méthodes statistiques multivariées d'analyse par composante principale et de classification hiérarchique ainsi qu'avec d'autres méthodes graphiques. Les données géochimiques et leur interprétation ont permis de 1) préciser le modèle conceptuel de l'écoulement de l'eau souterraine dans le système aquifère; 2) comprendre les processus géochimiques le long des voies d'écoulement; et 3) d'identifier la signature du panache de lixiviat dans l'eau naturelle et ses conditions d'atténuation naturelle. Les propriétés physiques et géochimiques des sédiments peuvent être liées à leur historique de déposition et ces propriétés ont fourni des indications sur le rôle de la chimie des sédiments sur les types d'eau souterraine, particulièrement au niveau du cycle du carbone. Cette étude démontre qu'il existe une grande hétérogénéité au niveau des sédiments, ce qui influence aussi la variabilité des types d'eau souterraine que l'on retrouve dans l'aquifère. Cette étude démontre aussi que ces conditions sont influencées par l'émergence d'eau souterraine provenant de l'aquifère rocheux fracturé sous-jacent à l'aquifère granulaire. Même si la géochimie de l'eau est hétérogène, il a été possible de définir les conditions naturelles dans l'aquifère et de reconnaitre l'eau influencée par l'émission de lixiviat. Cette géochimie naturelle variée fait en sorte que l'atténuation du panache

de lixiviat est différente selon les conditions oxydantes ou plus réductrices retrouvées le long d'une voie de migration. Ces résultats vont être intégrés à un modèle numérique de l'écoulement de l'eau souterraine et de la migration de lixiviat pour assurer la compréhension nécessaire à la gestion du risque environnemental de l'ancien site d'enfouissement sanitaire.

MOTS-CLÉS : Hétérogénéité géochimique, caractérisation hydrogéologique, aquifère granulaire, évolution de l'eau souterraine, site d'enfouissement, lixiviat.

ORIGINALITÉ ET PERSPECTIVE:

Plusieurs études ont fait l'utilisation des méthodes multivariées ou autres méthodes statistiques pour interpréter des données géochimiques de l'eau souterraine en lien avec un panache de lixiviat qui migre dans un aquifère granulaire. Plusieurs études ont aussi investigué les sols et les ont mis en relation avec la géochimie de l'eau. Par contre, peu d'étude ont considéré comment l'hétérogénéité des sédiments et de l'eau souterraine affecte l'évolution géochimique d'un panache de lixiviat lors de sa migration à l'échelle d'un sous-basin versant. Selon les auteurs, les approches décrites dans l'article ont les aspects novateurs et originaux suivants par rapport aux autres études sur ce sujet :

Échelle d'étude considérée

L'étude prend en considération un aquifère granulaire peu profond d'un sous-bassin versant de 12 km^2, incluant les zones sources et les récepteurs, pour y définir autant les conditions naturelles que les conditions du panache de lixiviat. De plus, l'étude prend aussi en considération les conditions à l'échelle régionale dans laquelle la région d'étude est située. L'étude à l'échelle régionale comprend des résultats de la géochimie de l'eau souterraine de l'aquifère rocheux pour toute la région du basin versant de la rivière chaudière. Ces données régionales ont permis de conclure que l'eau de l'aquifère au roc émerge dans l'aquifère granulaire sus-jacent à des endroits dans la région investigué ou le till est aminci. Cet apport d'eau du roc a des effets sur la géochimie de l'eau dans l'aquifère granulaire de même que sur l'évolution géochimique du panache de lixiviat. Peu d'étude sur le lixiviat ont été réalisés à une échelle de sous-bassins versants ou pris en considérations les contextes de différentes échelles d'investigation.

Méthodologie pour caractériser les sédiments

La méthodologie développée a permis de caractériser les sédiments de façon efficace tout en minimisant les temps d'attente et les coûts élevés pour certaines analyses de laboratoire. L'utilisation d'un XRF portable pour définir la géochimie des sédiments n'a pas été utilisée par d'autre étude en hydrogéologie. La compréhension de la géochimie des sols en contexte avec le type de matériaux aide aussi à comprendre la provenance des sédiments et l'historique de déposition ainsi que leur influence sur l'eau souterraine.

Reconnaissance des conditions géochimiques naturelles de l'eau souterraine et leur influence sur l'évolution du panache de lixiviat

L'ACP, qui est fréquemment utilisée pour élaguer des données, a été utilisé pour identifier les processus géochimiques et les origines de l'eau. L'identification des processus avec les graphiques géochimiques a permis de définir les deux voies distinctes de migration du lixiviat en conditions oxydantes et réductrices, qui influencent l'atténuation naturelle du lixiviat et son devenir. De plus les analyses ont démontré que les conditions géochimiques sont très hétérogènes. L'utilisation des isotopes du carbone a permis de faire le lien avec la nature des sédiments et d'ajouter à la compréhension en faisant le lien entre la géochimie des sédiments et celle de l'eau souterraine.

Intégration originale des divers indicateurs pour permettre une meilleure compréhension d'un système d'écoulement et pour la validation du modèle conceptuel hydrogéologique

L'originalité de cet article se retrouve dans l'approche de caractérisation géochimique des sédiments et de l'eau souterraine et dans la façon d'exploiter ces données afin de déterminer l'influence de l'hétérogénéité géochimique sur la migration et le devenir d'un panache de lixiviat. L'ensemble des données permet de préciser le modèle conceptuel du système aquifère granulaire peu profond déjà développé sur la base d'une caractérisation hydrogéologique intégrée. Ceci a des implications pour la compréhension du devenir du panache et sur la gestion du risque pour les milieux récepteurs. Cette phase de travaux de caractérisation sert aussi à valider le modèle conceptuel qui servira de base au modèle numérique. Les données géochimiques des éléments inorganiques et des traceurs vont aussi servir à contraindre le modèle numérique d'écoulement et les prédictions du transport de masse faites à l'aide de ce modèle numérique.

ABSTRACT:

This study aimed to describe the geochemical conditions that could influence leachate plume migration and attenuation in a 12 km^2 subwatershed encompassing a decommissioned and unlined landfill. Natural attenuation of a landfill leachate plume is influenced by the natural groundwater geochemistry of the aquifer in which it migrates. This natural geochemistry is related to the aquifer sediment's physical and chemical properties. Considering the importance of soil leaching on groundwater geochemistry, this study proposes a workflow to efficiently characterize aquifer sediment physical and chemical properties. A portable X-Ray fluorescence device was used for its ability to accurately analyze sediment chemistry. Groundwater geochemistry was also characterized with a full range of parameters. Soil and groundwater geochemical parameters were interpreted with multivariate statistical methods and the use of geochemical graphs. Geochemical data allowed 1) refinement of the aquifer system conceptual model, 2) better understanding of natural geochemical processes along flow paths, and 3) the identification of the leachate plume signature. Physical and chemical sediment properties were related to their depositional history and provided indications of the role of sediment chemistry on the groundwater chemical signature, including the carbon cycle. A high level of geochemical heterogeneity was seen in the aquifer sediments, which partly controls varied groundwater geochemical conditions. The study also showed that these conditions are influenced by groundwater upflow from the regional fractured rock aquifer into the granular aquifer. Although groundwater geochemistry was heterogeneous, it was possible to establish natural background conditions and recognize zones where groundwater quality was affected by leachate. These varied geochemical conditions lead to different natural attenuation processes of the leachate plume. Results will be used to validate a conceptual model which is the basis for a numerical model of groundwater flow and mass transport that will be used to predict the fate of the leachate plume.

KEYWORDS: Geochemical heterogeneities, field characterization, granular aquifer, natural groundwater evolution, landfill leachate plume.

6.1 Introduction

Currently, numerous decommissioned and unlined landfills remain solely managed by natural attenuation (MNA), relying on processes such as dilution, sorption, and microbial degradation to reduce the mass of contaminants to less harmful forms or concentrations (Christensen et al. 2001, 1994). Since not all contaminants can be remediated by natural attenuation or be decreased to acceptable concentration, contaminant mass emissions from such unlined waste disposal sites can pose a threat to shallow aquifers for decades to centuries (Milosevic et al. 2012; Bjerg et al. 2011, 2003; Cozzarelli et al. 2011; Park et al. 2008; Kjeldsen et al. 2002; Christensen et al. 2001). In the last decade, numerous scientific advances have been made regarding the occurrences and rates of several attenuation processes (physical, chemical and microbial) within plumes or at their fringe, which control the transport and ultimate fate of contaminants in aquifers. These studies demonstrated the complexity and variability of reactions occurring as plumes migrate through different types of geological and geochemical conditions (Robertson et al. 2011; Sudicky et al. 2011, 2010; Cozzarelli et al. 2011; Lorah et al. 2009; Bjerg et al. 2011, 2003; USEPA 2008, 2007; van Breukelen et al. 2003; van Breukelen 2003; Kjeldsen et al. 2002; Christensen et al. 2001, 1994). It has also been established that spatial variability in observed contaminant concentrations results from geological and geochemical heterogeneities (Anderson and McCray 2011; Allen-King et al. 2010; USEPA 2008, 2007; De Marsily et al. 2005). Some authors have even concluded that heterogeneity not only controls the physical spreading of contaminants by dispersion and advection but also influences chemical reactions and bacterial attachment rates as well as remediation effectiveness (Anderson and McCray 2011; Allen-King et al. 2010).

The natural geochemical composition of groundwater is closely related to weathering and dissolution of minerals within the aquifer, which releases some of the dissolved components found in groundwater (Appelo and Postma 2007). From recharge to discharge areas, rain water infiltrates the aquifer, reaches the water table and, as groundwater flows within permeable material, some minerals are dissolved, others precipitate and various chemical reactions occur (Hounslow 1995). Therefore, to comprehend a hydrogeological system, it is essential to acquire a good knowledge of the geochemistry of the aquifer matrix as well as the geochemistry of groundwater along flow paths. Moreover, redox processes affect groundwater quality, contribute to either the degradation or preservation of anthropogenic contaminants and lead to the presence of some undesirable components in groundwater, such as dissolved iron (McMahon and Chapelle 2008). Therefore the redox potential (ORP) of the aquifer matrix has to be investigated. While soil ORP is not the unique factor controlling the reactions that take place in an aquifer (Medina 2001), which are also controlled by pH, geochemistry, porosity and organic carbon content, ORP can provide a good indication of the main water and sediment redox species, and thus of the attenuation process that could take place in an aquifer (Appelo and Postma 2007; Patrick et al. 1996). The oxidation capacity (OXC) of a soil thus plays an important role in natural groundwater geochemistry and migration of pollutants in that groundwater. For example, as a leachate plume migrates, a slow rate of iron and manganese reduction would allow larger zones of methane and sulfate to form, whereas a fast rate would limit the development of such zones. Furthermore, one has to consider that redox conditions can vary substantially along different migration pathways of a leachate plume in a heterogeneous setting (van Breukelen 2003; Medina, 2001).

This study aims to identify what controls natural geochemical conditions (of sediments and water) in an aquifer, so as 1) to distinguish the leachate geochemical signature in the natural environment and 2) to infer the impact of these geochemical conditions on the leachate plume migration and attenuation. The study was carried out in a shallow granular aquifer, within a 12 km^2 subwatershed where a decommissioned landfill is located. The objectives of the study are to understand: 1) the natural groundwater geochemical evolution; 2) the relations between soil and sediment chemistry and natural groundwater geochemistry; and 3) how the natural groundwater geochemistry affects the natural attenuation of the leachate plume based on key geochemical tracers along its migration path. The purpose of the study is also to develop an efficient sediment characterization approach that facilitates the identification of relations between sediment and groundwater geochemistry. This geochemical investigation is the third phase of an integrated aquifer characterization program (Tremblay et al. 2013; Tremblay 2013). Results will be used to validate a conceptual model of the aquifer system, which is the basis for a numerical model of groundwater flow and mass transport. The numerical model will itself be used to quantitatively predict the fate of the plume and help support recommendations in relation to the environmental management of the landfill, especially the monitored natural attenuation (MNA) of the leachate plume.

6.2 Background

The 12 km^2 study area is located 35 km south of Quebec City, Canada (Figure 6.1). In the center of the study area, a decommissioned unlined landfill and an engineered municipal landfill are located on a 1 km^2 property managed by the “Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière” (RIGDCC), a public organization. The engineered landfill is in operation and meets current waste management standards. The decommissioned unlined landfill was in operation between 1974 and 1997, during which 9 million tonnes of waste from varied sources were deposited directly over permeable sands. From 1993 to 1997, the waste piles were reshaped and an impermeable cover was placed on its surface (RIGDCC 2013). Prior to closure, landfill leachate was detected in surrounding creeks (Gauthier 1993a). Since its closure, the decommissioned landfill is solely managed by natural attenuation, although no studies have verified the effectiveness of this management method.

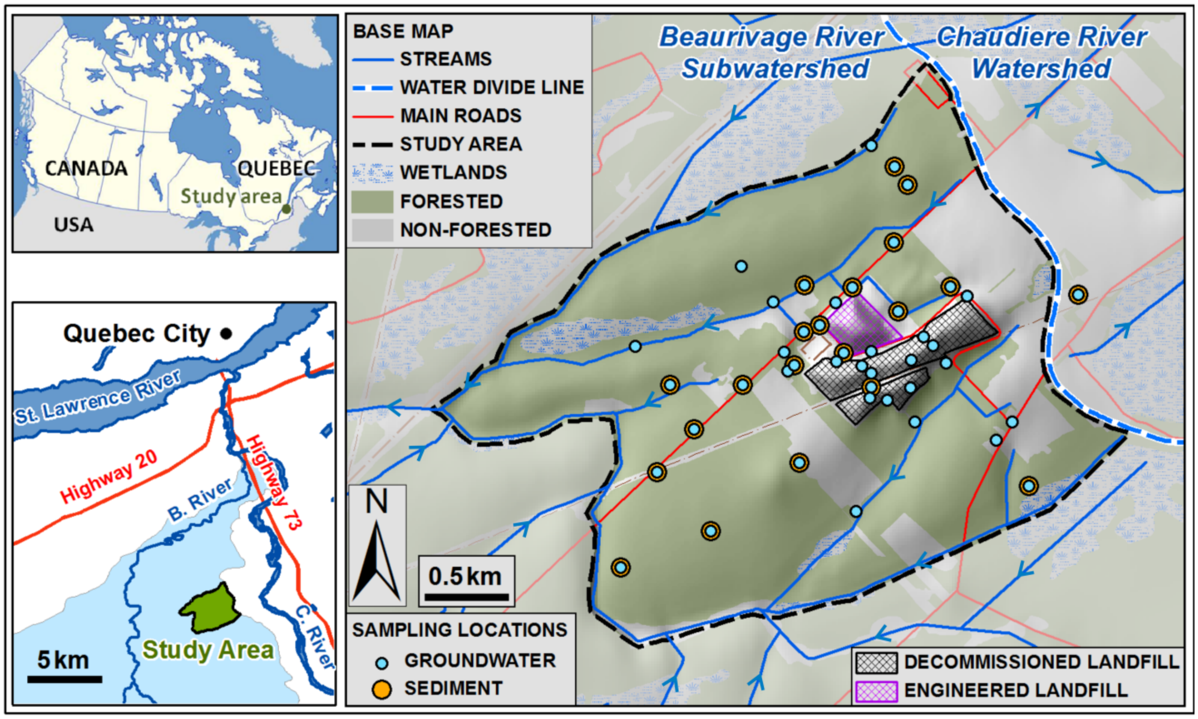


Figure 6.1 : Study area location, physiography and land use

On the map at the bottom left: B. River and C. River respectively stand for Beaurivage River and Chaudiere River. On the same map, the Beaurivage River subwatershed is shaded in blue. The map on the right shows the study area and the groundwater and sediment sampling locations (Modified from Tremblay 2013)

In 2006, our study was initiated to investigate the effectiveness of the natural attenuation of the leachate plume emitted by the decommissioned landfill within the granular aquifer. In 2007, an integrated multidisciplinary aquifer characterization was carried out. The extent of the study area was defined so as to encompass leachate source zones, near the decommissioned landfill, and receptors, which are mainly represented by streams in the study area. Tremblay et al. (2013) describe the characterization program designed on the basis of the specific hydrogeological setting of the study area, which includes three phases. The first phase involved : 1) surface geophysics including a) ground penetrating radar (GPR) and b) electrical tomography (ERT); 2) direct-push methods including a) cone penetration tests (CPT) with soil moisture resistivity (SMR), b) soil sampling and c) installation of fully-screened observation wells in the granular aquifer; 3) borehole operations including a) flowmeter measurements while pumping (Paradis et al. 2011), b) high vertical resolution (15 cm) packer slug tests, c) single well interference slug tests to assess vertical hydraulic conductivity (Paradis and Lefebvre 2013) and d) multilevel measurements of geochemical parameters and groundwater sampling. The integration of the field data allowed the definition of aquifer boundaries and thickness, geological heterogeneity of the aquifer material and the extent of the leachate plume. On the basis of the integrated data, an initial detailed conceptual model of the aquifer system was established (Tremblay et al. 2013). The initial conceptual model illustrated the high level of heterogeneity of the aquifer material and the great variability of the aquifer and aquitard surface topography, as well as the non-continuity of some units. A hot spot of higher concentrations of components originating from landfill leachate was also located (Tremblay et al. 2013). The initial conceptual model indicated locations of recharge and discharge zones on the basis of multiple parameters including groundwater temperature, conductivity, pH and dissolved oxygen, the relative topography and vertical hydraulic gradients, fluid conductivity and TDS derived from cone penetration tests (CPT) with soil moisture resistivity (SMR). The initial investigation demonstrated the absence of concentrations above provincial maximum concentration limits outside the landfill property. The second phase of field work further investigated recharge and discharge zones and the possible location of leachate discharge into streams. Spatially distributed recharge rates were inferred from the integration of multiple shallow indicators: $^{3}H/^{3}He$ groundwater ages, general water chemistry, $^{18}O/^{2}H$ on groundwater, well hydrographs, flowmeter measurements in streams at baseflow and an extensive study on soil and surface water connectivity to the aquifer (Tremblay 2013).

The study area under investigation received an average total yearly precipitation of 1200 mm from 1978 to 2012. A maximum recharge rate of 420 mm/year was found to occur at the groundwater divide line, where aquifer sediments are coarser and homogeneous, and in a few sand mounds distributed over the area. Elsewhere, surface soil and heterogeneous sediments control recharge rates estimated to vary from 250 mm/yr to none at discharge areas (Tremblay 2013). Important hypodermic flow and runoff zones were also identified throughout the study area. Streams were found to be directly connected to the aquifer in the northern part of the study area where stream beds are sandy, whereas they are indirectly connected in the southern and western zones where stream beds are composed of 1 to 3 m of compact silt or till. The aquifer is under semi-confined conditions in certain locations due to the presence of a silt layer (1-3 m) at the surface in the lower topographic areas surrounding the sand mounds. The top silt layer and organic soil at the surface of the aquifer induce anoxic conditions at the top of the water table. This paper documents results from the third phase of field and laboratory characterization work aiming to define geochemical properties of aquifer material and groundwater to meet the previously stated objectives of the study.

6.3 Physical and hydrogeologic setting

The study area is easily accessible by primary, secondary and forestry roads (not all indicated on Figure 6.1). The 12 km^2 subwatershed study area was defined on its eastern limit by a groundwater divide between the Beaurivage River subwatershed and the Chaudière River watershed. Elsewhere, limits were based on locations of the surrounding streams (Figure 6.1). The study area is mostly forested but there is also some peatland, agricultural fields and operational gravel pits. The natural topography of the study area varies from 125 m above sea level (asl) at the groundwater divide line to 109 m asl at the study area surface water outlet (Tremblay et al. 2013). The landfills, reaching 130 m in elevation, changed the local topography. The water table, which follows topography, is quite shallow, and generally found between 0.1 to 2 m below ground surface. Within the subwatershed, groundwater flows from higher elevations found under the landfill towards local streams in the southern and northern direction (Figure 6.1). In this relatively flat region, the surface soil is mostly made of poorly drained podzols of marine origin, sparse zones of organic material and well decomposed black peat (Laplante 1962). As a consequence of the flat topography and soil type, an important forestry and agricultural drainage network is in place and, in combination with natural streams, local hydrology strongly influences groundwater flow (Tremblay 2013).

The granular aquifer is composed of 10 to 20 m of highly heterogeneous sand interbedded with silt and clayey silt lenses. The aquifer overlies discontinuous layers of glaciomarine silt and till forming the relatively impermeable base of the aquifer. The till deposited by the last glaciation overlies the regional sedimentary rock aquifer composed of fractured shale and sandstone. The Quaternary fluvial and deltaic sediments making up the aquifer were deposited after the last glaciation (Bolduc 2003). The unconsolidated material originated from sea level changes during the regression of the Champlain Sea at the former Chaudière River outlet. At that time, the system was controlled by sea tides and current, followed by fluvial erosion and organic sedimentation. This littoral depositional environment is responsible for the high heterogeneity of the aquifer material (Parent and Occhietti 1999) as well as naturally occurring anoxic and oxidizing zones that influence the natural geochemical composition of groundwater (Tremblay et al. 2008). Added to this complex environment, is the presence of a leachate plume emitted by the decommissioned landfill which undergoes important geochemical transformations along its path (Tremblay et al. 2008), which is the object of the present study.

A regional hydrogeological characterization was carried out in the Chaudière River watershed, including the chemical analysis of 127 groundwater samples within the sedimentary rock aquifers (Benoit et al. 2012). The geochemical data supported the results obtained from a numerical flow model that was used to define regional groundwater flow (Brun Koné 2012, 2011). The water type found in the center of the Breakeyville formation underlying the study area is Na-HCO_3, contains no tritium, and has an average chloride concentration below 1 mg/L. The presence of this water type seems influenced by the effect of regional flow controlled by structures of orientation SW-NO within the formation and the presence of a thicker aquitard covering the bedrock in this area. Based on phase 1 of the characterization work (Tremblay et al., 2012) and on local hydrogeological investigations carried out by consultants on the landfill property (Gauthier et al., 1993b), the sedimentary rock aquifer is found at a an verage elevation of 100 m asl within the study area, but is also known to outcrop or to reach up to 110 m asl at many locations. Surface geophysical surveys demonstrated thinning of the impermeable unit over certain locations where the rock aquifer showed higher elevations (Tremblay et al. 2013). This has implications on the hydraulic connectivity between the granular aquifer and the rock aquifer (Tremblay et al, 2012b), which influences groundwater geochemistry in the study area as will be discussed later.

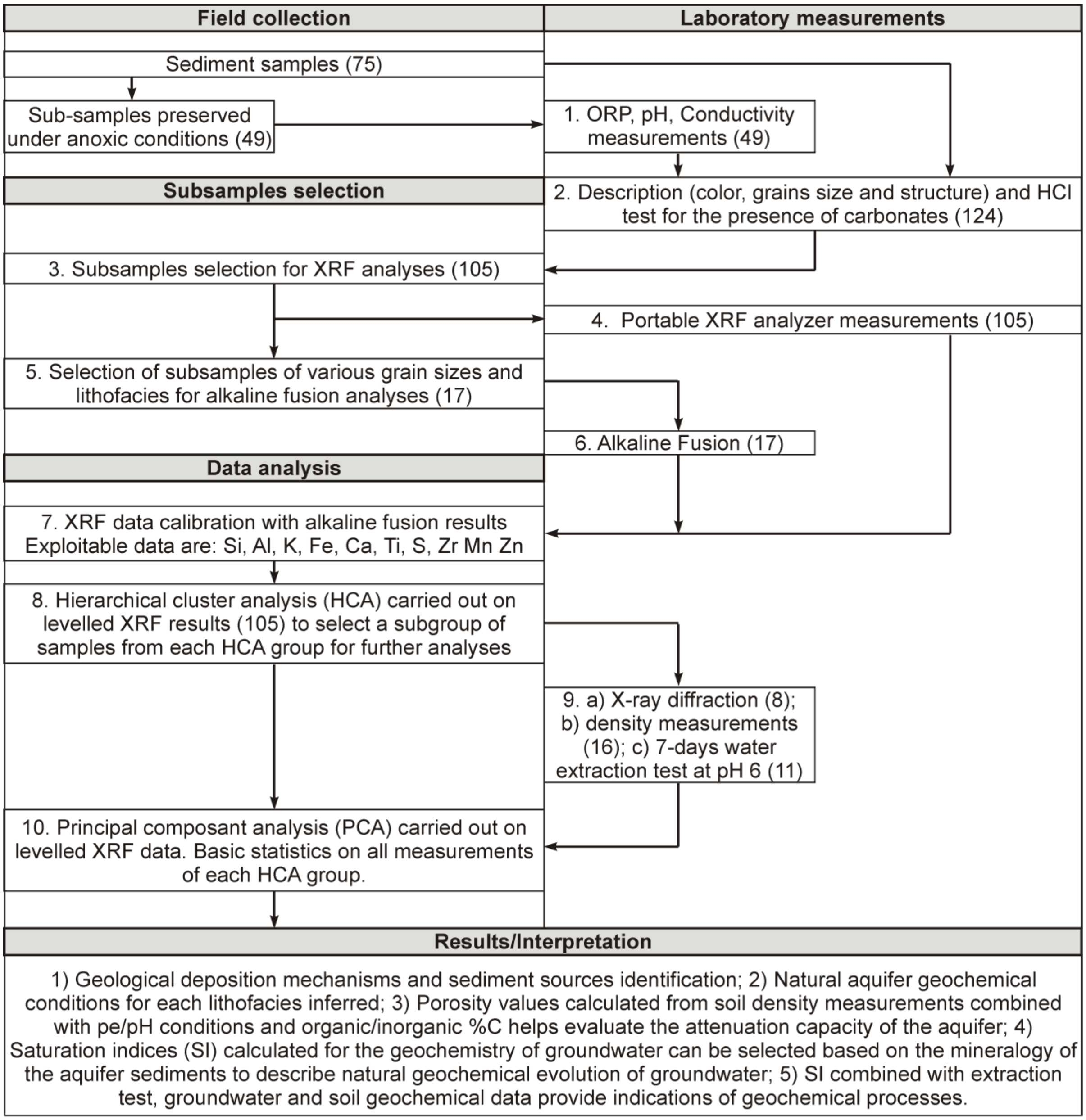
6.4 Methods and data processing

6.4.1 Sediment characterization and data processing

Seventy-five (75) soil cores of 2.5 cm diameter and 1.5 m length were collected with a direct-push system (Tremblay et al. 2012; Fauveau et al. 2005) and forty-nine (49) 15 cm long subsamples were preserved in nitrogen and frozen directly in the field (USEPA 2006). A workflow was developed to measure a variety of physical and geochemical parameters on the soil samples for the purpose of relating soil properties to the geochemical conditions of groundwater found in the aquifer (Figure 6.2). The use of a fast characterization method, such as a portable XRF analyzer, allowed the efficient identification of features distinguishing soil samples and the grouping of samples with similar characteristics. These results allowed the selection of fewer samples from similar groups for DRX analyses and leaching extraction tests.

Physical and geochemical properties of the aquifer sediments were characterized using various methods: X-Ray diffraction (XRD), X-Ray fluorescence (XRF), alkaline fusion, redox potential (ORP), pH, conductivity and soil density measurements to infer porosity. Permeability tests on soil columns using a lab permeameter and grain size analyses were also performed (Paradis et al. 2011). Moreover, 7-day soil leaching tests with neutral demineralized water were done (Centre d'expertise en analyse environnementale du Québec 2010). ORP, pH and conductivity measurements were carried out in an anoxic laboratory glove box on the 49 subsamples preserved in an anoxic environment. Sediments were extracted at a 1:1 ratio with nitrogen saturated water in soil paste (Rhodes 1982). Measurements were made with an YSI 556-MPS multiparameter probe (platinum ORP/pH probe; YSI 2012) while mixing the soluble salts.

Second, a visual description of structure, texture and color was done on the 75 samples and 49 subsamples. Sediments were air dried and a small amount of each sample was tested with HCl for the presence of carbonates. A subgroup of 17 samples spanning the range of lithologies and



1) Geological deposition mechanisms and sediment sources identification; 2) Natural aquifer geochemical conditions for each lithofacies inferred; 3) Porosity values calculated from soil density measurements combined with pe/pH conditions and organic/inorganic %C helps evaluate the attenuation capacity of the aquifer; 4) Saturation indices (SI) calculated for the geochemistry of groundwater can be selected based on the mineralogy of the aquifer sediments to describe natural geochemical evolution of groundwater; 5) SI combined with extraction test, groundwater and soil geochemical data provide indications of geochemical processes.

Figure 6.2 : Workflow for sediment analyses and data processing

The workflow is designed to provide a complete set of parameters from geochemical and physical analyses. Results from the multivariate data analysis provide indications on key processes: geological depositional environment; natural conditions (physical and geochemical) of groundwater; attenuation capacity of the aquifer based on groundwater and sediment geochemistry; mineralogy of aquifer sediments indicating relevant saturation indices to consider and thus constraining potential geochemical processes grain sizes found in the study area was selected for alkaline lithium metaborate fusion (laboratory of INRS, Centre Eau Terre Environnement, INRS-ETE). Sediments were oven dried at 60 °C and mechanically ground prior to analyses.

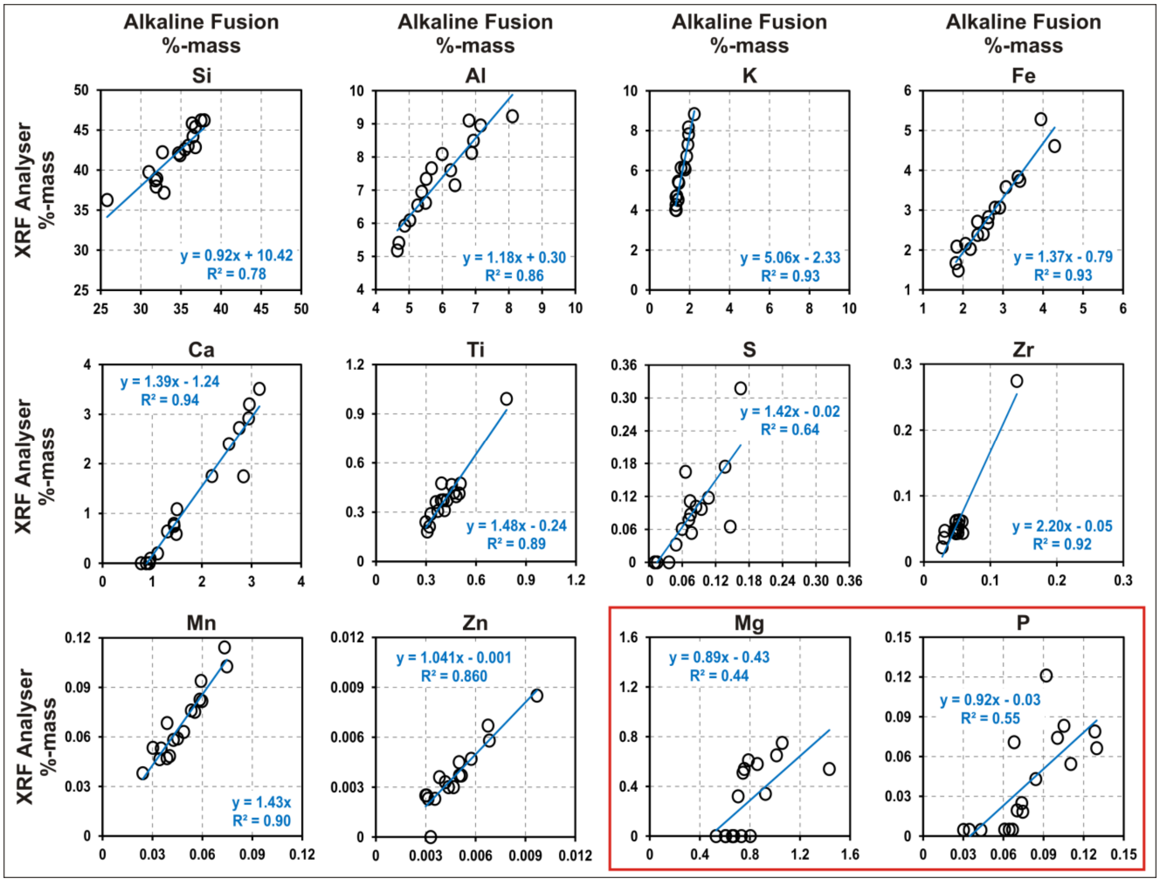


Figure 6.3 : Levelling sediments geochemical data sets

The adjustment of portable XRF analyzer data to alkaline fusion data is determined through regression. Alkaline fusion is the most reliable method of the two and a shift and a multiplier is used to obtain the best fit of XRF data. Levelling was impossible for some elements analyzed (Mg and P within red box). XRF results were corrected using the equations indicated on the graphs and only the elements shown were used for multivariate analysis

Third, measurements were taken on 105 samples with a portable Olympus Innov-X analyser (pXRF) DP-6000-C (Olympus 2013). The analyzer has to be in direct contact with the soil sample to avoid the dispersion of secondary X-rays and a thickness of at least 2 cm of sediment is required. Tests performed on the samples demonstrated that 45 seconds per measurement provided the best results for each element recorded. The XRF data had to be leveled as shown on

Figure 6.3 (XRF and chemistry) (Grunsky 2010). Afterward, the data were normalized, and centered using a logratio transformation prior to multivariate data analysis as required for compositional data analysis to avoid incorrect assessment of correlation (Pawlowsky-Glahn and Egozcue 2006; Egozcue et al. 2003; Aitchison 1986). Hierarchical clustering (HC) and principal component analysis (PCA) were carried out with Statistica 10 (StatSoft, 2010). Based on the clustering groups, a few samples representative of each HC group were selected for XRD and porosity measurements. XRD measurements were done at Laval University with a Siemens D5000. Semi-quantitative mineralogy was interpreted by Natural Resources Canada using EVA (Bruker AXS Inc.) with comparison to reference mineral patterns using Powder Diffraction Files (PDF) of the International Centre for Diffraction Data (ICDD) and other available databases. Semi-quantitative analysis was carried out using TOPAS (Bruker AXS Inc.), a PC-based program that performs Rietveld refinement (RR) of XRD spectra. This is based on a whole pattern-fitting algorithm. It relies on having a particular mineralogical structure such that the reference minerals are as closely matched as possible to the spectrum. Occasionally smear mounts are made in order to better identify clay minerals that are found in minor to trace amounts. Porosity calculations were also done based on dry weights of known volumes of sediments compacted in soil core sleeves and solids density measurements made with a Gay-Lussac 50 mL bottle (ASTM 2013; Saskatchewan Highway and Transportation 1993).

6.4.2 Groundwater sampling and data analysis

One hundred and twenty (120) groundwater samples were collected throughout the study area between 2010 and 2011 using three methods: 1) between packers in 29 wells screened over the full saturated thickness of the aquifer; 2) in 10 discrete shallow piezometers (1.5-2 m below ground surface); and 3) in 30 discrete observation wells screened at specific short intervals (typically between 0.2 and 1.5 m long). Wells were purged over three times their total volume before introducing the packers and the intervals isolated between packers were further purged over three times the total volume of the water in place. Low-flow groundwater sampling was done with a peristaltic pump to preserve sensitive geochemical conditions. Temperature, electrical conductivity, dissolved oxygen (DO), redox potential (ORP) and pH were recorded on site with a multi-parameter probe (YSI 556-MPS (YSI 2013)). When physicochemical parameters had stabilized (10 min to 30 min), groundwater was collected through 45 µm high capacity HDEP filters fitted directly at the outlet of the MFLEX FDA viton #16 7.6 m (25 ft) tubes. Groundwater was analyzed for major and minor elements as well as for stable (^{18}O, ^{2}H, ^{15}N, ^{3}He/^{4}He, ^{20}N, ^{21}N, ^{22}N) and radiogenic (^{14}C, ^{3}H) isotopes. Alkalinity was determined on site by titration with the total alkalinity method (AWWA 1995a). Fe^{2+} and S^{2-} were dosed directly in the field but readings were taken with a Thermo Spectronic Genesys 10 µv spectrophotometer in 1 cc cells in the lab at the end of each day (Voillier et al. 2000; AWWA 1995b). All samples were stored on ice after collection and during transportation to the laboratory. Teflon pipe 1/4-1/16 mur and teflon union straight connectors were cleaned between samples with oxalic acid 0.4 M and HCl 1%. Pipes were rinsed abundantly with demineralized water between uses of each cleaning product. Analyses were made on the demineralized rinsing water to confirm the absence of trace elements in the pipes from cleaning operations.

Major and minor elements were analyzed at an accredited commercial laboratory (MAXXAM Analytic). ^{14}C and ^{13}C analyses were prepared through the Environmental Isotope Laboratory at the University of Waterloo on 1 L groundwater samples and analyses were carried out at the National Isotope Center of the Institute of Geological and Nuclear Sciences Ltd in New-Zealand.

Groundwater geochemical data were interpreted using multivariate statistical methods as described by Cloutier et al. (2008) using Statistica version 10 (StatSoft Inc. 2010). Multivariate analysis was preferred over other methods since Hierarchical cluster analysis (HCA) was used to identify distinct groups and to distinguish leachate-affected water from natural groundwater and to classify the data in a relatively simple and direct manner. First, the geochemical data set was validated with the approach proposed by Hounslow (1995) and chemical balance and saturation indices were calculated using PHREEQCI 2.18.5570 (Parkhurst and Apello 1999). Based on the results obtained, a final data selection was made. For a same well, samples collected in August were favored, duplicates for all samples were removed, and samples showing a chemical balance error over >15% were eliminated. Eighty-four (84) samples thus remained for further analysis. Physicochemical parameters were included for the statistical analysis. Geochemical parameters used for statistical analysis only included the elements for which less than 10% of the groundwater samples were below the analytical detection limit (DL) (Farnham et al. 2002). Non-detected concentrations for selected elements were substituted by half the value assigned for their detection limit (Farnham 2002). Seventeen (17) variables (Ca, Mg, Na, K, Fe, Mn, NH_4,

SO_4, Cl, Sr, Ba, Al, Si, alkalinity, pH, pe and conductivity) were transformed for normality with the Box-Cox operation and standardized with Statistica 10 (StatSoft 2010).

6.5 Results

6.5.1 Sediment analysis

6.5.1.1 Multivariate analysis of the geochemical composition of aquifer sediments

Table 6.1 summarizes the sediment classification made on the basis of hierarchical cluster analysis (HCA) on sediment geochemical data (portable XRF analyzer on 105 samples). The classification of sediment groups seems highly related to particle size and somewhat to stratigraphy. Table 6.2 shows median values for XRF and alkaline fusion results for each HCA group. The data shows the same range of values for both methods. Table 6.2 also shows porosity estimated from soil density measurements, pe and pH for each group. Sediments with coarser texture have acidic pH and higher pe, whereas fine texture sediments have high porosity, alkaline pH and low pe.

Tableau 6.1 : Description of sediment HCA

Description of sediment classification results based on hierarchical cluster analysis (HCA) of geochemical data recorded with a portable XRF analyzer for 105 sediment samples. (*P03 is a borehole located on the east side of the groundwater divide line where ground elevation is higher and sediments coarser. Lithofacies found closer to the ground surface within the study area are found deeper below the ground surface in the vicinity of borehole P03)

HCA Groups			**Grain size**	**Color**	**Depth range (m)**
🟨	1	Medium sand	Medium to fine	Light brown	0-5 (*P03 6-8)
🟡	2	Medium to very fine sand	Medium to very fine	Light brown to grey	0-5 (*P03 9-10)
🔸	3	Fine sand	Fine to very fine	Light brown to grey	2-5 (*P03 13-14)
🔹	4	Sandy silt	Very fine	Grey	2-10 (*P03 15-16)
🔵	5	Silty sand	Very fine	Grey to pink	2-15
🟦	6	Silt	Coarse to fine	Grey to pink	3-15
🟩	7	Till matrix (sand and silt)	Medium to fine	Pink	8.5-13.5

Tableau 6.2 : Median values from XRF and alkaline fusion

The table compares the median for the two types of chemical analyses for each HCA group. The letter n stand for number of samples per analysis. The symbol “-“ stands for non-detected and DL for detection limits. Also, porosity inferred from soil density measurements and pe/pH are indicated for each group

			HCA groups													
			1		2		3		4		5		6		7	
		n	*2*	*16*	*1*	*9*	*2*	*18*	*4*	*23*	*1*	*15*	*2*	*15*	*5*	*9*
Elements	Units	DL	AF	XRF	AF	XRF	AF	XRF	AF	XRF	AF	XRF	AF	XRF	AF	XRF
Si	%-mass	0.0075	38	35	37	35	36	36	36	33	33	34	32	31	32	30
Al	%-mass	0.0022	4.76	4.89	4.69	4.86	5.44	5.13	5.39	5.89	6.38	5.74	6.93	6.83	6.84	6.14
Fe	%-mass	0.0015	1.85	2.02	1.84	2.10	2.43	2.34	2.28	2.71	3.95	2.63	3.37	3.19	3.00	2.74
Ca	%-mass	0.0039	0.86	0.92	0.88	0.93	1.04	1.12	1.44	1.46	2.55	1.58	2.75		3.06	
Na	%-mass	0.032	1.65		1.72		1.81		1.89		2.34		2.02		2.25	
K	%-mass	0.007	1.32	1.23	1.31	1.34	1.42	1.36	1.40	1.54	1.78	1.51	1.91	1.78	1.90	1.66
Mg	%-mass	0.0013	0.57		0.66		0.66		0.77		0.66		1.01		0.75	
Ti	%-mass	0.0006	0.31	0.31	0.30	0.35	0.40	0.38	0.36	0.41	0.78	0.42	0.47	0.44	0.48	0.44
C (inorg)	%-mass	0.02	0.08		-		0.03		0.08		0.18		0.37		0.37	
C (org)	%-mass	0.02	0.08		0.13		0.09		0.21		0.09		0.21		0.19	
S	%-mass	0.01	0.03	0.02	0.05	0.04	0.01	0.02	0.07	0.05	0.09	0.05	0.12	0.08	0.15	0.17
Zr	%-mass	0.0001	0.04	0.03	0.03	0.04	0.05	0.05	0.05	0.05	0.14	0.05	0.05	0.04	0.05	0.05
P	%-mass	0.01	0.04		0.03		0.07		0.07		0.09		0.11		0.10	
Ba	%-mass	0.0002	0.05		0.05		0.05		0.05		0.07		0.07		0.08	
Mn	%-mass	0.0002	0.03	0.03	0.03	0.03	0.04	0.04	0.04	0.05	0.07	0.05	0.06	0.05	0.05	0.05
Sr	%-mass	0.0002	0.02		0.02		0.02		0.02		0.03		0.03		0.04	
N	%-mass	0.01	0.02		0.01		0.02		0.03		-		0.03		0.02	
Zn	mg/Kg	2	32	35	30	36	42	42	39	54	50	48	67	62	362	53
Cr	mg/Kg	3	83		100		74		77		38		62		84	
Ni	mg/Kg	12	32		53		31		32		18		29		96	
V	mg/Kg	5	37		37		47		47		69		65		56	
As	mg/Kg	30	-		-		-		-		-		45		-	
La	mg/Kg	2	19		17		28		25		27		35		28	
Cu	mg/Kg	2	9		25		13		16		7		16		27	
Co	mg/Kg	4	4		8		5		7		6		7		10	
Sc	mg/Kg	4	-		-		5		-		8		8		7	
Cd	mg/Kg	1.5	1.9		-		-		-		4.0		3.6		3.5	
Porosity			0.41		0.39		0.40		0.40		0.40		0.43		0.36	
pH			4.69		6.46		5.43		7.14		8.50		7.56		8.70	
pe			5.3		-0.2		3.6		-0.4		0.5		-0.6		-1.5	

For the two principal components (PC’s) explaining a larger proportion of variance, Figure 6.4 shows results of soil samples distinguished by their HCA group. The loadings for these PC’s of each chemical element used for the analysis are also shown on the graph (the values of loadings are indicated by the locations of chemical symbols). Principal component 1 is related mostly to the relative proportions of silicate (dominating in coarse sand) versus carbonate (dominating in

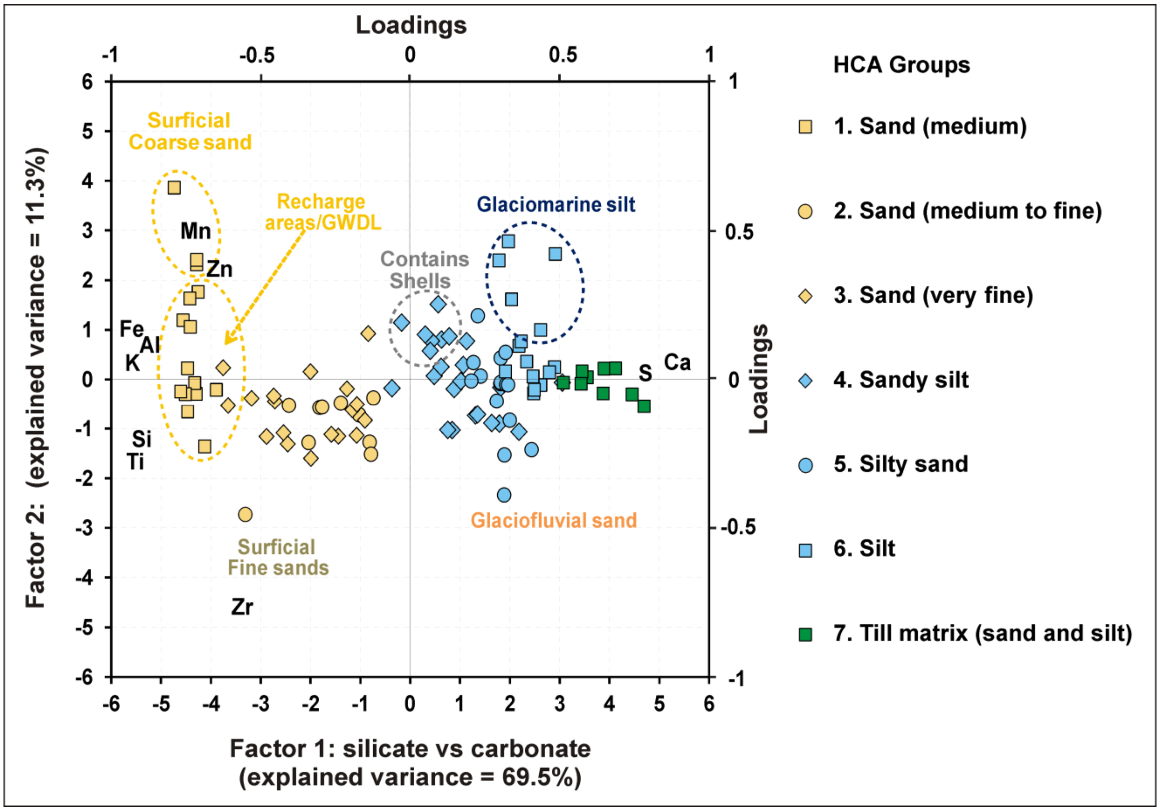


Figure 6.4 : Principal component analysis carried out on calibrated XRF data

Values of samples as well as relative loadings of chemical elements (black text) for the two main factors are shown on the graph. The classification is based on prior HCA analysis. PCA allows the recognition of many lithofacies and the chemical factors controlling the sediment groupings. GWDL stands for "groundwater divide line"

silt and till). Once again, the analysis demonstrates that chemical composition is highly related to particle size. Furthermore, these HCA groups also have specific spatial distributions in the study area. Coarser and homogeneous sediments containing no carbonates are localized along the eastern groundwater divide line where it is very likely that the sediments were deposited under a fluvial influence (Chaudière River). At the same location, coarse chunks of iron and manganese particles are clearly visible within the sediments at the soil surface. For example, P03 is a borehole located just east of the groundwater divide line where ground elevation is higher and sediments are coarser. At this location, the lithotypes found closer to the ground surface elsewhere in the study area are found deeper in the vicinity of borehole P03 (Table 6.1). Finally; the interpretation of PCA from the XRF geochemistry based on HCA groups indicates depositional mechanisms and sediment provenance, it was used to select samples for other analyses and, more importantly, it is correlated to the elements found in groundwater as will be discussed later.

6.5.1.2 Mineralogy of the aquifer matrix from XRD

Tableau 6.3 : Semi-quantitative X-ray diffraction analyses results

Semi-quantitative X-ray diffraction analyses results showing that the mineralogy is similar between samples. The symbol “-“ stands for non-detected. Results are consistent with the PCA showing that quartz diminishes in the finer sediments and layer silicates become predominant, carbonates also appear mostly at depth or in the silt interlayered with sand

Mineralogy (%)		**Sediment HCA Group**							
Mineral	Formula	**1**	**1**	**4**	**4**	**5**	**6**	**6**	**7**
Quartz	SiO_2	74	60	71	57	36	37	36	36
Plagioclase feldspar-albite	$NaAlSi_3O_8$	19	24	14	25	25	33	25	41
K-feldspar-microcline	$KAlSi_3O_8$	-	9	5	8	7	17	7	10
Amphibole-fero-tschermakite	$Ca_2Mg_3Al_3Fe^{3+}Si_6O_{22}(OH)_2$	-	tr	1	1	4	3	4	1
Mica-biotite	$K(Mg,Fe^{2+})_3(AlSi_3O_{10}(OH,F)_2)$	4	3	7	3	14	2	14	5
Chlorite-clinochloreII	$(Mg,Fe^{2+})_5Al(Si_3Al)O_{10}(OH)_8$	3	4	2	6	4	6	4	2
Calcite	$CaCO_3$	tr	tr	tr	-	7	tr	7	3
Dolomite	$CaMg(CO_3)_2$	-	tr	tr	-	3	2	3	2
nd = non-detected	Total	100	100	100	100	100	100	100	100

XRD analyses were done on eight (8) sediment samples selected from the HCA groups previously defined. Table 6.3 presents the 8 minerals identified in varying proportions by semi-quantitative analysis: quartz, plagioclase, k-feldspar, amphibole-fero-tschermakite, mica-biotite, chlorite-clinochloreII, calcite, and dolomite. XRD analysis detects mostly the minerals that are found above 5% of the total composition of a sample. Results reveal that the mineralogy is similar throughout the aquifer. However, the proportion calculated for each mineral type changes with grain size and location. Carbonate proportions increase with depth as sediments become finer, and in relation with the depositional environment. Once again, no carbonate minerals were detected in the recharge zone (Tremblay et al. 2013) where sand was deposited under the influence of a fluvial channel or in a beach environment sometimes reworked by wind. Furthermore, based on the underlying sedimentary rock mineralogical data (Brazeau 1999), sediment provenance is a combination between the St. Lawrence north shore granitic bedrock and the south shore sedimentary rocks. Deeper in the granular aquifer, the south shore rock signature is dominant, whereas the north shore granitic signature is predominant at local topographic highs.

From these results, conclusions can be made on the depositional environment causing the mineralogical variability within the aquifer and in relation to the local geographic location of sediments. The depositional environment of the unconsolidated material originated from sea level changes during the regression of the Champlain Sea at the former Chaudière River outlet. At that time, the system was controlled by sea tides and current, followed by fluvial erosion and organic sedimentation. This littoral depositional environment is responsible for the high heterogeneity of the aquifer material (Parent and Occhietti 1999). Other minerals were easily visible and identifiable in the sediments, such as magnetite, iron oxides and manganese oxides.

6.5.1.3 pH and ORP as indicators of the aquifer natural attenuation capacity

Contaminant migration and transformation rates are strongly dependent on redox conditions of an aquifer system (Medina 2001). Redox conditions of sediments are a good indicator of the oxidation capacity potential of an aquifer. The oxidation potential of an aquifer is a relative measurement that reveals its capacity to receive or transfer electrons. As the leachate migrate through an aquifer, the electron acceptors are used up, the aquifer becomes more reductive and the redox potential of groundwater decreases. Iron and manganese geochemistry of aquifer sediments is key to understand the occurrence of redox zones and attenuation processes as leachate progresses within an aquifer (Medina 2001).

pH and ORP measurements were carried out on 49 sediment subsamples preserved under anoxic conditions (Figure 6.5) (section 6.4.1). Figure 6.5 shows sediment pe and pH values in relation to theoretical stability fields for iron and manganese mineralogical forms (adapted from Stumm and Morgan 1981). These results thus provide indications of the likely Fe and Mn mineral forms in the aquifer, which will influence its attenuation capacity. The aquifer materials are mostly naturally reductive, which will affect groundwater signature and attenuation processes. Besides a few sediment samples located upstream from the landfill in a preferential recharge zone (medium sand and coarser surficial sand material where oxygen is present), most sediments are found between pH 7 and 8 and pe from 1 to -1. Therefore, pH and pe conditions of sediments indicate that the aquifer offers optimal biodegradation conditions (under ferrous iron compositions) and a natural anaerobic bacteriological environment for leachate attenuation (Sims et al. 1993; U.S. USEPA 1993; Cookson 1995). Moreover, concentrations of iron, manganese, carbon, sulfur and nitrogen in sediments (Table 6.2), their high porosity and the interstratification of the material in place (Tremblay et al. 2013) are other indicators that the aquifer offers the geochemical composition and physicochemical conditions to provide a high capacity for natural attenuation. However, these analyses are only indications of the conditions found within the aquifer material and further sediment extractions and analyses would have to be carried out to quantify the aquifer oxidation capacity and the forms and concentration of iron and manganese present in the sediments (Medina 2001).

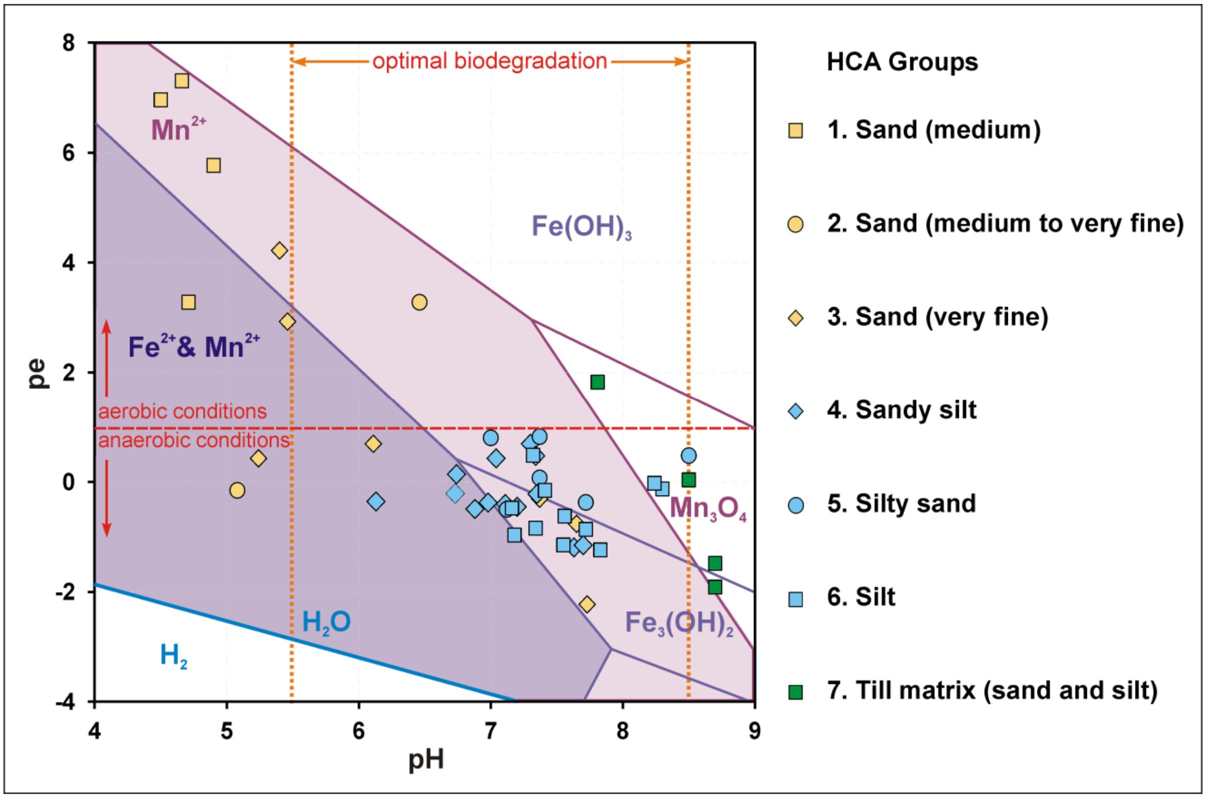


Figure 6.5 : pH and pe diagram for sediment samples

pH-pe diagram showing results of measurements on 49 sediment subsamples preserved under anoxic conditions. Stability fields for iron and manganese are represented (adapted from Stumm and Morgan 1981) to identify the Fe and Mn mineral forms likely present in the aquifer, which will influence its attenuation capacity. Away from sandy preferential recharge zones (yellow squares and circles), most aquifer sediments, both sands (yellow) and silty materials (blue) are naturally reductive, which affects the groundwater signature and attenuation processes

6.5.1.4 Sediment 7-day leaching tests

7 day-leaching tests were performed on samples from each HCA sediment group (section 6.4.1). These tests aimed to identify chemical components that would readily dissolve in water, plus determine distinct leaching behavior of sediment groups. These results will be compared later in the paper with groundwater analyses to help define the natural conditions of shallow groundwater and the effects of their potential interactions with evolved groundwater originating from the sedimentary rock aquifer or leachate from the landfill.

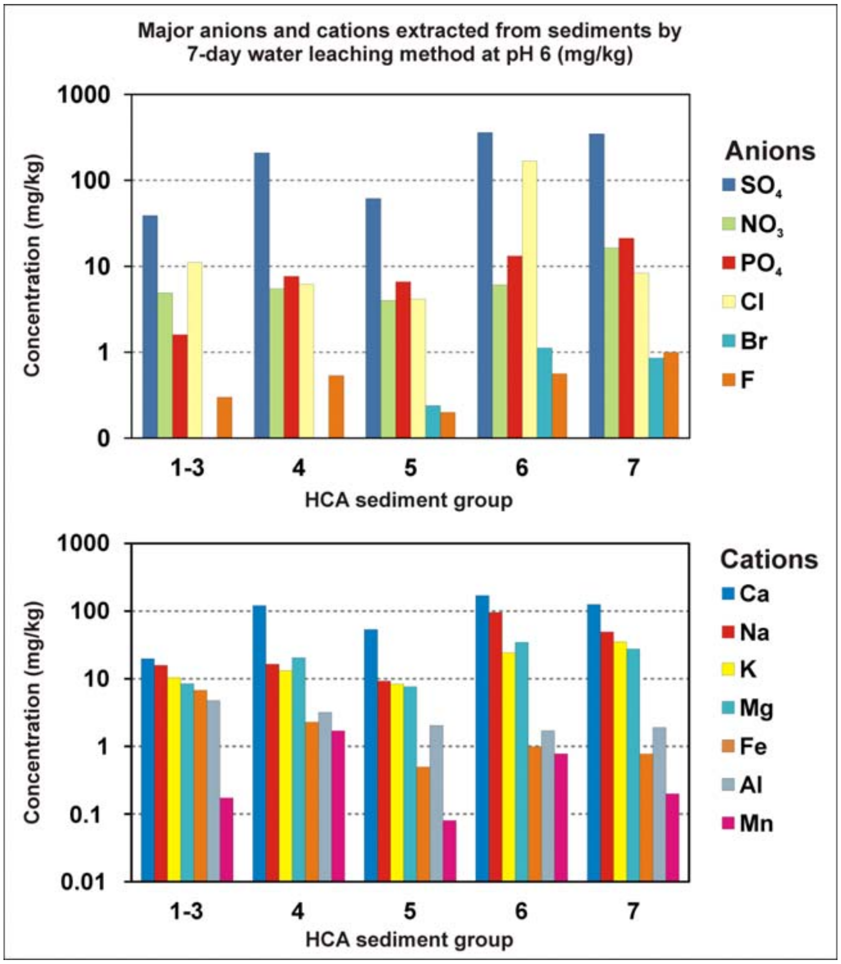


Figure 6.6 : Major anions and cations extracted from sediment leaching tests

Concentrations for iron, manganese and other elements might not be representative of aquifer conditions since the tests were not performed under anaerobic conditions. These results indicate components readily dissolved in the natural aquifer environment. One sediment sample from a well in group 6 is located within the leachate plume and thus shows anomalously elevated values for Cl, Br, Mg and Na

Tableau 6.4 : 7-day water extraction leaching test results

The letter n stand for number of sample per analysis. The symbol “-“ stands for non-detected and DL for detection limits. Average values obtained illustrating the anions and cations that will dissolve readily under the soil conditions found in the study area. An elevated concentration of Cl is recorded in group 6, however only one sample had a value higher than 24 and the sample showing an elevated value is located within the waste site property limits and is affected by leachate

			Sediments HCA Group				
			1-3	**4**	**5**	**6**	**7**
Elements	Units	DL	*n=3*	*n=3*	*n=1*	*n=2*	*n=2*
Ca	mg/kg	0.015	20	121	53	169	126
Na	mg/kg	0.004	16	16	9	96	49
S	mg/kg	0.03	15	76	25	133	127
K	mg/kg	0.009	10	13	8	24	35
Mg	mg/kg	0.004	8	20	8	35	27
Fe	mg/kg	0.0021	6.7	2.3	0.5	1.0	0.8
Al	mg/kg	0.006	4.8	3.2	2.0	1.7	1.9
Mn	mg/kg	0.0008	0.17	1.70	0.08	0.78	0.20
SO_4	mg/kg	1	39	210	62	360	347
NO_3	mg/kg	1	5	5	4	6	16
Cl	mg/kg	0.1	11	6	4	167	8
Br	mg/kg	0.1	-	-	0.24	1.12	0.85
PO_4	mg/kg	1	1.60	7.64	6.59	13.18	21.17
F	mg/kg	0.1	0.30	0.53	0.20	0.56	1.00
As	mg/kg	0.01	-	0.04	0.04	0.021	0.04
Ba	mg/kg	0.0002	0.10	0.34	0.07	0.37	1.67
Cd	mg/kg	0.0004	0.0009	0.0010	-	-	-
Co	mg/kg	0.0021	0.008	0.005	-	-	-
Cr	mg/kg	0.001	0.013	0.012	-	0.005	0.007
Cu	mg/kg	0.0018	0.025	0.028	0.015	0.046	0.023
Mo	mg/kg	0.0021	0.025	0.026	0.010	0.037	0.266
Ni	mg/kg	0.002	0.049	0.021	0.005	0.014	0.008
P	mg/kg	0.013	0.205	0.183	0.270	0.206	0.265
Pb	mg/kg	0.005	0.021	0.026	0.011	0.015	0.019
Sr	mg/kg	0.0004	0.113	0.779	0.317	1.125	1.307
Ti	mg/kg	0.0005	0.184	0.202	0.043	0.061	0.076
Zn	mg/kg	0.0009	0.066	0.023	0.012	0.033	0.109
pH		0.1	7.87	7.93	7.87	7.87	7.52
Total alc ($CaHCO_3$)	mg/L	1	26	41	27	42	43

Leaching test results are presented in Table 6.4 and illustrated in Figure 6.6. Once again the results are consistent with the mineralogical analyses and sediment grain sizes. As grain sizes are smaller, the concentrations of dissolved components increase in the collected water. Calcium is the dominant cation for all samples and sulfate is the dominant anion. Iron and aluminum concentrations decrease as sediments become finer, whereas magnesium, potassium, fluoride and phosphate increase. Bromide concentrations are detected in finer sediments (groups 5-7). Also chloride concentration is high for group 6; however, one sample from this group is known to be within the extent of the leachate plume. Leaching tests were done under normal atmospheric conditions, while sediments in the aquifer are under anaerobic conditions and different pressures (Figure 6.5). Thus, while these results may not be exactly representative of what could leach from sediments under aquifer conditions, they indicate which components are readily soluble for each sediment type. An example of the difference between the leaching test results and the aquifer is that nitrate is not detected in groundwater but ammonium is, contrary to leaching tests. Multiplying the average porosity of 0.4 measured for most sediments with the concentration obtained in mg/kg can provide an approximate concentration that could be found in groundwater. Therefore, concentrations would be very low with the exception of sediment groups 4, 6 and 7. These data will be later compared with the geochemical analysis on groundwater and put in context with the mineralogy of each sediment group. Moreover, since the tests were performed under pH 6 (and that rain and recharge water are more acidic in reality), it was taken into consideration that not all constituents from the sediments were able to dissolve representatively.

6.5.2 Groundwater geochemistry

6.5.2.1 Multivariate and graphical analysis

Multivariate methods are useful for interpreting simultaneous changes in several properties (Davis 2002). HCA and PCA were done on 84 groundwater samples, including two samples from the sedimentary rock aquifer. HCA was done using Ward's method as a linkage rule and Euclidean distance measurements (Cloutier et al. 2008). Figure 6.7 shows the HCA results that distinguish five main groundwater groups below a Phenon line set at a linkage distance of 16. Under the graph, Figure 6.7 also indicates the names of water groups, the symbols used on following graphs to identify the groups (by color) and subgroups (by symbols), the major ions representing the water types related to the groups, and Stiff diagrams illustrating the concentrations of major ions in the subgroups. The geochemical conditions and spatial distributions of the HCA groups were used to recognize natural groundwater from groundwater related to landfill leachate or impacted by leachate. Natural groundwater groups were named according to their distinct pH ranges (Alkaline, Neutral, and Acidic). Table 6.5 provides statistics on groundwater geochemical parameters for each HCA water group. Table 6.5 and the Stiff diagrams on Figure 6.7 clearly show that HCA water groups are geochemically quite distinct in terms of concentrations of both major and minor ions as well as physico-chemical conditions.

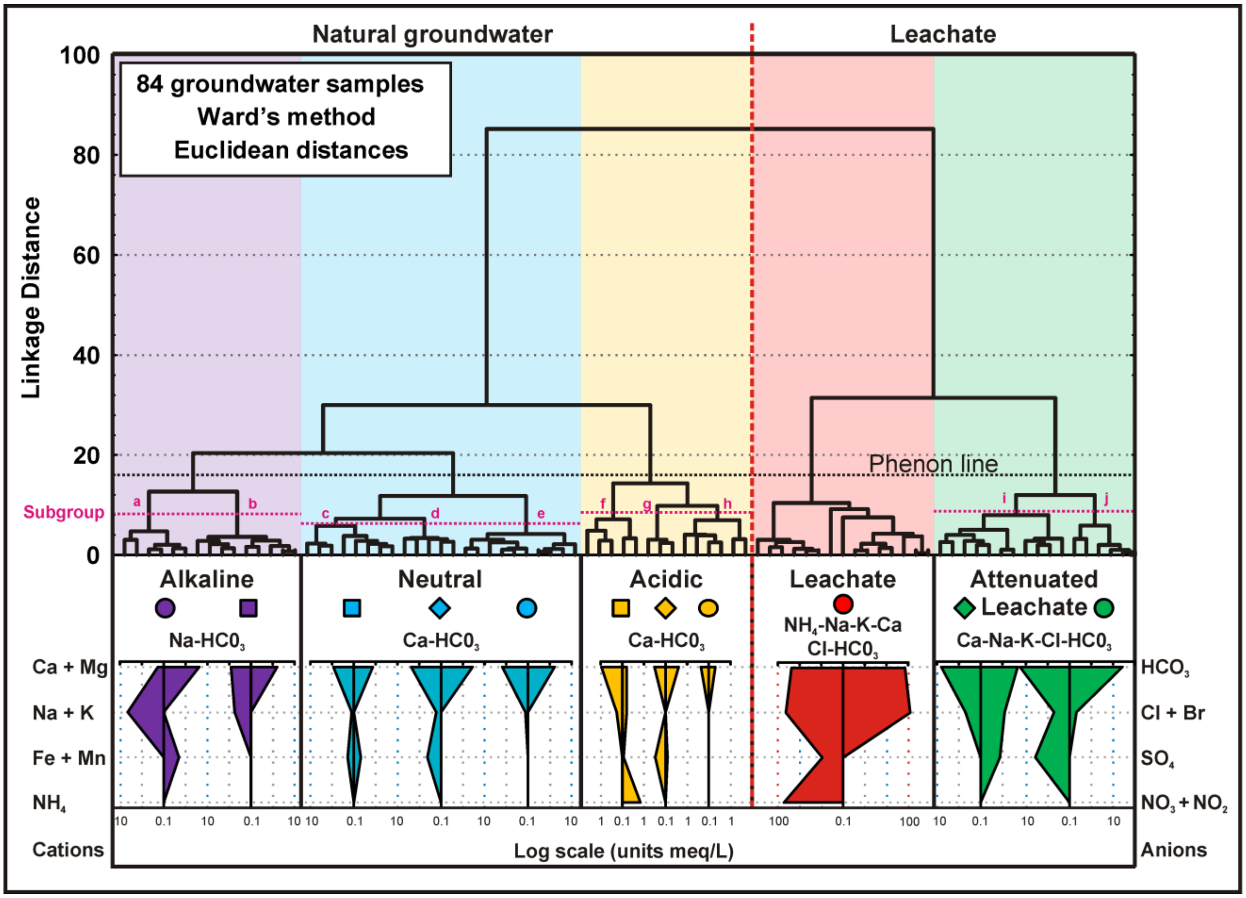


Figure 6.7 : Hierarchical cluster analysis (HCA) for groundwater

The phenon line set at 16 classified the groundwater into 5 main groups. Subgroups were selected based on HCA and by specific characteristics defining these groups that are illustrated by stiff diagrams on the lower part of the figure. Geochemical compositions of HCA Groups are summarized in Table 6.5, whereas Table 6.6 describes the subgroups and the geochemical processes affecting their chemical composition

Tableau 6.5 : Groundwater geochemistry for each HCA group

		Acidic			Neutral			Alkaline			Leachate			Attenuated lechate		
	n	*14*			*23*			*15*			*15*			*17*		
	DL	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max
Ca	0.02	1.2	4.4	18	6.1	16	38	2.1	12	27	8.7	110	1900	11	46	290
Mg	0.01	0.32	1.7	9.3	2	3.9	10	0.35	2.7	6.4	16	74	300	4.5	14	65
Na	0.03	0.62	1.7	7.4	0.64	2.3	9.7	4.9	13	100	14	400	1400	2.1	9.1	140
K	0.1	-	-	4.3	0.35	0.66	8.9	1.7	3.7	7.4	22	340	1600	0.7	3.3	89
Fe	0.03	-	-	9.6	-	-	43	-	-	0.6	5.1	29	1400	0	8.2	120
Mn	0.0004	0.001	0.06	9.4	0.07	0.3	5.5	0.01	0.04	0.17	0.03	0.21	57	0.14	1.3	10
N-NH4	0.02	-	-	0.75	-	-	1.2	0.37	0.61	1.5	31	360	1500	0	1.1	92
HCO3	1	1.2	10	52	37	74	168	89	132	252	432	2712	12161	83	312	1620
Cl^-	0.05	0.25	1.1	16	0.29	0.5	8.8	0.31	0.58	11	7.2	470	1800	0.35	12	160
Br^-	0.1	-	-	-	-	-	-	-	-	-	-	-	2	-	-	0.7
SO_4^{2+}	0.1	-	-	11	-	-	28	-	-	33	-	-	1500	-	-	88
N-NO^{3-}	0.01	-	-	20	-	-	0.02	-	-	0.08	-	-	3.5	-	-	0.13
N-NO^{2-}	0.01	-	-	0	-	-	-	-	-	-	-	-	0.84	-	-	-
Al	0.001	-	-	1.2	-	-	0.14	-	-	0.08	0.01	0.03	0.31	-	-	0.14
As	0.001	-	-	0.017	-	-	0.03	-	-	0.065	0.003	0.028	0.12	-	-	0.053
Ba	0.002	0.005	0.03	0.39	0.01	0.02	0.27	0.01	0.03	0.16	0.14	0.55	4.6	0.03	0.13	1.5
B	0.005	-	-	0.026	-	-	0.028	0.02	0.1	0.24	0.18	1	4.7	0.01	0.019	0.29
Cr	0.0005	-	-	0.0092	-	-	0.001	-	-	-	0.003	0.008	0.11	-	-	0.003
Co	0.0005	-	-	0.056	-	-	0.058	-	-	-	0.001	0.004	0.45	-	-	0.039
Cu	0.0005	-	-	0.028	-	-	0.004	-	-	0.004	-	-	0.002	-	-	0.003
F	0.1	-	-	0.1	-	-	0.1	-	-	1.4	-	-	0.4	-	-	0.1
Li	0.001	-	-	-	-	-	-	-	-	-	-	-	0.024	-	-	0.017
Mo	0.0005	-	-	0.001	-	-	0.004	-	-	0.0031	-	-	0.012	-	-	0.001
Ni	0.001	-	-	0.14	-	-	0.031	-	-	0.0029	0.005	0.073	0.5	-	-	0.034
Pb	0.0001	-	-	0.002	-	-	0.002	-	-	0.0001	-	-	0.0007	-	-	0.001
Si	0.02	1.7	5.4	11	3.6	7.5	15	4.3	6.3	11	0.1	9.7	25	3	9.2	26
Sr	0.002	0.02	0.04	0.17	0.05	0.11	0.24	0.03	0.13	0.35	0.33	0.99	9.8	0.10	0.58	3.1
S^{2-}	0.02	-	-	0.7	-	-	0.2	-	-	0.5	-	-	0.1	-	-	2.1
Ti	0.001	-	-	0.015	-	-	-	-	-	-	-	-	0.41	-	-	0.003
V	0.002	-	-	0.025	-	-	0.002	-	-	0.002	-	-	0.100	-	-	0.005
Zn	0.001	-	-	0.04	-	-	0.09	-	-	0.01	-	-	0.09	-	-	0.32
P	0.01	-	-	0.05	-	-	0.10	0.03	0.23	0.47	-	-	2.10	-	-	0.04
COD	10	-	-	77	-	-	140	-	-	10	81	460	40000	-	-	680
BOD$_5$	4	-	-	10	-	-	10	-	-	5	4	37	28000	-	-	420
C-DIC	0.1	1	7	22	7	14	30	17	33	47	115	519	2395	24	64	263
C-DOC	0.1	0.3	0.7	2.0	0.4	1.6	36	0.1	1.3	5.4	20	239	12240	2	12	241
CS	mS/cm^c	0.02	0.05	0.57	0.05	0.12	0.93	0.11	0.20	0.48	1.93	5.27	19.63	0.21	0.66	2.23
DO	-	0.55	9	11	0.16	0.56	9	0.11	0.36	8	0.15	3	71	0.10	0.33	14
pH	-	4.31	4.61	5.63	5.37	6.76	7.92	7.05	8.05	8.93	5.41	6.62	7.37	6.04	6.61	7.86
pe	-	0.4	4.3	10.0	0.4	2.8	5.9	1.2	2.5	4.5	1.3	2.5	4.7	-0.7	2.3	5.1
CH$_4$	0.5	-	-	-	-	-	7	-	-	-	0.63	4	39	-	-	-
^{2}H	-	-107.6	-85.5	-77.9	-88.9	-82.1	-77.3	-87.7	-83.6	-77.5	-82.4	-72.5	-49.3	-92.7	-87.5	-69.2
^{18}O	-	-16.1	-12.5	-11.9	-13.2	-12.4	-11.7	-13.3	-12.1	-11.5	-12.6	-11.2	-10.9	-17.4	-12.9	-11.7
^{15}N	-	-	-	-	-	-	-	-2.75	0.25	3.25	2.81	4.80	7.88	4.81	5.38	9.95
^{3}H	0.8	9	11	15	8	11	13	-	-	-	134	155	600	11	12	13

n = number of sample; DL = detection limit; "-" = non-detected

Tableau 6.6 : HCA group and subgroup descriptions and geochemical processes at their origin

GW HCA Group and Subgroup			**Distinctive Features**	**Geochemical processes**
Alkaline	🟣	a	Na-HCO_3-SO_4. Two samples are from the fractured bedrock and four samples are from the granular aquifer. GW contains diagnostic minor elements F, P,B.	Carbonate weathering (limestone and/or dolomite), cations exchange, sodium from albite and/or ions exchange, **plagioclase weathering not likely**
	🟪	b	Ca-Na-HCO_3 Groundwater from the bedrock aquifer emerging into the granular aquifer carrying its signature and mixing with evolved fresh water GW contains diagnostic minor elements F, P,B.	Carbonate weathering (limestone and/or dolomite), cations exchange, sodium from albite and/or ions exchange**, plagioclase weathering possible**
Neutral	🟦	c	Ca-HCO_3-SO_4 (presence of Fe variable)	Silicate and carbonate weathering, ferromagnesian minerals dissolution, sodium from albite and/or ion exchange
	🔷	d	Ca-HCO_3 (Fe) anoxic superficial confining silt unit (most samples are from 1 to 5 m depth)	Carbonate weathering (limestone-dolomite) and dissolution of ferromagnesian minerals, **plagioclase weathering possible for superficial GW** but not for sample below 5 m depth.
	🔵	e	Ca-HCO_3 samples found >7m depth	Silicate and carbonate weathering, sodium from albite/ions exchange, dissolution of ferromagnesian minerals
Acidic	🟨	f	Wells located at the GW divide line. Ca-HCO_3 type but contains anthropogenic NO_3 and Cl from agricultural activities	Silicate weathering and cation exchange
	🔶	g	Superficial wells underneath anoxic soil unit contain S and Fe	Silicate weathering and ferromagnesian mineral dissolution, plagioclase weathering possible
	🟡	h	Recharge water in SiO_2 medium sands, located on local higher topographic elevation. Contains higher concentrations of metals such as Al.	Silicate weathering and ferromagnesian mineral dissolution, pyrite oxidation, plagioclase weathering possible, sodium from albite and/or ion exchange
Leachate	🔴		Wells located underneath the waste pile or within 20 m. Elevated concentrations for all major ions. NH_4 and Cl are characteristic.	Dolomite dissolution, calcite precipitation
Attenuated Leachate	◆	i	Leachate mixing with neutral semi-anoxic water in a heterogeneous geological setting, “anoxic path” samples are located >100 m from the former waste pile	Ion exchange, ions precipitation, sorption, iron reduction, calcite precipitation
	🟢	j	Leachate mixing with oxic and acidic water in a homogeneous geological setting “oxic path” samples are located within 100 m from the former waste pile	Ion exchange, iron precipitation, calcite solution

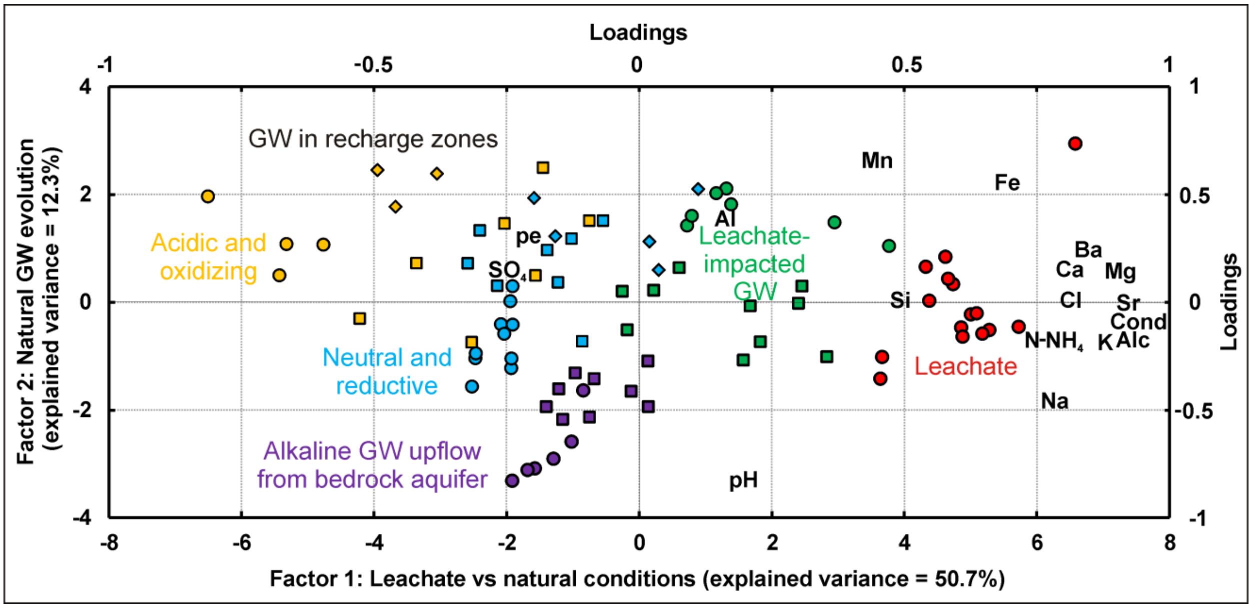


Figure 6.8 : Principal component analysis (PCA) of groundwater

PCA of groundwater in the study area distinguished by HCA groups (Figure 6.7; symbols are also defined in Table 6.6). The graphs show the values of the main factors for groundwater samples as well as the relative loadings of chemical elements (black text)

PCA is commonly used to evaluate similarities between elements or to diminish the number of variables for further statistical analysis (Farnham 2002), but it was used in this study more to identify the variables that control HCA groups and to gain insight on the processes that drive natural groundwater evolution and leachate plume attenuation. Figure 6.8 illustrates the results for PCA of the same transformed and standardized groundwater samples as HCA. Figure 6.8 shows both the loadings of samples for the two first principal components and for the geochemical variables (shown by the location of black symbols on the graph). Since many chemical parameters have high values in the leachate plume, the first principal component, which explains 50% of the sample variance, is related to the distinct concentrations between natural groundwater and leachate, with leachate-impacted groundwater located at an intermediate position between natural groundwater and leachate. Natural groundwater and leachate-related groundwater are respectively on the negative and positive side of principal component 1. Principal component 2 is more related to natural groundwater conditions and its evolution represented by changes in pH and pe conditions (Figure 6.9). Acidic groundwater containing dissolved metals, such as iron, manganese or aluminum, has positive values of principal component 2 (vertical axis). In comparison, alkaline groundwater with higher concentrations of sodium and potassium has negative values of principal component 2. Figure 6.8 clearly shows the different origins of the groundwater found in the aquifer: 1) high recharge zone groundwater that is acidic and oxidizing; 2) upflow of alkaline and reducing groundwater from the sedimentary rock aquifer; and 3) landfill leachate. Groundwater and leachate geochemically evolve along different paths due to their encountering different redox and pH conditions as well as mixing with waters of different compositions. HCA subgroups provide indications of these processes.

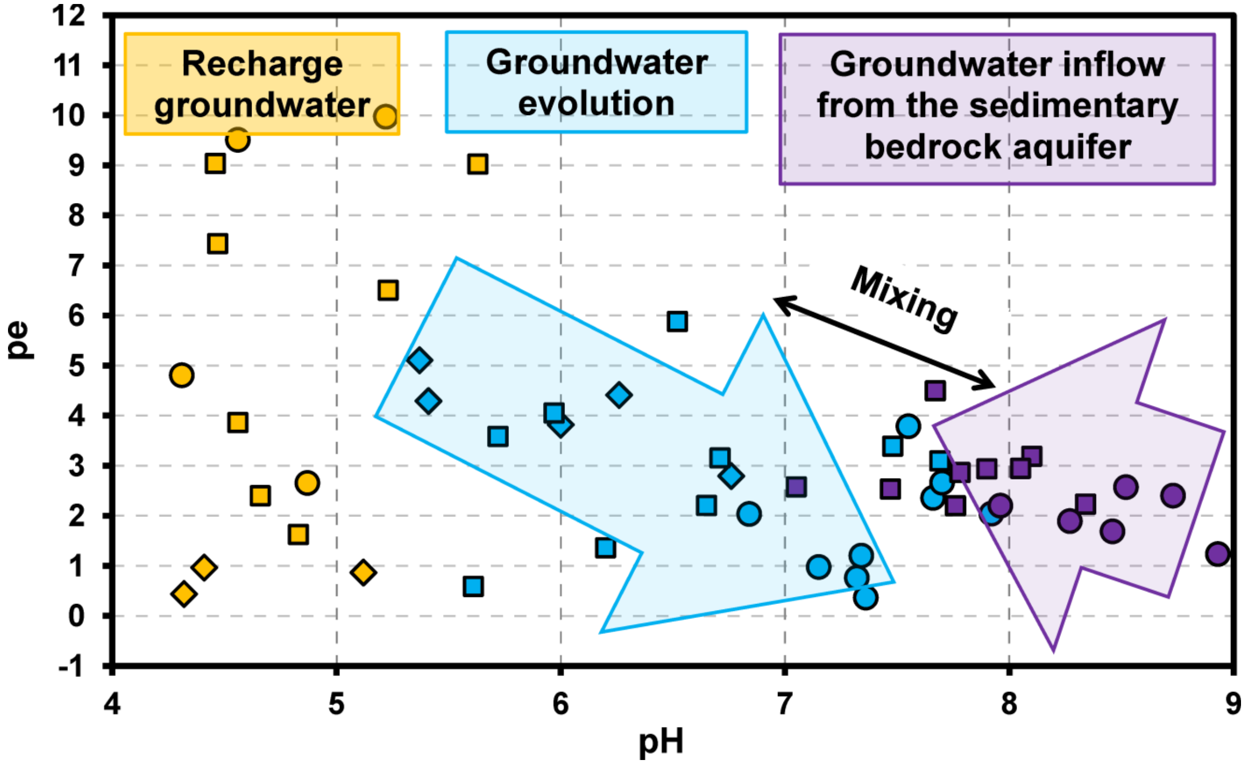


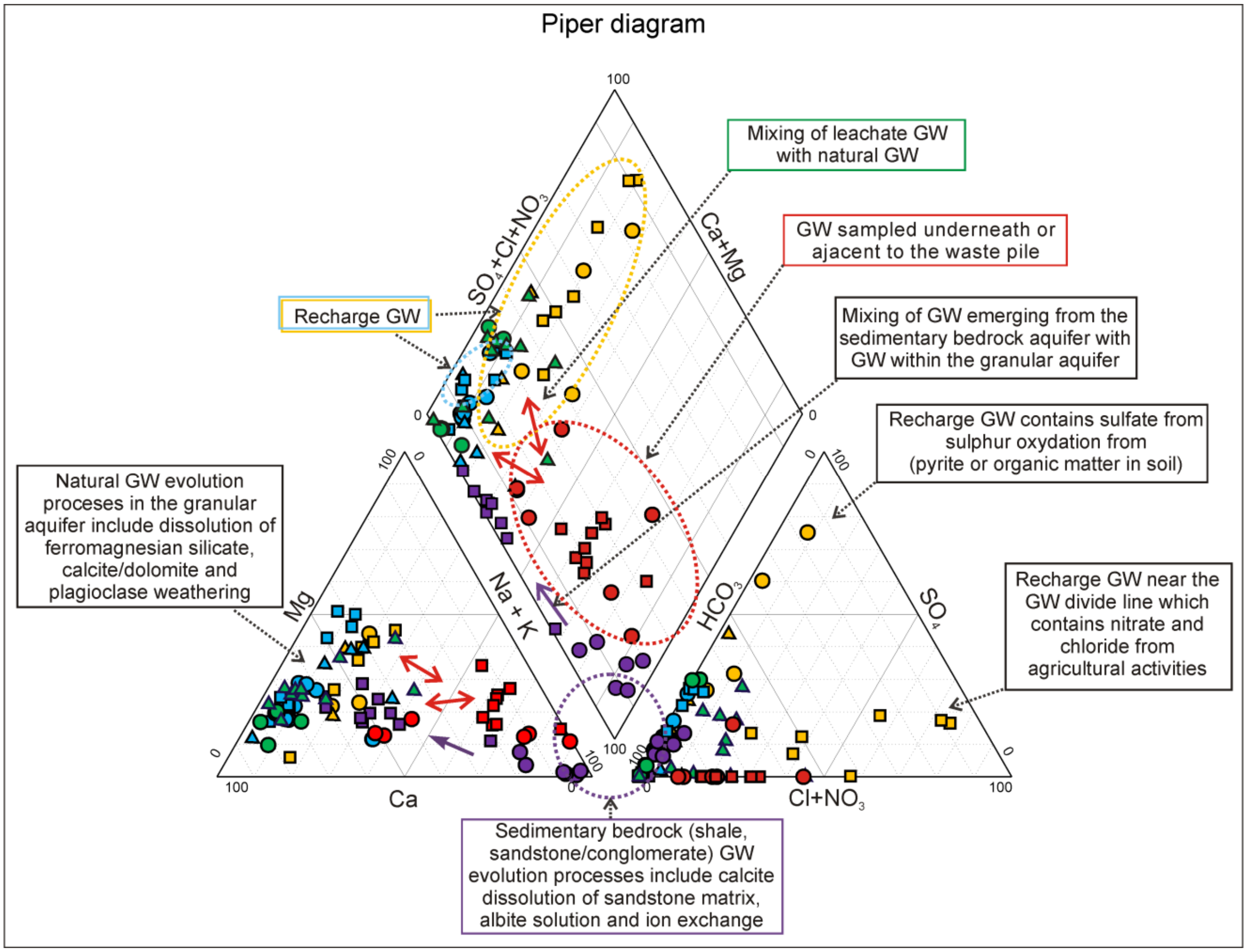
Figure 6.9 : Natural groundwater pH-pe diagram

pH-pe diagram illustrating changes in conditions due to the evolution of natural groundwater. Natural groundwater composition evolves from two origins: recharge zones and upflow from the underlying sedimentary rock aquifer

HCA groups can be further subdivided into subgroup based on sample similarities. Table 6.6 describes the main distinguishing features between HCA groups and subgroups as well as possible chemical processes that control the geochemistry of these groundwaters. Figure 6.9 illustrates pH-pe conditions for natural groundwater in the aquifer system with symbols related to their HCA subgroups (Figure 6.7 and Table 6.6). pH-pe conditions are quite diagnostic of natural groundwater evolution: 1) groundwater from recharge zones is acidic and mostly oxidizing and groundwater becomes more neutral and less oxidizing as it evolves; and 2) the upflow of anoxic and alkaline groundwater from the sedimentary rock aquifer imparts a distinct signature to pH and pe conditions as it mixes with groundwater in the aquifer. pH-pe conditions also identify two distinctive leachate attenuation paths, depending on the conditions of groundwater in which leachate migrates (Table 6.6): 1) "oxic path": a leachate-impacted groundwater subgroup originates from the mixing of leachate with oxic and acidic groundwater in a relatively homogeneous geological setting suggested from samples within 100 m from the former waste pile; 2) "anoxic path": a leachate-impacted groundwater subgroup is related to leachate mixing with neutral semi-anoxic water in a heterogeneous geological setting farther than 100 m from the former waste pile. As a result, since natural groundwater conditions are highly variable at the subwatershed scale, depending on the natural conditions found along the leachate migration path, different reactions will take place and different geochemical signatures will be found downstream from the landfill. The effects of hydrogeological and geochemical heterogeneity on the natural attenuation of landfill leachate are further discussed by Tremblay (2013; chapter 7).

All samples are plotted on a piper diagram (Figure 6.10) to further illustrate the diverse conditions found in the aquifer and the geochemical processes controlling groundwater evolution and leachate plume attenuation (the main processes related to HCA Subgroups are also summarized in Table 6.6):

- Recharge groundwater and natural groundwater in the aquifer have a variety of compositions, depending on surficial conditions at recharge locations. Natural groundwater also exhibits many geochemical evolution trends depending on the nature of aquifer materials and conditions along flow paths. On the diamond of the Piper diagram, recharge groundwater plots on the left side, which corresponds to the Ca-HCO_3 water type typical of most recharge waters. Subgroups c, d and e of the Neutral HCA Group remain at that position, but are distinguished by the occurrence of iron or sulfate. Groundwater related to HCA Acidic subgroups show two distinct evolution paths related to increases in SO_4 due to pyrite oxidation or in Cl and NO_3 related to infiltration through agricultural land. Natural groundwater geochemical processes affecting recharge groundwater are wide-ranging: silicate weathering and ferromagnesian mineral dissolution, pyrite oxidation, calcite/dolomite dissolution and plagioclase weathering (sodium from albite and/or ion exchange) (Table 6.6).


Piper diagram

Mixing of leachate GW with natural GW

GW sampled underneath or ajacent to the waste pile

Recharge GW

Mixing of GW emerging from the sedimentary bedrock aquifer with GW within the granular aquifer

Recharge GW contains sulfate from sulphur oxydation from (pyrite or organic matter in soil)

Natural GW evolution processes in the granular aquifer include dissolution of ferromagnesian silicate, calcite/dolomite and plagioclase weathering

Recharge GW near the GW divide line which contains nitrate and chloride from agricultural activities

Sedimentary bedrock (shale, sandstone/conglomerate) GW evolution processes include calcite dissolution of sandstone matrix, albite solution and ion exchange

Figure 6.10 : Piper diagram

Piper diagram representing the proportions of major ions in groundwater distinguished by HCA Groups. Figure 6.7 and Table 6.6 provide the definitions of symbols, but triangles replace diamonds for every group, red circles represent wells installed just below the waste and red squares are wells located near the pile (not distinguished by the HCA Subgroup symbols). Boxes provide descriptions of the main geochemical processes controlling GW evolution. Red and green color markers represent groundwater affected by leachate and the proportions of ions on the Piper diagram for these samples are not representative due to the presence of high concentrations of iron and ammonium that are not considered on the diagram

- Besides recharge groundwater, another source of groundwater in the aquifer system is HCA Alkaline Group groundwater emerging from the regional sedimentary rock aquifer. At their origin, this evolved groundwater is of Na-HCO_3 type and thus plots on the Piper diagram at the merging point of the two triangles and the lower tip of the diamond. Mixing of groundwater from the rock aquifer with the Neutral group groundwater in the aquifer makes it evolve with an increase in Ca. (and to a lesser extent Mg) at the expense of Na. The main processes affecting the natural evolution of the Alkaline group groundwater involve carbonate weathering (limestone or dolomite) and cation exchange (Na may be from albite and/or ion exchange) (Table 6.6).

- Finally, the other source of groundwater in the aquifer is leachate, which is recognized as HCA Leachate and Attenuated Leachate groups (Table 6.6). The occurrence of leachate waters is restricted to the waste pile or within about 20 m of it. Like Alkaline water, Leachate water is characterized at its origin by a high proportion of Ca, but it is distinct from Alkaline water by a larger proportion of Mg and especially by the presence of Cl. Leachate groundwater mixes with each of the other water types (Acidic, Neutral, Alkaline) depending on the geographical location, leading to a wide distribution of Leachate water composition on the Piper diagram and, eventually, to the Attenuated Leachate group plotting on the Piper diagram at a similar location as recharge groundwater (both Acidic and Neutral). However, Attenuated Leachate waters are distinct from natural groundwater in terms of minor components and, like Leachate waters, their composition cannot be well represented on the Piper diagram due to the presence of high concentrations of iron and ammonium that are not considered on the Piper diagram. Leachate waters are affected by dolomite dissolution and calcite precipitation, whereas Attenuated Leachate is affected by ion exchange, ion precipitation, sorption, iron reduction or precipitation and either calcite precipitation or dissolution. The processes affecting Attenuated Leachate depend on distance from the waste pile (Table 6.6).

6.5.2.2 Effect of aquifer matrix on geochemical groundwater evolution

Jankowski and Beck (2000) demonstrated that hydraulic conductivity and groundwater chemistry vary in relation to changes in depositional environment and that chemical reactions resulting from water-sediment interaction produced significant changes in groundwater chemistry, which is a function of mineralogy, lithology and residence time in the aquifer. Moreover, McMahon and Chapelle (2008) stated that geological factors such as sediment source and depositional environment can influence the availability of natural electron acceptors and donors in the subsurface. Therefore, to initially assess the impact of sediment heterogeneity on groundwater geochemical signatures and evolution in the study area, carbonate dissolution in groundwater and saturation indices (SI) for minerals identified in sediments with XRD were further investigated.

Carbonate minerals are ubiquitous and the degree of carbonate weathering is largely controlled by the availability of CO_2, while limited by the saturation of calcite (Clark 2012). Tremblay et al. (2013) investigated this issue in two distinct recharge zones in the study area, finding that the dissolution of carbonate minerals is highly related to the material in place and its heterogeneity. Figure 6.11 shows the evolution of pH and pCO_2 for groundwater samples in the study area distinguished by their HCA Group (Figure 6.7 and Table 6.6). DIC and pH values were used to calculate pCO_2 for all samples. Figure 6.11 shows the dissolution of carbonates under open and closed system conditions. The evolution of pH and pCO_2 is linked to calcite dissolution (Clark 2012), as groundwater dissolves calcite under open or closed system condition. The graph distinguishes the HCA water subgroups to explain the impact of calcite dissolution on these different water types (HCA symbols are defined in Figure 6.7 and Table 6.6). P03 is a well located just east of the groundwater divide (groundwater samples from 5 to 11 m depth are illustrated by yellow squares in Figure 6.11), close to a former channel formed by the Chaudière River. Ground elevation is higher and sediments are homogeneous and coarser (with higher hydraulic conductivities; Paradis et al. 2011) compared to sediments found elsewhere in the study area. Moreover, as seen in Figure 6.4 there is a smaller proportion or no carbonate at all in the sand at this location. Therefore, there is no buffer for the acidic groundwater, and groundwater travels faster and the pH remains the same along groundwater flowpaths at this location in the aquifer. As seen in Figure 6.11, pCO_2 and pH conditions (as well as Ca concentrations; not shown) remain similar at this location. Symbols on the graph also distinguish recharge zones where sediments are homogeneous (yellow circles), shallow groundwater sampled just below the unsaturated zone in intermediate or in discharge zones (yellow diamonds), groundwater at shallow depth where the aquifer is composed of silty material (blue diamonds), groundwater above 8 m depth containing dissolved iron (blue squares), groundwater below 8 m depth in heterogeneous settings without dissolved iron (blue circles), and groundwater found at various depths representative of upflow from the sedimentary rock aquifer (purple symbols). Leachate and leachate-impacted groundwaters clearly have pH-pCO_2 conditions distinct from natural conditions, which show quite a range of pH-pCO_2 conditions. Therefore, the graph also confirms that HCA groups accurately distinguish anthropogenic versus natural conditions. The pH-pCO_2 evolution of natural groundwater also provides indications of confining conditions, as the amount of dissolved calcite is ultimately controlled by the partial pressure of CO_2 of the system, which depends on the open or closed nature of the system. As groundwater below the water table consumes carbonic acid through weathering, its pCO_2 drops and carbonate alkalinity increases (Clark 2012). Once again, the graph shows that the type of material in place controls carbonate dissolution in groundwater in relation to the types of sediments present in the aquifer.

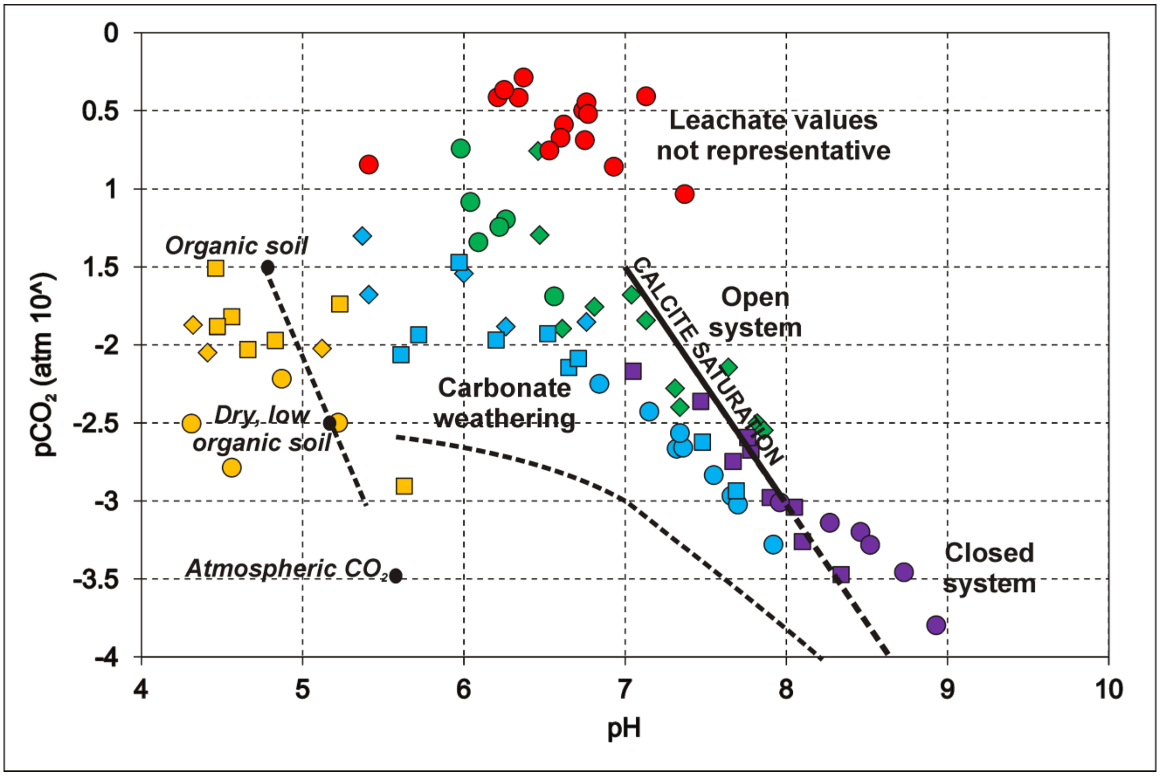


Figure 6.11 : Carbonate cycle within the hydrogeological system

Carbonate cycle within the system under study for all groundwater samples, as illustrated by pCO_2 versus pH, which distinguishes between closed and open systems (adapted from Clark 2012) (HCA groups represented by the symbols are defined in Table 6.6)

Saturation indices of minerals present in aquifer sediments, based on XRD, are illustrated in Figure 6.12 by HCA group. As expected, quartz is supersaturated, which does not imply that quartz will precipitate (Zhu and Anderson 2002). Otherwise, all natural groundwaters are undersaturated relative to minerals found in the aquifer sediments. However, natural groundwater is supersaturated relative to various aluminum and iron oxides, such as hematite (Figure 6.12).

Even though SI may indicate that a mineral will tend to dissolve or precipitate, it may not occur because of kinetic constraints and other factors, such as solid solutions complicating the mineral composition, the chemical formula of the minerals that may increase the magnitude of the SI, errors in analyses, the reliability of the thermodynamic data, etc. (Zhu and Anderson 2002). Furthermore, the SI does not provide information about reaction rate or the mass of the mineral involved (Zhu and Anderson 2002). Therefore, this analysis only provides indications of potential mineral reactions in relation to the evolution of natural groundwater or leachate in the aquifer system.

Some general observations from Figure 6.12 are that fluoride and phosphorous are only found in natural groundwater group 3, which represents the sedimentary rock groundwater signature. All copper-bearing minerals also show supersaturation for group 3 groundwater. Saturation indices increase for most minerals from group 1 to 3, accordingly with the natural groundwater geochemical evolution. SI's are higher in leachate groundwater for Ca-, K- and Na-bearing minerals.

The groundwater geochemistry also provides indications about redox-sensitive mineral species. Precipitation of FeS, FeS_2 and $FeCO_3$ may deplete groundwater from sulfur or iron and $MnCO_3$ may deplete groundwater from Mn (Christenson 1994). Other studies on leachate plumes emitted by landfills demonstrated that groundwater affected by leachate is often of surpersaturated relative to calcite and siderite. Since iron-reduction kinetics is important for assessing long-term potential for aquifer attenuation (van Breukelen 2003; Keldsen 2002), and that siderite and calcite are key minerals that precipitate along leachate migration, SI in relation to iron, manganese and calcite should be further investigated. Finally, in general, it is difficult to identify key saturation indices specific to a site for geochemical modeling. The chemical elements found in high concentrations in leachate also have elevated SI values for all the mineral indices containing these same chemical elements. Therefore a detailed description of the chemistry of the aquifer material can help define the metal species that can participate in reactions.

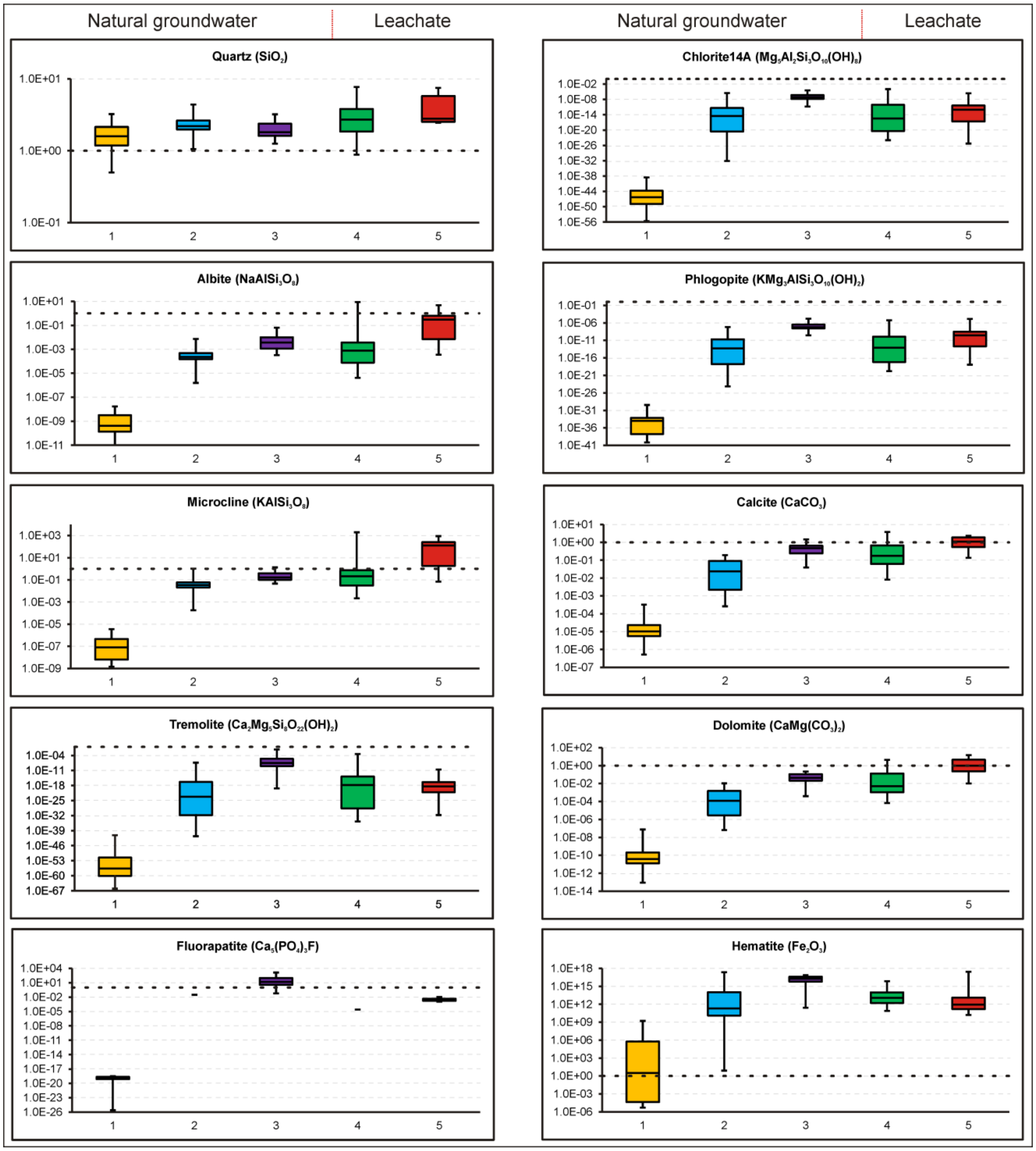


Figure 6.12 : Groundwater saturation indices

Saturation indices for the minerals identified in aquifer sediments with XRD. The saturation indices for hematite are also shown to illustrate that groundwater is supersaturated relative to many iron and aluminum oxides as well as potassium-bearing minerals. Fluor and phosphorous are only found in natural groundwater group 3, which represents the sedimentary rock aquifer signature. Saturation indices increase for most minerals from group 1 to 3, which represent groundwater evolution. Concentrations are higher in leachate groundwater (group 5) for Ca-, K- and Na-bearing minerals. Figure 6.7 and Table 6.6 describe HCA water Groups and Subgroups

6.5.3 Conceptual model revisited based on sediment and groundwater geochemistry

Figure 6.13 illustrates the detailed conceptual model of the aquifer system in the study area developed on the basis of the integrated characterization data conducted in three phases (see respectively Tremblay et al. 2013 and Tremblay 2013 for phases 1 and 2). The geochemistry of sediments and groundwater types allows refining the model by confirming flowpaths, the evolution of natural groundwater, as well as the leachate plume extent and its two natural attenuation paths (Figures 6.9 and 6.11). Figure 6.13 also illustrates the hydrogeological conceptual model with geochemical conditions and components related to different sediment types. Numbers refer to observation well locations and Stiff diagrams are located in the general area where samples were collected. The groundwater geochemical signature of the sedimentary rock aquifer confirmed the provenance of this water type which emerges locally in the granular aquifer.

In Figure 6.13, the extent of ammonium and chloride concentrations exceeding 100 mg/l illustrates that the leachate plume is readily attenuated and remains within the property limits. Chloride is a conservative contaminant only attenuated by dilution. From plume mapping data, degradation of organic matter may be evaluated by comparing the organic matter plume with the chloride plume, the latter expressing the degree of dilution. However, such a comparison assumes that the organic matter does not significantly sorb onto the aquifer material and that the organic matter to chloride ratio has been constant in the infiltration of leachate during the period in question (Ford et al. 2008; Christensen et al. 1994). Based on the interpretation of geochemical data and processes, the detailed conceptual model already established was refined. The final conceptual model describes in detail the conditions under which the leachate migrates and the distinct natural conditions under which it will attenuate. The conceptual model also considered the influence of the geology (physical and geochemical conditions) in the evolution of natural groundwater and for its capacity to immobilize contaminants and attenuate the leachate in relation to concentrations found at the source. Ford et al. (2008) stated that the underlying basis of a good conceptual model resides from a site characterization plan that considers potential reactions of the contaminant within the solid phase, as well as changes to the solids that may impact contaminants mobility. Ford et al. (2008) also added that acquisition of site specific information is critical for developing regulatory and public acceptance of MNA as a viable remedy.

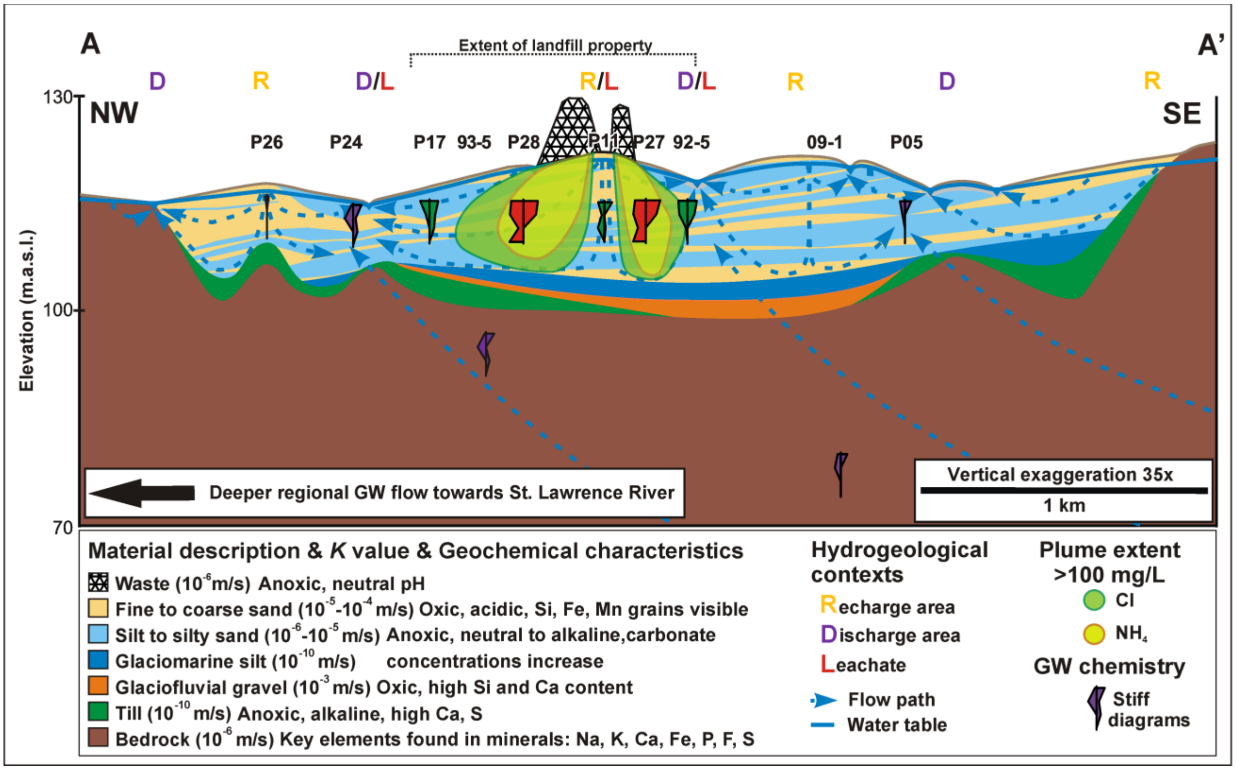


Figure 6.13 : Hydrogeological conceptual model of the study area

Hydrogeological conceptual model of the study area with geochemical conditions and components related to different sediment types. Numbers refer to observation well locations and Stiff diagrams are located in the general area where samples were collected. The groundwater geochemical signature of the sedimentary rock aquifer confirmed the provenance of this water type which emerges locally within the granular aquifer. The extent of ammonium and chloride concentrations exceeding 100 mg/l illustrates that the leachate plume is readily attenuated and remains within the landfill site property limits (modified from Tremblay et al. 2013)

6.6 Discussion

6.6.1 Field collection, analytical method and data interpretation

6.6.1.1 Sediments

The mineralogy of soils and aquifer sediments is not often considered as part of groundwater studies (Zhu and Anderson 2002). However, the present study shows that mineralogical data can help understand the geochemical conditions of an aquifer system. In the present study, the combination of a few analytical methods to a logical selection of samples provided explanations for the influence of the soils and aquifer sediment geochemistry on natural composition of groundwater. Furthermore, these analyses provided insights on the controls of leachate natural attenuation in the aquifer system.

The chemical and mineralogical data were obtained through an efficient combination of methods. Even though alkaline fusion analysis provides a complete data set compared to the XRF method, XRF measurements with a portable analyzer are much faster and require less sample preparation and lab time. Therefore, combining the two methods allows the efficient characterization of the aquifer matrix and the data set can be completed with results from alkaline fusion data used to calibrate the XRF data. The methodological framework employed to evaluate the general geochemical and physical properties of aquifer sediments (Figure 6.2) provides the range of values of aquifer sediment proprieties at the subwatershed scale, while reducing laboratory costs, sample preparation and analytical delays.

Interpretation of sediment chemical data through multivariate analysis allowed the understanding of the depositional environment that was related to their physical and geochemical properties. pH and pe measurements on sediments preserved from oxidation provided complementary information on the potential for the aquifer to attenuate and biodegrade contaminants. The investigation demonstrated that the geological environment not only controls redox conditions found in the aquifer, but also pH conditions that also greatly influence reactions that can take place and the element dissolved in groundwater. The mineralogical analyses also showed some variability between HCA groups but demonstrated that the aquifer matrix had mostly the same mineralogical composition, but in differing proportions. Porosity measurements showed high values, offering a good potential for important adsorption sites. The overall analysis provided the data required to understand spatial variability in the geological environment as well as associated chemical and physical characteristics that influence groundwater flow and contaminant migration and attenuation. Moreover, results provided the needed information to select saturation indices for further geochemical interpretation. Overall, sediment characteristics imply a high aquifer capacity to sustain attenuation. However, the data obtained are still incomplete, as they do not provide the information required to quantify the mass of electron acceptors, the rates of attenuation reactions, nor the long-term stability of immobilized contaminants. Still, the obtained sediment data provided valuable information on the spatial variability of geochemical conditions.

6.6.1.2 Groundwater

Groundwater was sampled in different geological contexts at a subwatershead scale in a study area encompassing source zone and receptors (mainly streams). Fully-screened direct push wells were preferred since sampling could be easily repeated at selected depths. The 5 cm (2 in) diameter wells also allowed the measurement of multiple parameters with a variety of instruments (flowmeter, YSI for geochemical profiling and borehole geophysics). Though the fully-screened wells have limitations, depending on the magnitude of vertical hydraulic gradients and hydraulic conductivity, the long screened wells can still provide the general trend for hydraulic conductivity and geochemistry at a specific location and orient the understanding of the study site where more depth-specific surveys can be carried out. Long screened wells can induce cross-aquifer groundwater exchanges in multilayered aquifers as groundwater from the horizon with the greatest hydraulic head flows into the wellbore and displaces water from a horizon with lower hydraulic head. Water can move up or down the wellbore, depending on the direction of vertical head gradients. While pumping can remove the mixed groundwater from the wells and contaminated horizon, the required volume of water purged from a well to help ensure a representative sample of the aquifer is unknown since the true extent of cross-contamination is difficult to quantify. Long screen wells dilute water drawn from contaminated horizons with water from non-contaminated horizons, while short screen wells and discrete sampling may bias the sample by either missing contaminated or non-contaminated horizons (Hofmann 2010; Mayo 2010; Appelo and Postma 2005). However, McMahon and Chapelle (2008) stated that although it might be expected that wells with longer screens would produce more mixed redox diagnoses than wells with shorter screens, the opposite pattern was observed in their study on redox processes and water quality. In the present study, a well that intersects a more permeable unit at the base of the aquifer, where the pressure gradient is greater (strong reverse gradient), will give the geochemical signature of that specific interval through the groundwater at shallower depth of the well. Therefore the geochemical signal recorded at each location, combined with the flowmeter response, is an indication of the local conditions which, when combined with other data, help understand the flow dynamics at a small scale. Multiple wells with discrete (1 m) screen intervals were also installed throughout the study area at specific depths to validate the geochemical data obtained previously from long screens. As wells were left open for a few years before geochemical sampling occurred, mixing in the wells with strong vertical hydraulic gradients took place. To minimize the impact of cross-contamination of geochemical parameters with a long screened borehole, wells should be completely sealed between sampling events.

The collected groundwater geochemical data showed significant variability in groundwater types within a small study area. Such information on the spatial distribution of geochemical conditions is required to understand the impact of a leachate plume on the natural environment. The interpretation of groundwater data provided the information required to understand varied natural conditions and groundwater evolution, as well as the geochemical composition of the leachate plume and its variability. However, there was a single geochemical sampling campaign in 2010. There is thus no temporal geochemical data available to assess groundwater composition variability through time. This study thus provides a snapshot of groundwater geochemical conditions that still allows an understanding of both natural and leachate-impacted groundwaters.

6.6.2 Relation between aquifer matrix and groundwater geochemistry

Groundwater geochemistry was found to be controlled in large part by surface conditions and the chemical properties of aquifer sediments. These surface conditions and sediment chemistry not only affected the presence of major or minor ions, but also controlled the redox and pH conditions of the groundwater. Furthermore, since the heterogeneity, expressed mostly as an interstratification of fine and coarse materials, also affected groundwater recharge and flow rates, it also affected the residence time of groundwater which impacts the level of geochemical evolution that groundwater can attain in the aquifer.

Redox processes affect the chemistry and quality of groundwater, as they largely control the mobility of metals associated with naturally occurring materials. Redox conditions also influence the degradation, or the preservation, of anthropogenic contaminants and generate undesirable by-products, such as dissolved ferrous iron, hydrogen sulfide and methane. McMahon and Chapelle (2008) state that geological factors such as sediment source and depositional environment can influence the availability of natural electron acceptors and donors in the subsurface. Determining the kinds of redox processes occurring in an aquifer system, documenting their spatial distribution, and understanding how they affect concentrations of natural or anthropogenic contaminants are thus essential to assess the chemical quality of groundwater (McMahon and Chapelle 2008).

6.6.3 Implications for plume migration and natural attenuation

The groundwater geochemical data allowed the identification of the extent of the leachate plume, and leachate-impacted groundwater, and showed the variability of concentrations found within the plume. The geochemical data show that attenuation processes lead to sharp devrease in concentration along flowpaths, so that no concentrations exceeding provincial standards were found outside the landfill property limits. Moreover, heavy metals and dissolved organic carbon decrease readily.

The investigation of groundwater ages and residence time by Tremblay (2013) (paper 2) demonstrated that it would take 40 years for groundwater to reach the base of the aquifer from the center of the landfill. Since the decommissioned landfill started its operations almost 50 years ago, the leachate plume has clearly reached the base of the aquifer. However, there is some uncertainty regarding the future behavior of the plume. The behavior of cations in leachate plumes is strongly influenced by exchange with sediments; exchange reactions and dilution create a very complicated migration pattern for cations in plumes (Christensen 1994). High iron and manganese concentrations, as indicated by reddish oxic aquifer sediments (high oxidation capacity), will strongly reduce the methanogenic and sulfate reducing zone and lead to a larger iron and manganese reduction zone. Therefore, the aquifer geochemistry, including iron and manganese minerals, is a key factor in understanding the formation of redox zones. The aquifer capacity to attenuate contaminants is hard to quantify. However, some indicators provide strong indications on the efficiency for the aquifer to attenuate contaminants. Contaminant concentrations that strongly decrease in the flow direction and the natural conditions of the aquifer indicate that the aquifer has a high attenuation capacity. The identification of redox zones based on aquifer sediment mineralogy and groundwater geochemistry is also important. The distribution of contaminants between solid-bound (sorbed) and aqueous phases is an important aspect of risk assessment and remediation design in the context of both groundwater and surface water resources. In the study area, sediment mineralogy proved to be a major factor determining the relative importance of oxic and anoxic conditions and processes. The data also indicated two distinct leachate attenuation paths, (oxidizing path vs anaerobic path) that is discussed in more detail by Tremblay (2013; chapter 7). Thus, in the same area, leachate attenuation processes are different depending on heterogeneous natural geochemical conditions in the aquifer that in turn depend on heterogeneous hydrogeological conditions.

6.6.4 Further work needed for leachate plume risk management

When properly assessed and employed, Monitored Natural Attenuation (MNA) may provide an effective contamination management approach. The assessment of MNA requires a characterization documenting many conditions: mass reduction, the occurrence and rates of specific MNA processes and the occurrence of microbial degradation. The present study has limitations for the assessment of MNA since it did not provide the knowledge of all potential mechanisms and rates of contaminants attenuation, the capacity of the aquifer to sustain attenuation of the released mass of contaminant and the long-term stability of immobilized contaminants, and the spatial variation and types of solid-phase reactants. Furthermore, the long-term stability of immobilized contaminants needs to be verified, which could not be done without temporal monitoring of geochemical conditions. Moreover, if recharge conditions or landuse were to change, a reevaluation of MNA and monitoring through time at strategic locations would be required. As a next step in the assessment of MNA in the study area, a numerical model of groundwater flow and mass transport will be developed to evaluate the fate of the leachate plume through time, as well as a more detailed interpretation of geochemical data (Tremblay et al. 2013).

6.7 CONCLUSIONS

This study makes scientific contributions to the understanding of factors controlling the heterogeneous geochemical conditions that can be found in granular aquifers, and the interrelation between sediment properties and geochemical conditions, which themselves influence both the natural evolution of groundwater and the attenuation of landfill leachate.

Chemical heterogeneities of sediments and groundwater in a granular aquifer were investigated in a 12 km^2 subwatershed where a decommissioned unlined landfill is located. An efficient workflow for soil and aquifer sediment characterization was developed to gain information about the chemical and physical variability of aquifer material. Groundwater geochemical conditions across the aquifer were also defined through the chemical analyses of a wide range of parameters. Multivariate analysis of the sediments and groundwater chemical data involved hierarchical cluster analysis and principal component analysis. The multivariate analysis allowed the definition of groups whose conditions were related either to sediments or groundwater conditions.

Results showed a great variability in geochemical conditions in the aquifer that is controlled by the nature of surface conditions and sediments properties. The sediments and groundwater geochemical data provided evidence of groundwater origin through different recharge conditions but also from the upflow of groundwater from the underlying regional sedimentary rock aquifer. The data analysis thus showed quite heterogeneous natural geochemical conditions that could be distinguished from leachate or leachate-impacted waters. Finally, the data allow the recognition of geochemical evolution of natural groundwater as well as leachate and the processes likely responsible for this evolution. The leachate was found to follow two attenuation paths related to redox conditions of the groundwater it comes in contact with. Results show that the aquifer has a good capacity to attenuate the leachate plume.

In view of the varied redox conditions of natural groundwater that influence leachate attenuation, ORP and pH measurements of aquifer sediments proved very important as controls on groundwater conditions but also to infer the ability of the aquifer to attenuate and biodegrade contaminants. Sediment properties also provide insight into the history and conditions of deposition of the sediments, and thus relate sedimentology, geochemistry and hydrogeological contexts.

Geochemical data were finally useful in refining the conceptual model of groundwater flow and in understanding natural geochemical processes that occur along flow paths as well as leachate plume attenuation processes. This improved knowledge of the site conditions will lead to a better assessment of the environmental risk to groundwater receptors with respect to the leachate plume.

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Chapitre 7

Effects of Hydrogeological and Geochemical Heterogeneity on Natural Attenuation of a Leachate Plume

TITRE FRANÇAIS :

Effets de l'hétérogénéité hydrogéologique et géochimique sur l'atténuation naturelle d'un panache de lixiviat

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CONTRIBUTIONS:

Laurie Tremblay a planifié et coordonné l'acquisition des données de terrain. Elle était personnellement responsable de l'acquisition, du traitement et de l'interprétation des données géochimiques. Elle a aussi réalisé la modélisation numérique de l'écoulement et du transport de masse. Au niveau de l'article, elle a réalisé les figures et écrit la version préliminaire de l'article.

René Lefebvre a coordonné le projet, encadré les étudiants impliqués et révisé l'article.

Vincent Cloutier a encadré l'étudiante par rapport aux aspects géochimiques et a révisé la version préliminaire de l'article.

John Molson a supervisé la réalisation du modèle numérique et a révisé la version préliminaire de l'article.

REVUES CONSIDÉRÉES POUR LA PUBLICATION :

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RÉSUMÉ

Un panache de lixiviat émis par un ancien site d'enfouissement sanitaire dans un aquifère granulaire peu profond est géré par atténuation naturelle. Afin d'évaluer l'efficacité de ce mode de gestion, des travaux antérieurs ont porté sur la caractérisation détaillée du sous-bassin versant de 12 km^2 entourant l'ancien site d'enfouissement. Ces travaux ont montré des conditions hydrogéologiques et géochimiques hétérogènes et ont permis de développer un modèle conceptuel détaillé du système aquifère. Sur cette base, la présente étude a pour objectif d'évaluer les effets de ces différents types d'hétérogénéité sur l'atténuation naturelle du panache de lixiviat. Les effets de l'hétérogénéité hydrogéologique ont été évalués à l'aide d'un modèle numérique 2D coupe traversant le site d'enfouissement le long des voies d'écoulement et de migration du lixiviat. Des efforts particuliers ont été dédiés à la vérification de la représentativité du modèle numérique à l'aide de données hydrauliques mais aussi de données géochimiques (âge de l'eau et traceurs géochimiques). Le modèle numérique a montré que l'alternance de couches fines et grossières limitait l'écoulement et conduisait à des temps de résidence relativement longs (40-60 ans) dans la partie inférieure de l'aquifère où migre le panache. De plus, le panache de lixiviat est fortement dilué lors de sa lente migration; ce qui favorise son atténuation naturelle avant son émergence dans les cours d'eau. L'analyse graphique des données géochimiques a permis d'identifier deux voies d'atténuation naturelle du lixiviat en fonction des conditions géochimiques de l'eau souterraine dans laquelle migre le lixiviat. Les mécanismes identifiés d'atténuation du lixiviat réduisent les concentrations en ions majeurs et métaux sur de courtes distances, de sorte que l'eau souterraine n'excède aucune norme en dehors des limites de propriété du site d'enfouissement. La compréhension des conditions de l'aquifère permet une meilleure évaluation du risque environnemental que représente le panache de lixiviat sur les milieux récepteurs, particulièrement les cours d'eau de la région d'étude.

MOTS-CLÉS: hydrogéochimie, processus d'atténuation naturelle, modélisation numérique, aquifère granulaire, panache de lixiviat.

ORIGINALITÉ ET PERSPECTIVE :

Des études antérieures ont évalué les effets de l'hétérogénéité en relation avec des problèmes de contamination de l'eau souterraine par la modélisation numérique. De plus, plusieurs études ont modélisé l’atténuation naturelle d’un panache de lixiviat. Par la même occasion, ces études ont évalué les réactions géochimiques et quantifié les taux d’atténuation des composés d’intérêts à une échelle très locale (< 200 m). Par contre, peu d’étude ont évalué à la fois l’hétérogénéité des conditions hydrogéologiques et géochimiques et considéré leurs effets sur l’atténuation naturelle du lixiviat à l’échelle d’un sous-basin versant incluant les zones sources et les récepteurs. De plus, peu d’études ont considéré la migration d’un panache de lixiviat à travers différentes conditions géochimiques dans un même aquifère. Les travaux décrits dans l’article ont ainsi les aspects novateurs et originaux suivants par rapport aux études antérieures sur ce sujet :

Échelle d’étude considérée

L’étude prend en considération un aquifère granulaire peu profond compris dans un sous-bassin versant de 12 km^2, incluant les zones sources et les récepteurs, pour considérer autant les conditions naturelles que les conditions du panache de lixiviat. Peu d’études sur le lixiviat ont été réalisées à une échelle de sous-bassins versants ou pris en considération les contextes de différentes échelles d’investigation. Cette échelle d’étude permet de comprendre la migration et l’atténuation naturelle d’un panache de lixiviat à partir de son origine jusqu’aux récepteurs que sont les cours d’eau, permettant ainsi de mieux évaluer le risque environnemental sur ces récepteurs.

Considération de l’ensemble des composantes d’un système hydrogéologique

L’étude intègre toutes les données de la caractérisation multidisciplinaire réalisée précédemment en trois étapes pour développer et valider un modèle conceptuel représentatif du système aquifère investigué. Les composantes intégrées représentent les aspects principaux qui peuvent influencer l’écoulement de l’eau souterraine et la migration d’un panache de lixiviat, notamment la recharge spatialement distribuée. Les données de sources variées ont été intégrées pour développer un modèle numérique de l’écoulement et du transport de masse représentatif de l’hétérogénéité de l’aquifère (géologie des sédiments et géochimie de l’eau souterraine). De plus, la localisation de la coupe 2D représentée et utilisée pour la modélisation numérique a été choisie en fonction des voies naturelles de migration du panache. Cette approche multidisciplinaire itérative pour considérer tous les aspects d’un système aquifère est originale.

Approche de modélisation

Cet article présente l'évaluation des effets de l'hétérogénéité hydrogéologique et géochimique sur l'atténuation naturelle du lixiviat émis par un site d'enfouissement. Ces travaux sont originaux par leur l'utilisation complémentaire de la modélisation numérique et des données géochimiques; méthodes qui permettent d'évaluer les effets des conditions géochimiques variables sur deux voies distinctes d'atténuation naturelle du lixiviat. Le modèle numérique reproduit toutes les variables identifiées lors de la caractérisation et de l'intégration des données de terrain à l'échelle du sous-basin versant. Le modèle est ensuite ajusté aux données obtenues de l'âge de l'eau et des temps de résidence. Ceci permet d'augmenter la représentativité du modèle par rapport au système aquifère, donnant un certain niveau de confiance aux prédictions du modèle. Ensuite le modèle a été vérifié en fonction de bien représenter l'étendue du panache par rapport aux concentrations de chlorure observées dans l'aquifère. Les données géochimiques ont été utilisées pour contraindre le modèle numérique en vérifiant les voies de migration du panache simulées et en contraignant les propriétés hydrauliques afin de reproduire les observations aux niveaux des temps de résidence et de l'étendue et des concentrations du panache de chlorure. Ensuite, la modélisation du transport de masse pour le chlorure a été effectuée afin de prédire le devenir du panache de lixiviat. Cette approche permet d'assurer la représentativité du système aquifère et des conditions du panache pour la détermination de l'atténuation naturelle en tant que mode de gestion. Cette approche pour évaluer l'impact d'une source de contamination sur les milieux récepteurs est novatrice.

Définition des conditions d'atténuation naturelle des contaminants

Suite à la modélisation numérique, les données géochimiques de l'eau souterraine ont été analysées davantage par des méthodes graphiques pour permettre l'identification et la compréhension des mécanismes d'atténuation. Ceci a permis de reconnaître deux voix distinctes de migration et d'atténuation du lixiviat : 1) migration dans des eaux souterraines acides dans une portion d'aquifère homogène et 2) migration dans des eaux souterraines neutres dans une portion d'aquifère hétérogène. Cela a aussi permis de comprendre l'évolution naturelle de l'eau souterraine et les plages de concentrations anormales pour les conditions naturelles. Les résultats démontrent finalement que la méthodologie de caractérisation hydrogéologique intégrée qui a été développée dans le cadre de la présente recherche doctorale pourrait être utilisée pour évaluer efficacement l’atténuation naturelle d’autres sites contaminés dans des environnements géologiques complexes. Les résultats obtenus par l’approche utilisée dans cet article et dans l’ensemble de la thèse fournissent les informations nécessaires à l’identification des principaux mécanismes d’atténuation qui sont propre à un site spécifique, ce qui permet une gestion environnementale rationnelle.

ABSTRACT:

A leachate plume emitted by a decommissioned landfill in a shallow granular aquifer is being managed by natural attenuation. To evaluate the attenuation efficiency, an earlier detailed characterization was carried out within the 12 km^2 sub-watershed surrounding the landfill. This characterization revealed highly heterogeneous hydrogeological and geochemical conditions and allowed the development of a detailed conceptual model of the aquifer. On this basis, the present study aims to evaluate the effects of these types of heterogeneity on natural attenuation of the leachate plume. The effects of hydrogeological heterogeneities were investigated using a 2D (vertical section) numerical model passing through the landfill along natural groundwater flow and leachate migration paths. Particular analyses were made to verify if the numerical model is representative on the basis of observed hydraulic heads and geochemical data (groundwater age and geochemical tracers). The numerical model shows that the layering of silts and sands limits groundwater flow and leads to its relatively long residence time (40-60 years) in the deepest part of the aquifer where the plume migrates. Moreover, the leachate plume is shown to undergo strong mixing during its migration; which favors its attenuation prior to its discharge into streams. Graphical analysis of geochemical data allowed the identification of two distinct leachate natural attenuation paths related to differing geochemical conditions of the groundwater through which the plume migrates. The identified leachate attenuation mechanisms decrease concentrations of major ions and metals over short distances, so that groundwater is not exceeding quality standards outside the landfill property limits. The understanding of aquifer conditions thus allowed a better assessment of the leachate environmental risk to receptors, especially streams.

KEYWORDS: Hydrogeochemistry, granular aquifer, landfill leachate, natural attenuation processes, numerical modeling.

7.1 Introduction

Decommissioned and unlined municipal landfills solely managed by natural attenuation can leach contaminants to surface and groundwater for decades to centuries (Cozzarelli et al. 2011; Milosevic et al. 2009; Bjerg et al. 2011, 2003; Kjeldsen et al. 2002; Christensen et al. 2001, 1994). Monitored natural attenuation (MNA) refers to the reliance on natural attenuation processes, such as dilution, sorption, and microbial degradation, to reduce the mass of contaminants to less harmful forms or to acceptable concentrations within a reasonable time frame when compared to active remediation methods (Cozzarelli et al. 2011; USEPA 2008, 2007; Christensen et al. 2001). When properly assessed and employed, MNA may provide an effective, non intrusive and lower cost approach for the management of landfill contaminants emitted in groundwater (Bjerg et al. 2011; USEPA 2008, 2007). It is generally agreed that the effectiveness of natural attenuation processes is site-specific, both in terms of landfill setting and hydrogeological conditions, and thus MNA requires a local characterization, monitoring and revaluation through time (Cozzarelli et al. 2011; Bjerg et al. 2011; USEPA 2008, 2007; Kjeldsen et al. 2002; Christensen et al. 2001; EA 2003, 1999; EC 2000, 1999).

Over the past decade, scientific advances have been made regarding the occurrence and rates of several attenuation processes within leachate plumes or at their fringe (physical, chemical and microbial), which control the fate and transport of contaminants in aquifers. Table 7.1 summarizes natural attenuation reactions and processes acting upon the different types of chemical compounds found in leachate plumes. Despite knowledge of the general attenuation processes affecting leachate plumes, previous studies have demonstrated the complexity and variability of reactions occurring as plumes migrate through different types of geological conditions (Robertson et al. 2011; Cozzarelli et al. 2011, 2009; Gutierrez-Neri et al. 2009; Lorah et al. 2009; Bjerg et al. 2011, 2003; USEPA 2008, 2007; van Breukelen et al. 2003; Kjeldsen et al. 2002; Christensen et al. 2001, 1994). Some of these studies have also shown that many components can also be remobilized within the plume and migrate further along flowpaths (Cozzarelli et al. 2011; 2009; Lorah et al. 2009; Bjerg et al. 2011, 2003; van Breukelen et al. 2003; Kjeldsen et al. 2002). More importantly, these studies confirmed that ammonium is a persistent contaminant that does not decrease in concentration with time under anaerobic conditions, which allows ammonium to continue its migration within the plume at elevated concentrations (Cozzarelli et al. 2011; Lorah et al. 2009; USEPA 2008, 2007; Böhlke et al. 2006). Under such conditions, numerous decommissioned and unmonitored waste sites may pose a risk to the quality of surface and groundwater, where those sites are not properly monitored (Cozzarelli et al. 2011; Butt et al. 2008; EA 2003, 1999; EC 2000, 1999).

Tableau 7.1 : Summary of landfill leachate natural attenuation reactions (modified from Christensen et al. 2001)

Chemical compounds	Characteristics	Attenuation mechanisms and principal reactions
Dissolved organic matter	Expressed as total organic carbon, dissolved organic carbon, non-volatile organic carbon or chemical O_2 demand in leachate, since it is difficult to differentiate the types of organic compounds present in dissolved organic matter. Therefore it is difficult to be specific in relation to the attenuation mechanisms that take place for these contaminants. The portion of dissolved organic carbon in the leachate acid phase was reported to degrade easily (volatile fatty acid). However, the fraction of dissolved organic matter from the leachate methanogenic phase is persistent and does not degrade easily (humic and fulvic acid, refractory compounds). Field evidence suggests that dissolved organic matter decreases readily within the first few hundreds of meters from landfills.	Dilution, sorption (not significant and lacks field observations), degradation
Xenobiotic organic compounds	Most compounds seem to degrade in the plume. All compounds from this group degrade in a different manner and specific studies in relation to certain compounds are available in the literature.	Dilution, sorption, degradation
Inorganic macrocomponents	The degree of reaction mechanisms depends on the concentration of each component and pH. Anions are important because of their capacity to form complexes and to participate in precipitation and dissolution reactions. However, complexes formation can increase cations and heavy metals mobility. Cations attenuation is principally through ionic exchange.	Dilution, redox reactions, ionic exchange, precipitation
NH_4^+	Participates in complex redox reactions, the mechanisms of attenuation of this compound are not well understood. This compound is toxic and persistent in the environment, is mostly retarded and remains a long term contaminant.	Ionic exchange (sorption), oxidation, anammox reaction under anaerobic conditions
Fe, Mn, S	Actively involved in redox reactions. Will highly contribute to natural attenuation reactions depending on the ionic form available (speciation) and quantity. Fe and Mn are associated to sediments as oxides or hydroxides (Fe(III) & Mn(IV)). Fe reduces under the form Fe(II) and is an important electron acceptor for the leachate plume.	Redox reactions, dilution, abiotic dissolution of minerals in sediments, ionic exchange, precipitation, complex formation with organic carbon
Heavy metals	Heavy metals do not represent a problem for groundwater contamination related to landfills, since heavy metals actively participate in sorption and precipitation reactions very locally at the source of the leachate plume and are not present in great concentrations. However, their presence in leachate complicates the understanding of natural attenuation since diverse metals will react differently and will be distributed in the plume in relation to pH conditions, which can increase metal solubility or metal mobility.	Sorption, precipitation, dilution

Despite advances in the scientific understanding of mechanisms and rates of contaminant attenuation in leachate plumes, a debate still exists within the remediation community regarding the adequate assessment of MNA based on EPA guidance and on an acceptable level of uncertainty. Moreover, among numerous regulatory agencies throughout the world, there is no consensus on parameters that should be monitored and their associated maximum acceptable concentration in groundwater (Butt et al. 2008; Wilson et al. 2005; Vangelas et al. 2005; Norris et al. 2004; Daugherty et al. 2004; Rittmann et al. 2004; Crumbling et al. 2003; Lee et al. 2003; EA 2003, 1999; EC 2000, 1999). On the other hand, it is currently well accepted that much of the spatial variability in observed contaminant concentrations around contaminated sites results from geological heterogeneity and, consequently, a three-dimensional characterization of aquifer systems is required to understand the transport and ultimately the fate of contaminants (Bjerg et al. 2011; Sudicky and Illman 2011; Sudicky et al. 2010; Maxwell et al. 2008; USEPA 2008, 2007). In addition to the development of a detailed knowledge of the hydrogeological system needed to establish transport pathways, groundwater recharge and discharge characteristics must also be defined since they exert dominant controls on flow (USEPA 2008, 2007; Healy and Cook 2002; Kim 2002; Arnold and Allen 1999). Bjerg et al. (2011) advocated the need for new approaches, the development of in situ indicators to document mass removal and numerical modeling to test hypotheses in order to assess the potential efficiency of MNA for contaminated site management. Furthermore, Cozzarelli et al. (2011) stated that accurate description of biochemical processes affecting the transport of contaminant in aquifers affected by landfill leachate requires an understanding of the aquifer geological and hydrodynamic framework. Many authors have concluded that heterogeneity not only controls the physical spreading of contaminants by dispersion and advection but also influences chemical reactions and bacterial attachment rates as well as remediation effectiveness (Anderson and McCray 2011; Allen-King et al. 2010).

The present study aims to evaluate the effects of both hydrogeological and geochemical heterogeneity on the natural attenuation of a leachate plume. The study area is located in the province of Quebec, Canada, where monitoring is not required for unlined waste sites under two conditions; 1) if the site was definitively closed before the legislation for engineered landfills entered into effect in 2006 (MDDEP, 2012a); and 2) on the date of the site closure, the decommissioned landfill demonstrated conformity to the requirements of the previous legislation on solid waste. Nonetheless, this previous legislation on solid waste included no standard values for contaminants in groundwater, or for ammonium in surface water, and very few restrictions related to site hydrogeology (MDDEP, 2012b).

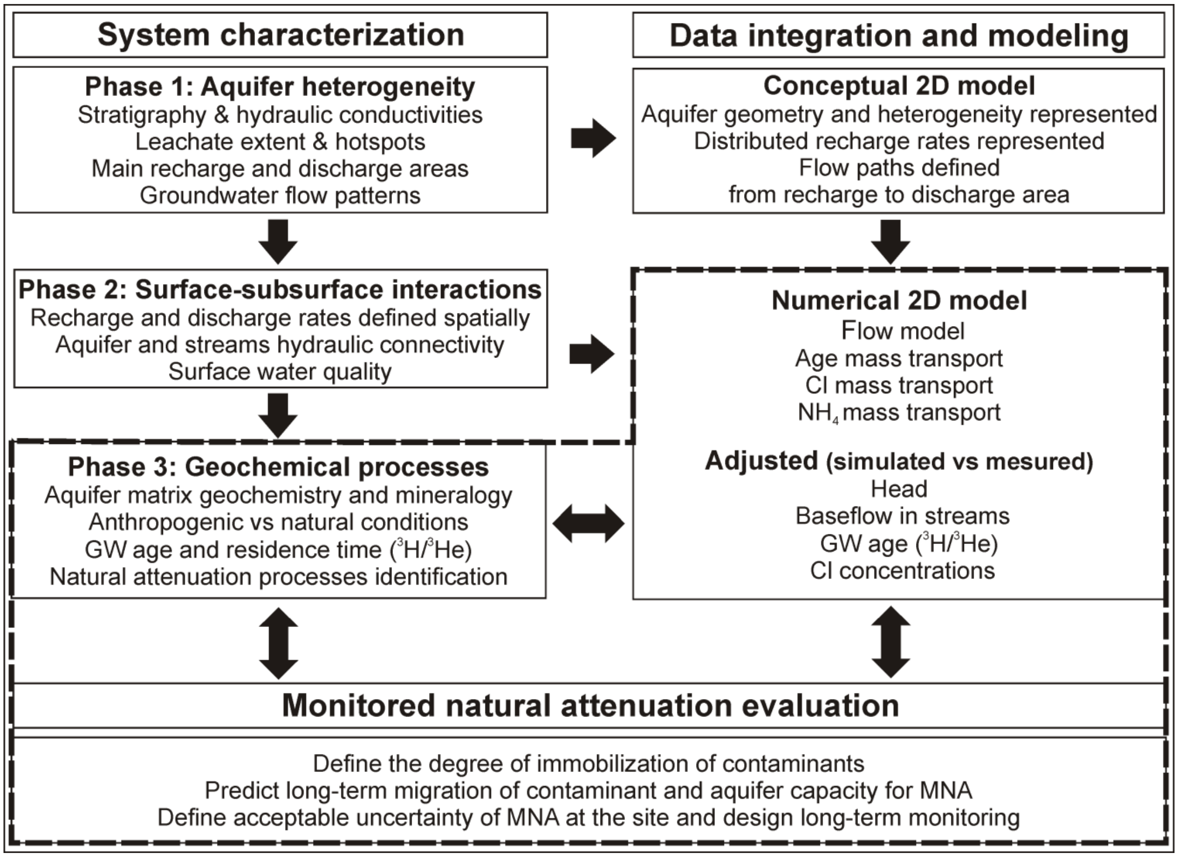

Figure 7.1 : General approach and scope of the present study

Flowchart illustrating the general approach used to assess the efficiency of monitored natural attenuation (MNA) as a leachate plume management approach in the study area. The objectives and interrelations of the current (within bold dashed line) and preceding steps of the general study are shown. Characterization was previously carried out in three phases. The present study involves the development of a flow and transport numerical model and further interpretation of geochemical processes

The hydrogeological and geochemical heterogeneities of the study area were previously defined by a detailed characterization (Figure 7.1). The present study thus builds upon a three-phase characterization intended to provide detailed knowledge and understanding of the study area in the perspective of assessing the effectiveness of MNA as a leachate plume management approach. The first phase focused on the development and application of an integrated characterization approach allowing the definition of hydrogeological (physical) heterogeneity at the sub-watershed scale (Tremblay et al. 2013). This phase also allowed the general delineation of the leachate plume and the development of an initial conceptual model of the aquifer system in the study area. The second phase involved the use of surficial and shallow direct and indirect indicators to delineate zones and rates of groundwater recharge and discharge and thus refine the conceptual model (Tremblay 2013; chapter 5). The third phase allowed the definition of the heterogeneous natural geochemical conditions of groundwater in the aquifer system and the identification of two distinct leachate attenuation paths, depending on the geochemical conditions through which the leachate plume migrates (Tremblay 2013; chapter 6).

The present study thus integrates and further analyzes the available data, within the framework of the detailed conceptual model of the aquifer system, to assess the effects of hydrogeological and geochemical heterogeneities on the natural attenuation of the leachate plume. The effects of hydrogeological heterogeneities are investigated using a 2D (vertical section) numerical model passing through the landfill along groundwater flow and leachate migration paths. Particular efforts are made to verify if the numerical is representative on the basis of observed hydraulic heads and geochemical data (groundwater age and geochemical tracers). The numerical model is then used to assess flow and leachate migration paths, leachate residence time and the importance of dispersion on concentrations of conservative compounds in the leachate. Geochemical indicators are then graphically interpreted to identify the geochemical processes affecting leachate attenuation under the heterogeneous geochemical conditions of the aquifer. This paper thus aims to make a contribution to the approaches used to study the fate of contaminants: integrated characterization, combination of numerical modeling and geochemical data analysis, numerical model history-matching approaches, and distinction of varied natural geochemical conditions and their effects on natural attenuation.

7.2 Study area and previous work

7.2.1 Study area location and context

The 12 km^2 study area is located 35 km south of Quebec City, Canada (Figure 7.2). A decommissioned landfill and an engineered landfill are located on a 1 km^2 property managed by the “Régie intermunicipale de gestion des déchets des Chutes-de-la-Chaudière” (RIGDCC), a public organization. The engineered landfill is in operation and meets current waste management standards. The decommissioned landfill was, and still is, unlined at the base when it was in operation between 1974 and 1997, during which 9 million tonnes of waste from varied sources were disposed directly over permeable sands. From 1993 to 1997, the waste piles were reshaped and an impermeable cover was placed on its surface (RIGDCC 2013). Prior to closure, landfill leachate was detected in surrounding creeks (Gauthier 1993a). Since its closure, as allowed by current regulations, the decommissioned landfill is solely managed by natural attenuation, although no studies have verified the effectiveness of this management method.

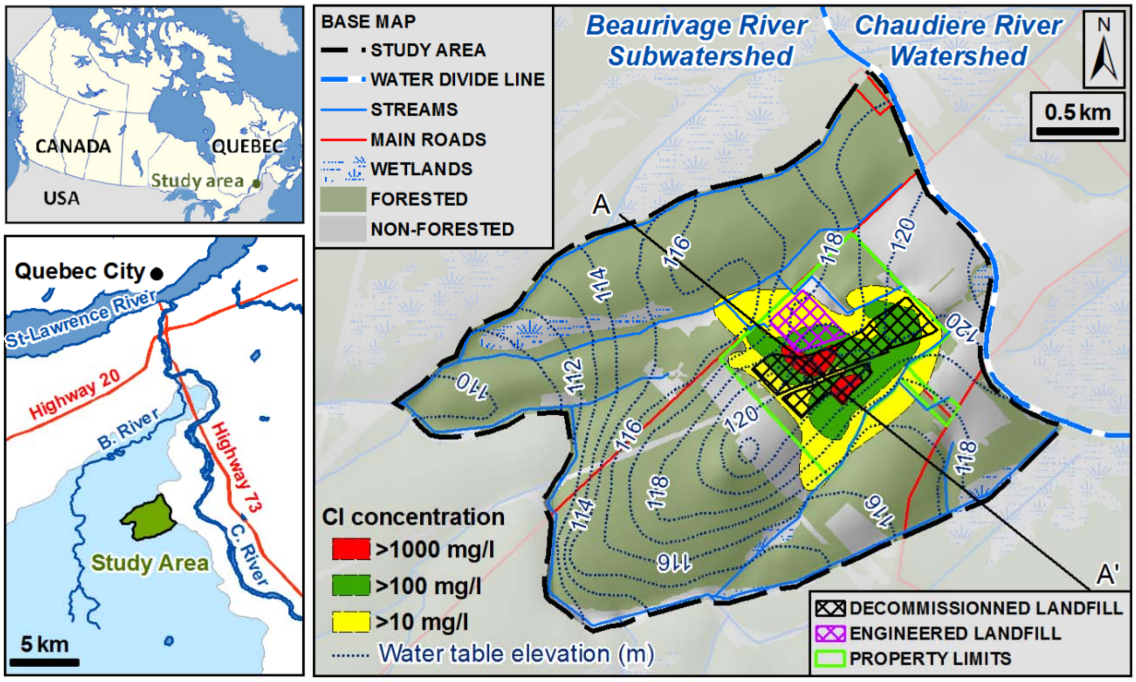

Figure 7.2 : Study area location, physiography and land use

Map on bottom left: B. River and C. River respectively stand for Beaurivage River and Chaudiere River; the Beaurivage River sub-watershed is shaded in blue. The map on the right shows the water table elevation, the leachate plume extent as indicated by chloride concentrations and the location of the cross-section (Figure 7.3) used as a basis for the 2D (vertical section) numerical model (Figure 7.5) (modified from Tremblay et al. 2013)

7.2.2 Study area physical and hydrogeological setting

The study area is easily accessible by primary, secondary and forestry roads (not all indicated on Figure 7.2). The 12 km^2 sub-watershed study area was defined on its eastern limit by a groundwater divide between the Beaurivage River sub-watershed and the Chaudière River watershed. Elsewhere, limits were based on locations of streams surrounding the landfill site (Figure 7.2). The study area is mostly forested but also includes peatlands, agricultural fields and operational gravel pits. The natural topography of the study area is relatively flat and varies from 125 m (asl) at the groundwater divide line to 109 m asl at the study area surface water outlet (Tremblay et al. 2013). The landfills, reaching 130 m in elevation, changed the local topography. The water table, which follows topography, is quite shallow, and generally found between 0.1 to 2 m below ground surface. Within the sub-watershed, groundwater flows from higher elevations found under the landfill towards local streams in the southern and northern direction (Figure 7.2). The surface soil is mostly made of poorly drained podzols of marine origin, sparse zones of organic material and well decomposed black peat (Laplante 1962). As a consequence of the flat topography and soil type, an important forestry and agricultural drainage network is in place and, in combination with natural streams, local hydrology strongly influences groundwater flow (Tremblay 2013; chapter 5).

The granular aquifer is composed of 10 to 20 m of highly heterogeneous sands interbedded with silt and clayey silt layers or lenses. The aquifer overlies discontinuous layers of glaciomarine silt and till forming the relatively impermeable base of the aquifer. The till deposited by the last glaciation overlies the regional sedimentary rock aquifer composed of fractured shale and sandstone. The Quaternary fluvial and deltaic sediments making up the aquifer were deposited after the last glaciation (Bolduc 2003). The unconsolidated material originated from sea level changes during the regression of the Champlain Sea at the former Chaudière River outlet. At that time, the system was controlled by sea tides and currents, followed by fluvial erosion and organic sedimentation. This littoral depositional environment is responsible for the high heterogeneity of the aquifer material (Parent and Occhietti 1988), as well as naturally occurring anoxic and oxidizing zones that influence the natural geochemical composition of groundwater (Tremblay et al. 2008). Added to this complex environment, is the presence of a leachate plume emitted by the decommissioned landfill which undergoes important geochemical transformations along its path (Tremblay et al. 2008), which is the object of the present study.

A regional hydrogeological characterization was carried out in the Chaudière River watershed, including the chemical analysis of 127 groundwater samples within the sedimentary rock aquifers (Benoit et al. 2012). The geochemical data supported the results obtained from a numerical flow model that was used to define regional groundwater flow (Brun Koné 2012; Brun Koné et al. 2011). The water type found in the center of the Breakeyville formation underlying the study area is Na-HCO_3, and it contains no tritium and has an average chloride concentration below 1 mg/l. The presence of this water type seems influenced by the effect of regional flow controlled by structures of orientation SW-NO within the formation and the presence of a thicker aquitard covering the bedrock in this area. Based on phase 1 of the characterization work (Figure 7.1; Tremblay et al. 2013, 2012) and on local hydrogeological investigations carried out by consultants on the landfill property (Gauthier et al. 1993b), the sedimentary rock aquifer is found at an average elevation of 100 m asl within the study area, but it is also known to outcrop or to reach up to 110 m asl at some locations. The sedimentary rock aquifer underneath the landfill property has hydraulic conductivities in the order of 10^{-7} m/s for fissile black shale and 10^{-5} m/ s for sandstone (Gauthier et al. 1993). Surface geophysical surveys demonstrated the local thinning of the impermeable units separating the sedimentary rock aquifer from the granular aquifer (Tremblay et al. 2013). This has implications on the hydraulic connectivity between the granular and the rock aquifers (Tremblay et al, 2012b), which influences groundwater geochemistry in the study area as will be discussed later.

7.2.3 Previous integrated characterization of the granular aquifer

In 2006, the present study was initiated to investigate the effectiveness of the natural attenuation of the leachate plume emitted by the decommissioned landfill. Prior field work and historical hydrogeological data on the granular aquifer (1985-1998) were sparse and did not extend outside the landfill site property limit of 1 km^2. In 2007, an integrated multidisciplinary aquifer characterization was carried out. The extent of the study area was defined so as to encompass leachate source zones, near the decommissioned landfill, and receptors, which are mainly represented by streams in the study area. Tremblay et al. (2013) describe the characterization program designed on the basis of the specific hydrogeological setting of the study area, which includes three phases (Figure 7.1). The first phase involved : 1) surface geophysics including a) ground penetrating radar (GPR) and b) electrical tomography (ERT); 2) direct-push methods including a) cone penetration tests (CPT) with soil moisture resistivity (SMR), b) soil sampling and c) installation of fully-screened observation wells in the granular aquifer; 3) borehole operations including a) flowmeter measurements while pumping (Paradis et al. 2011), b) high vertical resolution (15 cm) packer slug tests, c) single well interference slug tests to assess vertical hydraulic conductivity (Paradis and Lefebvre 2013) and d) multilevel measurements of geochemical parameters and groundwater sampling. The integration of multi-source data allowed the representation of aquifer heterogeneity and the delineation of the leachate plumes (extent shown on Figure 7.2).

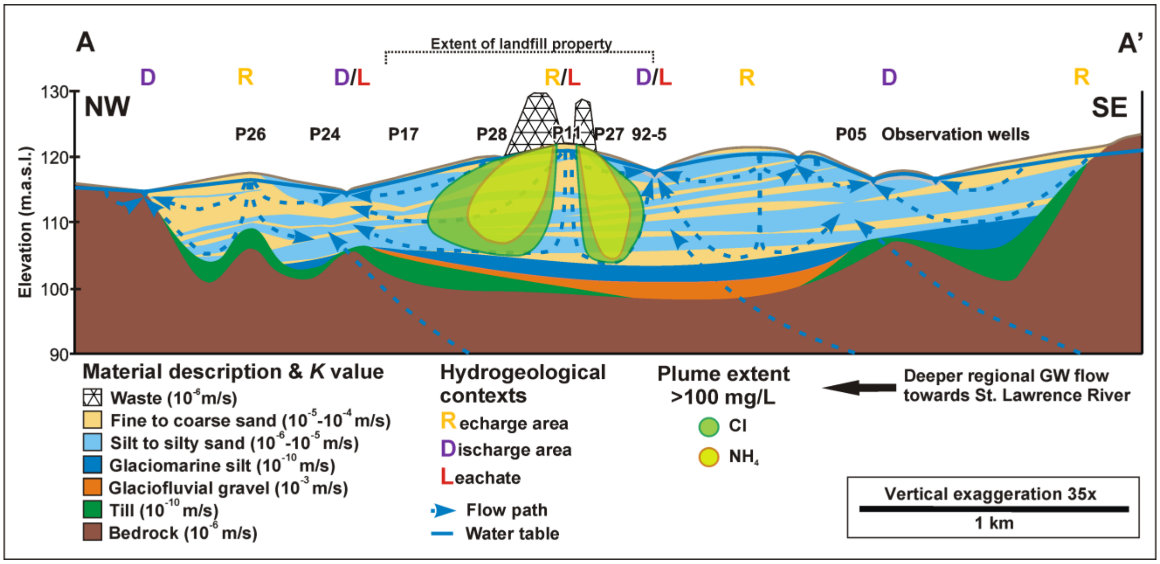

Figure 7.3 : Detailed hydrogeological conceptual model

Figure 7.2 shows the location of section A-A'. This section is the basis of the 2D (vertical section) numerical model (Figure 7.5) and indicates the actual conditions for the chloride and the ammonium plumes. Numbers indicate observation well locations. The extent of ammonium and chloride concentrations exceeding 100 mg/l illustrates that the leachate plume is readily attenuated and remains within the landfill property limits (modified from Tremblay et al. 2013)

Figure 7.3 shows a detailed conceptual model of the aquifer system in the study area (based on Tremblay et al. 2013). The cross-section representing the conceptual model passes through the decommissioned landfill along groundwater flow and leachate migration paths (location shown in Figure 7.2). The hydrostratigraphy of the granular aquifer shown on the conceptual model was inferred from the Fellenius and Eslami (2000) CPT profiling chart validated with soil samples. CPT/SMR soundings provide bulk electrical conductivity measurements that were correlated to water conductivity (Tremblay et al. 2013), thus confirming the extent of the leachate plume and providing further indications on the main flow and leachate migration paths (Figures 7.2 and 7.3). Additionally, the combination of GPR, ERT and CPT surveys indicated the discontinuity of the impermeable base of the aquifer (till and glaciomarine silt), which has implication for potential hydraulic connectivity between the superficial granular aquifer and the fractured bedrock aquifer as well as for the dispersion of the leachate plume in the study area.

The integration of phase 1 characterization data (Figure 7.1) provided an initial detailed conceptual model of the aquifer system (Figure 7.3; Tremblay et al. 2013). The initial conceptual model illustrated the high level of heterogeneity of the aquifer material and the great variability of the aquifer and aquitard surface topography, as well as the non-continuity of some units. The hot spots of higher concentrations of components originating from landfill leachate were also located (Tremblay et al. 2013). The initial conceptual model provided indications of recharge and discharge zones on the basis of multiple parameters (groundwater temperature, conductivity, pH and dissolved oxygen; the relative topography and vertical hydraulic gradients; fluid conductivity and TDS derived from cone penetration tests (CPT) with soil moisture resistivity (SMR)).

Phase 2 of the field characterization further investigated recharge and discharge zones and the possible location of leachate discharge into streams (Figure 7.1). Spatially distributed recharge rates were inferred from the integration of multiple shallow indicators: $^{3}H/^{3}He$ groundwater ages, general water chemistry, $^{18}O/^{2}H$ on groundwater, well hydrographs, flowmeter measurements in streams at baseflow and an extensive study on soils and surface water connectivity to the aquifer (Tremblay 2013; chapter 5). Results from phase 2 show that the study area received average total yearly precipitation of 1200 mm from 1978 to 2012. A maximum recharge rate of 420 mm/year occurs at the groundwater divide line at the eastern limit of the study area, where aquifer sediments are coarser and homogeneous, and in a few sand mounds distributed over the area. Elsewhere, surface soil and heterogeneous sediments control recharge rates estimated to vary from 250 mm/yr to zero at discharge areas (Tremblay 2013; chapter 5). Important hypodermic

Tableau 7.2 : Description of groundwater HCA groups

Distinctive features of groundwater groups defined through hierarchical cluster analysis (HCA) in the study area (modified from Tremblay et al. 2013). Symbols for subgroups are used in the geochemical graphs of Figures 7.8, 7.9, 7.10, 7.11 and 7.12

GW HCA Group and Subgroup			**Distinctive Features**
Alkaline	●	a	Na-HCO_3-SO_4. Two samples are from the fractured bedrock and four samples are from the granular aquifer GW contains diagnostic minor elements F, P,B and low concentration of Si
	■	b	Ca-Na-HCO_3 Groundwater from the bedrock aquifer emerging into the granular aquifer carrying its signature and mixing with evolved fresh water GW contains diagnostic minor elements F, P,B and high concentration of Si
Neutral	■	c	Ca-HCO_3-SO_4 (presence of Fe variable)
	◆	d	Ca-HCO_3 (Fe) anoxic superficial confining silt unit (most samples are from 1 to 5 m depth)
	●	e	Ca-HCO_3 samples found >7m depth
Acidic	■	f	Wells located at the GW divide line. Ca-HCO_3 type but contain anthropogenic NO_3 and Cl from agricultural activities
	◆	g	Superficial wells underneath anoxic soil unit contain S and Fe
	●	h	Recharge water in SiO_2 medium sands, located on local higher topographic elevation. Contains higher concentrations of metals such as Al.
Leachate	●		Wells located underneath the waste pile or within 20 m. Elevated concentrations in all major ions. NH_4 and Cl are characteristic
Attenuated Leachate	◆	i	Leachate mixing with neutral semi-anoxic water in a heterogeneous geological setting “anoxic path” samples are located >100 m from the former waste pile
	●	j	Leachate mixing with oxic and acidic water in a homogenous geological setting “oxic path” samples are located within 100 m from the former waste pile

flow and runoff zones were also identified throughout the study area. Streams were found to be directly connected to the aquifer in the northern part of the study area where stream beds are sandy, whereas they are indirectly connected in the southern and western zones where stream beds are composed of 1 to 3 m of compact silt or till. The aquifer is under semi-confined conditions in certain locations due to the presence of a silt layer (1-3 m) at the surface in the lower topographic areas surrounding the sand mounds. The top silt layer and organic soil at the surface of the aquifer induce anoxic conditions at the top of the water table.

Phase 3 of field characterization (Figure 7.1) investigated geochemical heterogeneity of aquifer sediments and groundwater. Multivariate methods (Davis 2000) were used to interpret 84 groundwater samples, including two samples from the sedimentary rock aquifer (Tremblay 2013; paper 3). Table 7.2 shows the Hierarchical Cluster Analysis (HCA) water types including five main groundwater groups, which are subdivided in subgroups. Table 7.2 indicates the symbols used on following graphs in this paper to distinguish water groups (by color) and subgroups (by symbol shapes). Table 7.2 also lists the major ions associated with water groups. The geochemical conditions and spatial distributions of HCA water groups were used to recognize natural groundwater from landfill leachate or leachate-impacted groundwater. Natural groundwater groups were named according to their distinct pH ranges (Alkaline, Neutral, Acidic). Table 7.3 describes the general chemical composition of natural groundwater and leachate-related water. Chemical data thus show quite heterogeneous natural geochemical conditions, besides leachate or leachate-impacted waters. Furthermore, HCA groups provide indications of the diverse origins and geochemical evolution of natural groundwater (Tremblay 2013; chapter 6; Table 7.2): acidic waters associated with preferential recharge areas, alkaline waters originating from upflow from the underlying sedimentary rock aquifer, and neutral waters representing mixtures or evolution of diverse groundwater origins. Leachate and leachate-impacted groundwater could also be distinguished from HCA groups, which showed two distinct leachate evolution paths associated with leachate migration into different natural groundwater related to redox conditions labeled as “oxic path” and “anoxic path”. Figure 7.4 illustrates the spatial distribution of natural geochemical conditions over the study area. The great variability in geochemical conditions is controlled by the nature of surface conditions and aquifer sediments.

Tableau 7.3 : Typical geochemical composition of groundwater and representative tracers for natural groundwater evolution or leachate plume detection in the study area. Grey shading indicates diagnostic parameters for each group

		Granular aquifer		Bedrock aquifer		1.Leachate attenuation path		2. Leachate attenuation path		Leachate	
		Background values		Background values		Acidic & oxic		Neutral & anoxic		Source	
	DL	MIN	MAX	MIN	MAX	MIN	MAX	MIN	MAX	MIN	MAX
Ca	0.02	1.2	38	2.1	5.8	37	220	11	290	8.7	1900
Mg	0.01	0.32	10	0.97	1.7	5.5	16	4.5	52	16	300
Na	0.03	0.62	9.7	29	97	2.1	14	2.7	80	14	1400
K	0.1	0.35	8.9	6.5	7.3	2.8	22	0.7	11	22	1600
Fe	0.03	-	43	-	0.051	8.2	120	-	100	5.1	1400
Mn	0.0004	0.0011	9.4	0.011	0.018	1.1	10	0.14	3	0.03	57
$N\text{-}NH_4$	0.02	-	1.2	0.77	1.2	0.16	31	-	1.8	31	1500
HCO_3	1	1	170	116	256	264	516	82.8	1620	432	12161
Cl^-	0.05	0.25	16	0.37	0.44	0.35	7.2	6	160	7.2	1800
Br^-	0.1	-	-	-	-	-	0.1	-	0.6	-	2
SO_4^{2+}	0.1	-	28	3.4	24	2.1	150	-	49	-	1500
$N\text{-}NO_3^-$	0.01	-	-	-	-	-	0.02	-	0.13	-	3.5
$N\text{-}NO_2^-$		-	-	-	-	-	0	-	-	-	0.84
Al	0.001	-	1.2	0.0052	0.078	0.012	0.049	-	0.01	0.01	0.31
As	0.001	-	0.023	-	-	0.013	0.042	-	0.053	0.0028	0.12
Ba	0.002	0.0045	0.39	0.087	0.16	0.081	0.33	0.034	0.52	0.14	4.6
B	0.005	-	0.028	0.13	0.24	0.018	0.77	0.005	0.062	0.18	4.7
Cr	0.0005	-	0.009	-	-	-	0.0025	-	-	0.0025	0.11
Co	0.0005	-	0.058	-	-	0.00073	0.039	-	-	0.001	0.45
Cu	0.0005	-	0.028	-	0.001	-	0.0031	-	0.0019	-	0.0021
F	0.1	-	0.1	0.2	0.6	-	0.1	-	0.1	-	0.4
Li	0.001	-	-	-	-	-	0	-	0.017	-	0.024
Mo	0.0005	-	0.004	0.0008	0.0009	-	0.0012	-	0.0006	-	0.012
Ni	0.001	-	0.14	0.002	0.003	0.0048	0.034	-	0.0068	0.0048	0.5
Pb	0.0001	-	0.002	-	-	-	0.0006	-	0.0001	-	0.0007
Si	0.02	1.7	15	4.3	4.7	3	9.2	6.3	15	0.1	25
Sr	0.002	0.02	0.24	0.03	0.17	0.17	1.1	0.095	3.1	0.33	9.8
S^{2-}	0.02	-	0.7	-	0.2	-	0	-	2.1	-	0.1
Ti	0.001	-	0.015	-	-	-	0.018	-	0.003	-	0.41
V	0.002	-	0.025	-	-	-	0.003	-	-	-	0.100
Zn	0.001	-	0.091	-	0.007	0.0076	0.047	-	0.32	-	0.09
P	0.01	-	0.1	0.22	0.32	-	0.1	-	0.04	-	2.10
COD	10	-	77	-	-	45	81	-	680	81	40000
BOD_5	4	-	10	-	-	-	12	-	420	4	28000
C-DIC	0.1	1	30	22	47	33	173	24	263	115	2395
C-DOC	0.1	0.3	7.1	0.1	5.4	9	19.6	2	241	20	12240
CS	mS/cm^c	0.02	0.93	0.19	0.41	0.48	1.93	0.21	2.21	1.93	19.63
pH	-	4.31	7.92	7.96	8.73	5.98	6.56	6.46	7.86	5.41	7.37
Pe	-	0.36	9.97	2.20	2.40	2.5	5.1	-0.7	2.8	1.3	4.7

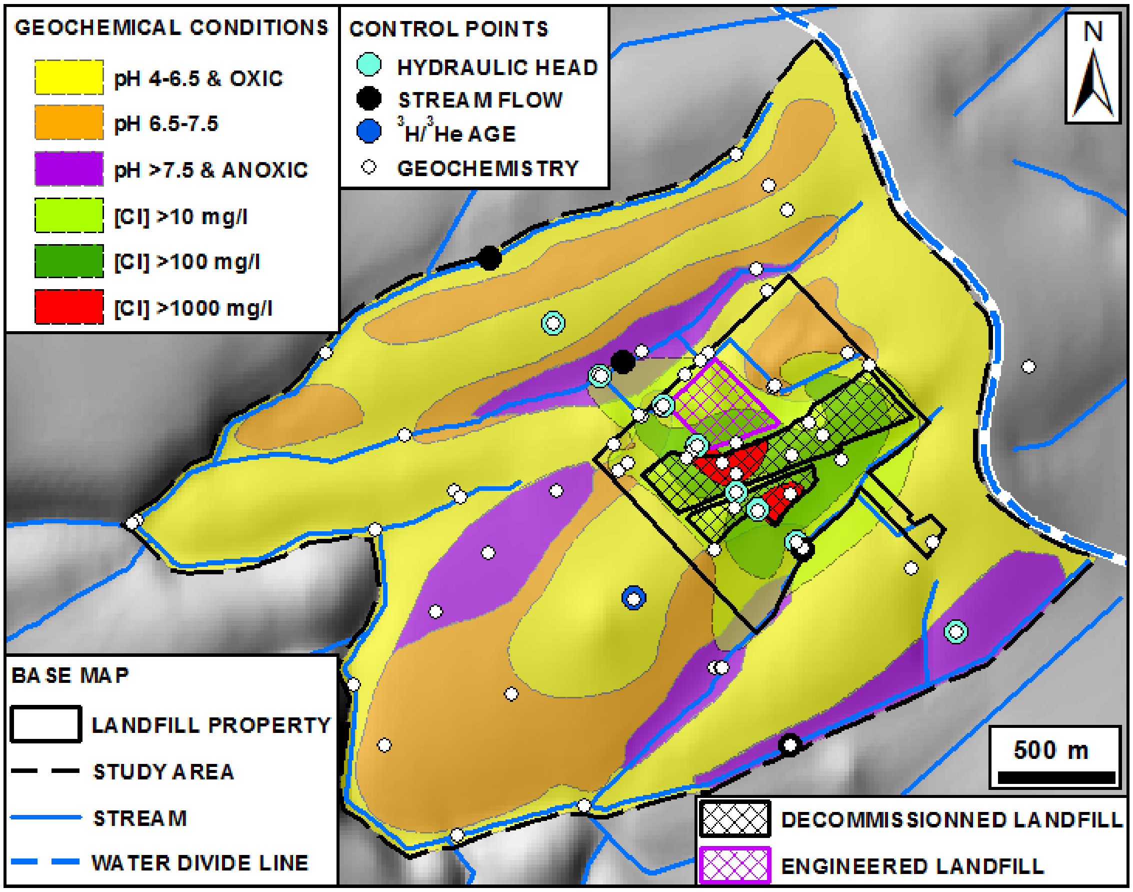

Figure 7.4 : Spatial distribution of geochemical conditions in the study area
Groundwater geochemical conditions in relation to the leachate plume that is migrating through distinct geochemical environments. Small circles indicate the control points used to define geochemical conditions. Large circles indicate control points used for ajusting the numerical model (Figure 7.6). The geochemical polygons in color on this map were drawed on the basis of all data and knowledge acquired through the integrated characterization.

The aquifer has a good capacity to attenuate the leachate plume inorganic and organic components. In Figure 7.3, the extent of ammonium and chloride concentrations exceeding 100 mg/l illustrates that the leachate plume is readily attenuated and remains within the property limits. The previous investigations demonstrated the absence of concentrations above provincial maximum concentration limits outside the landfill property. Plume extent in terms of chloride concentration is illustrated in Figure 7.2 and Figure 7.4. The decommissioned landfill is located on a local water divide line and leachate migrates on either side of the waste pile as shown with water table elevation contours on Figure 7.2. Moreover, the map shows that groundwater hydraulic gradients are stronger on both sides of the former landfill. Since the landfill is located on a groundwater divide, there is also a strong vertical flow component under the landfill. $^3H/^3He$ groundwater dating shows that it takes about 40 years for the groundwater to reach the base of the 15 m thick granular aquifer at the highest water table elevation located on the cross section on the landfill property (Figure 7.3). Since the landfill started its operation in 1974, the leachate emitted has not yet had time to spread outside the landfill property limits and concentrations of inorganic elements found farther than 100 m from the waste piles are relatively low. Organic elements are not detected behong this limits. The effects of hydrogeological and geochemical heterogeneities on the leachate plume migration and attenuation are further investigated in the following two sections dealing respectively with numerical modeling and the interpretation of geochemical data.

7.3 Effect of hydrogeological heterogeneity

7.3.1 Numerical model development

The objective of numerical modeling is to assess the impacts of hydrogeological heterogeneities on leachate natural attenuation. In that perspective, groundwater flow and mass transport simulations were thus planned to obtain the following results:

- Steady-state flow simulations were completed to ajust the model with hydraulic heads and stream baseflow. These simulations then provided groundwater flow paths and discharge points;
- Steady-state groundwater age mass transport simulation (Goode 1996) were carried out in the steady-state flow field to history-match the model with a vertical isotopic ($^3He/^3H$) age profile. These simulations then provided the spatial distribution of the residence time of groundwater;
- Transient mass transport of a conservative tracer was simulated in the steady-state flow field for a time period corresponding to the interval between the start of operation of the landfill and now (35 years). These results are then used to history-match the model with measured chloride concentrations. The simulations allowed the assessment of the migration paths of the leachate plume and the effect of dispersion on concentration

reductions between leachate sources at the landfill and the discharge of the leachate plume in streams, which are the main receptors in the study area.

The numerical site model was developed using FLONET/TR2 (Molson and Frind 2002). Groundwater flow was simulated in FLONET; a 2D steady-state flow finite element simulator. Contaminant mass transport was simulated with TR2. FLONET/TR2 represents a saturated domain and can generate a structured rectangular mesh that allows grid elements to deform, thus providing a more accurate representation of the water table geometry (Molson and Frind 2002). Moreover the simulator allows the simulations of mean groundwater age through dispersive age mass transport (Molson and Frind 2012, 2002).

7.3.1.1 Numerical grid

The numerical grid for all simulation was developed to represent the detailed conceptual model and the geometry of the aquifers and aquitard units (Figure 7.5) based on the conceptual model cross-section (Figure 7.3). The vertical section is 4 km long and has fine layers over the about 20 m thickness of the granular aquifer. The uppermost elevation of the model is about 130 m. The sedimentary rock aquifer starts at 0 m asl and outcrops on each side of the section above 115 m asl. The bedrock aquifer is composed of 30 layers, the aquitard units have 10 layers and the granular aquifer system has 50 layers. The horizontal section is divided in 290 vertical units for a total of 58 580 elements. On both ends of the vertical section, where the bedrock outcrops, the 50 layers composing the granular aquifer are allowed to deform to represent the water table geometry.

7.3.1.2 Initial and boundary conditions

The initial conditions for the numerical flow model were selected to represent the conceptual model developed from the integration of field data (Figure 7.5). Spatially distributed recharge values were based on Tremblay (2013; chapter 5). A fluid flux was applied on the right boundary to represent groundwater flow within the bedrock aquifer. Fixed heads were imposed at stream locations at known elevations, and no-flow boundaries were imposed elsewhere. Hydraulic conductivities were based on measured values and hydrofacies defined previously (Tremblay et al. 2013). The effective porosities used were set to 0.14 for the silt, 0.30 for the sands and 0.20 for the sedimentary rock aquifer. The unsaturated zone was not considered, since the water table is very shallow.

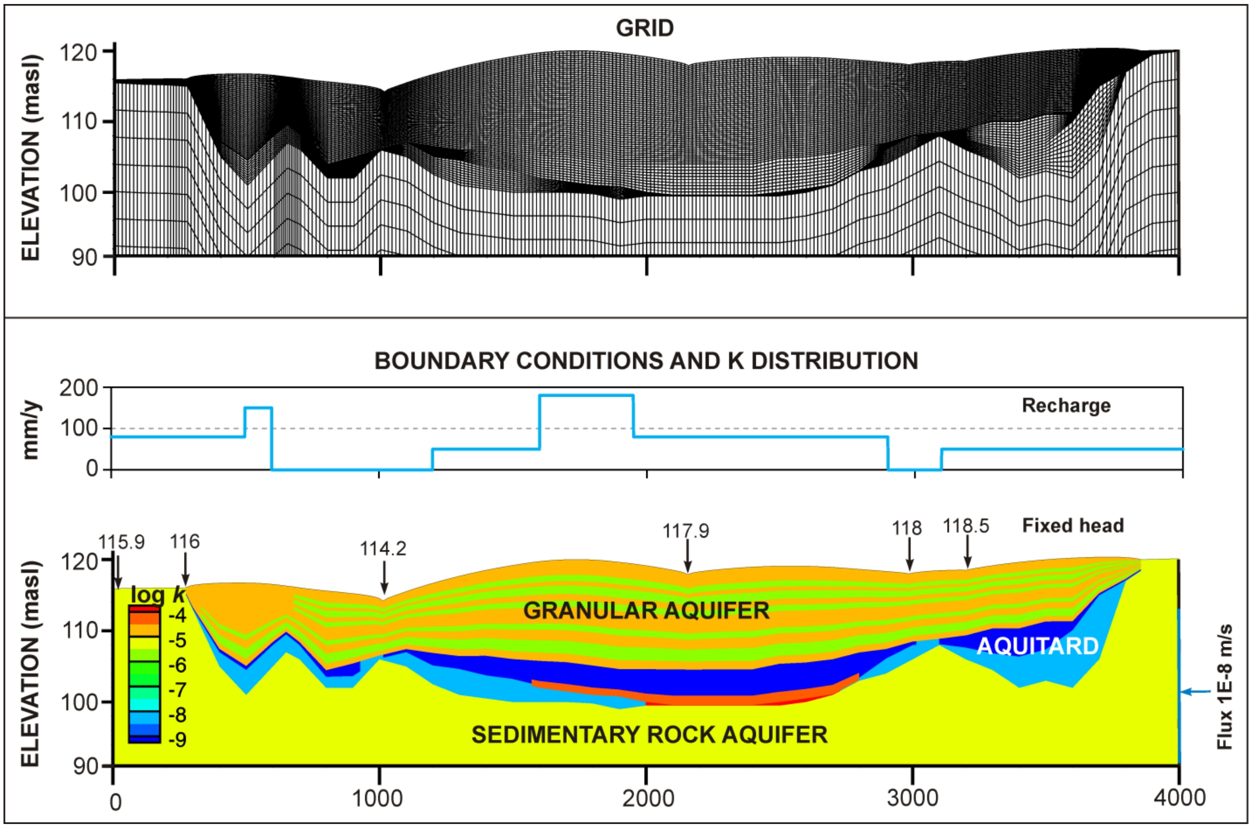

Figure 7.5 : Groundwater flow numerical model grid and initial conditions

The top graph shows an enlargement of the numerical grid, which has fine layers in the granular aquifer in order to represent the alternating layers of sands and silt. The numerical grid actually extends to depth 0 m into the regional sedimentary rock aquifer underlying the granular aquifer. Groundwater recharge applied to the top of the model is based on a previous assessment of the spatial distribution of recharge (Tremblay 2013; Chapter 5). The lower graph shows the spatial distribution of hydraulic properties based on the types of materials present in the aquifer system

7.3.2 Groundwater flow and mass transport - History matching/calibration

Parameters of the groundwater flow model were adjusted based on the comparison of simulated to measured hydraulic heads and stream baseflows. Figure 7.6 (a and b) shows that hydraulic heads were matched well with the model, while simulated stream flow rates are in the same range as measured values. Groundwater age mass transport and conservative tracer mass transport results were used to adjust horizontal and vertical hydraulic conductivity values so that simulated ages and conservative tracer concentrations would respectively match measured $^{3}H/^{3}He$ groundwater ages and chloride concentrations found in the aquifer. In an iterative process, hydraulic properties were also selected to ensure that conservative (chloride) mass transport would also match observed chloride concentrations. Longitudinal dispersivity was set to 10 m, transverse dispersivity to 0.01 m and a diffusion coefficient similar to a conservative tracer of $8.64 \cdot 10^{-9}$ m^2/s was used. The grid and the time steps were checked to satisfy respectively the Peclet and the Courant criterion (Molson and Frind 2002). Figure 7.6 (c and d) illustrates the match for the mass transport model. Initial age conditions were set to 0 y at the top of the model in recharge area illustrated on Figure 7.5 and to 3000 y for the bedrock aquifer on

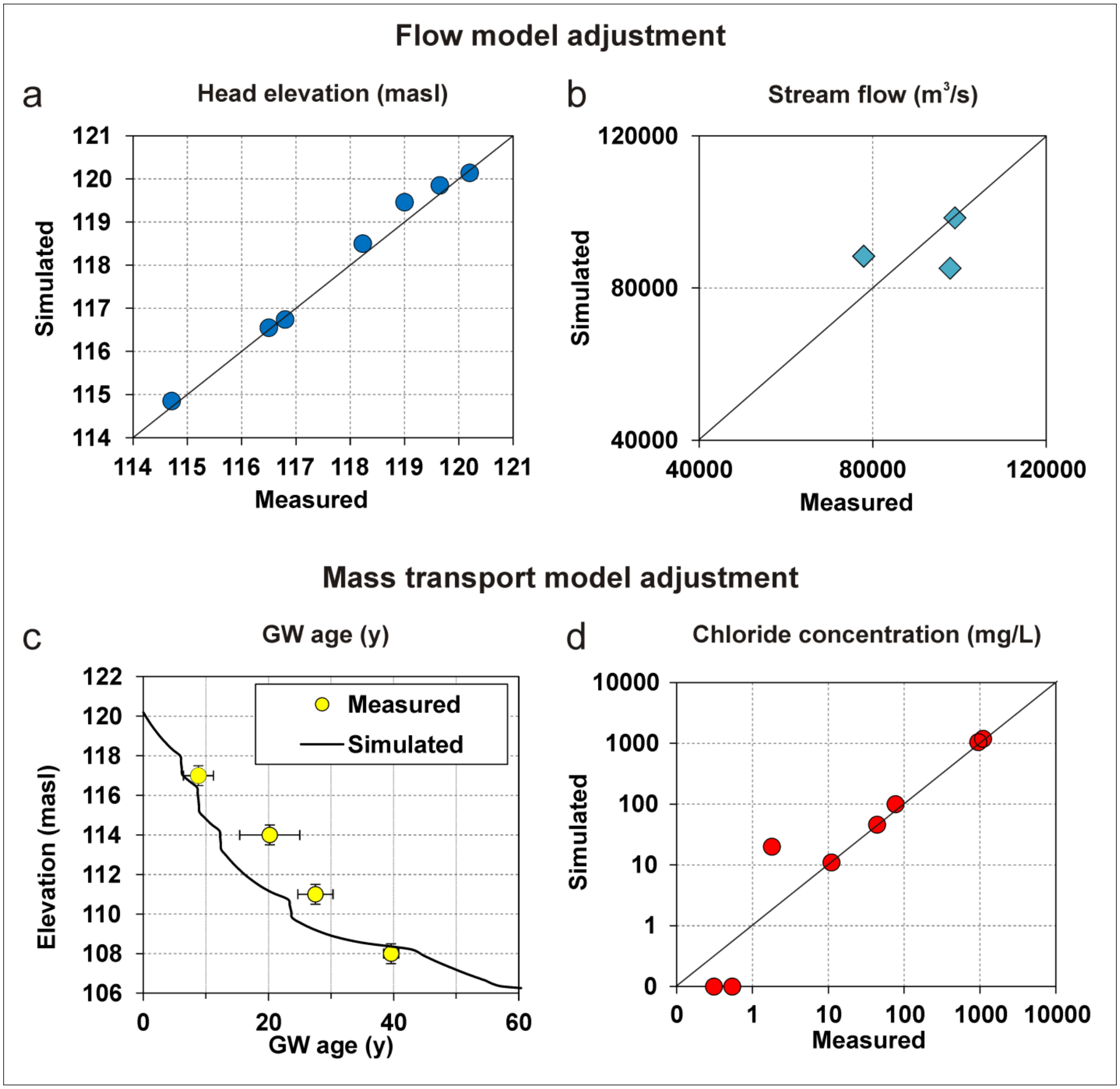

Figure 7.6 : Numerical model adjustment

the right vertical boundary of the model. Mean groundwater age was simulated for a very long period so that it represents steady state conditions (a million days with a time step of 100 days). A measured vertical profile of $^3H/^3He$ groundwater ages at a location with a sediment sequence equivalent to that underlying the landfill was used to compare model results. $^3H/^3He$ groundwater ages could not be measured under the landfill because landfills have anomalously high concentrations of tritium. After flow and mean age simulations were matched, mass transport of a conservative tracer equivalent to chloride was simulated under the same conditions, adding a source of 1000 mg/L of chloride at the landfill location. The model was first run under the same conditions under transient state for 20 years followed by another 20 years but under a capped landfill condition with recharge set to zero under the landfill location. An impermeable barrier with very low K_v and K_h values was defined on the first two layers to simulate the landfill under capped conditions. When simulated chloride concentrations were matching observations and other history-matching parameters (hydraulic heads, groundwater age, stream flow) were also reproduced satisfactorily, simulations to predict the fate of the leachate plume were done. The model was not sensitive to small changes of initial conditions.

7.3.3 Prediction of plume migration for risk management

Groundwater flow and mass transport simulations were run under the conditions described in previous sections. Figure 7.7a illustrates simulated groundwater streamfunctions (flowlines). FLONET/TR2 calculates steady-state stream functions (flowlines) that are convertible into stream fluxes by summing the fluxes from each stream tube that discharges at a stream location and by multiplying results by the length of the stream (Molson and Frind 2002). Since aquifer conditions are difficult to represent at the model limits, results considered for the mass transport model are limited to the window outlined in Figure 7.7a. Flowlines indicate that an important downward flow component is present under the location of the landfill, which is due to its location at a groundwater divide line (local high of the water table). Groundwater originating from the landfill thus flows both north and south (left and right on the model), which implies that leachate emitted by the landfill will also move north and south (confirmed by the observed leachate plume spatial distribution; Figures 7.2 and 7.3). Flowlines also indicate local exchanges between the sedimentary rock aquifer and the granular aquifer (for example at the horizontal distance of about 1000 m along the geological cross-section at a stream location). Otherwise, groundwater recharging the granular aquifer remains within that aquifer and discharge into streams.

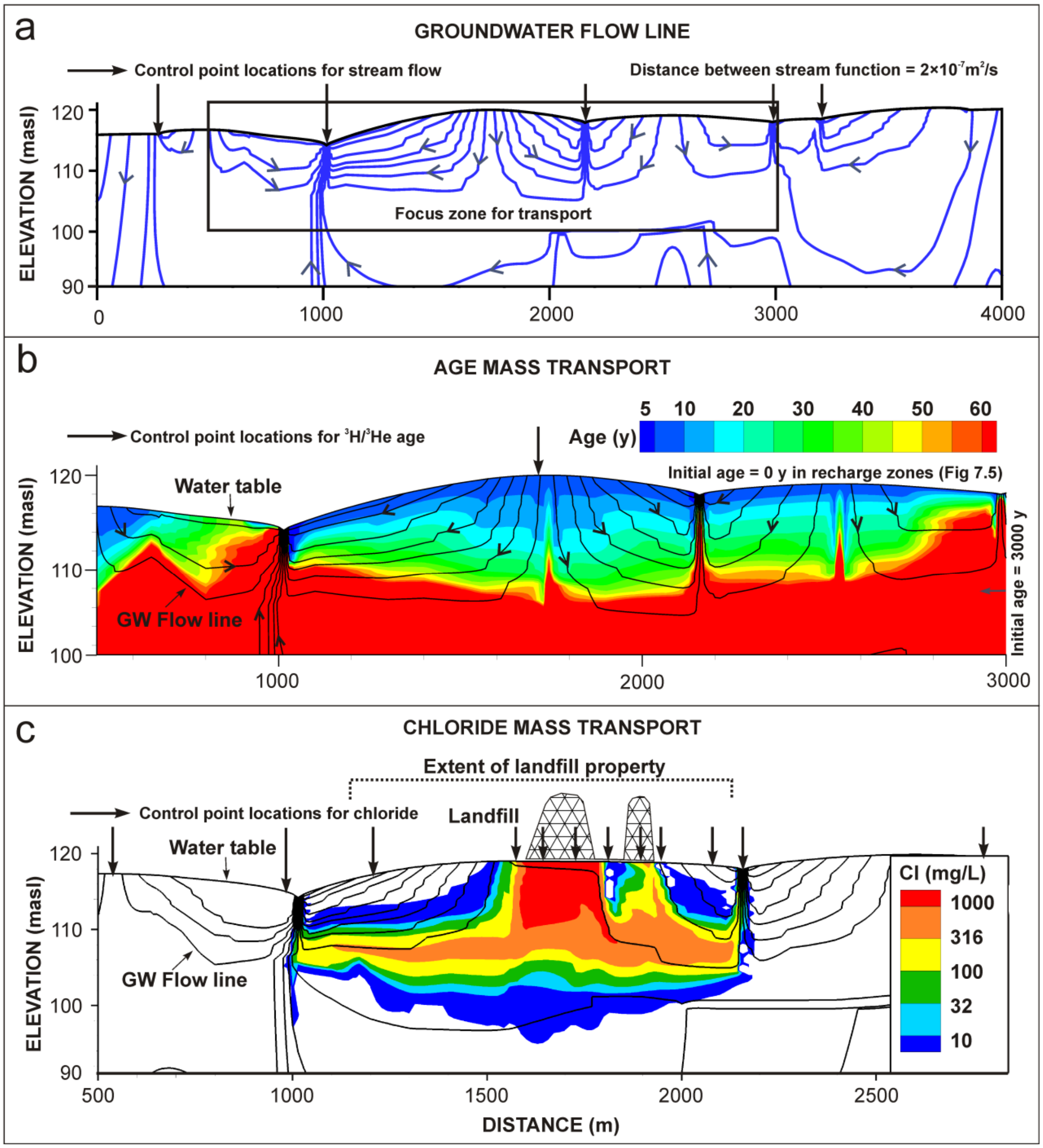

Figure 7.7 : Numerical model results

a) Groundwater flow lines under steady-state flow conditions. b) Steady-state mean groundwater age mass transport under steady-state flow. c) Chloride concentrations after 35 years from transient mass transport under steady-state flow conditions. The numerical simulation actually extends to depth 0 m into the regional sedimentary rock aquifer underlying the granular aquifer.

The model was also used for transient conservative tracer (chloride) mass transport from present time to 35 years in the future to predict the evolution of chloride concentrations (Figure 7.7c). Chloride mass transport simulations show that chloride preferentially migrates downward at first and that recharge on both sides of the capped landfill contributes to the important dilution of the chloride plume, which is dispersed and mixed with recharging groundwater. Chloride preferentially migrates near the base of the aquifer and its concentration remains below the provincial standard at the landfill property limits. Therefore, based on model predictions, it can be inferred that dispersive processes will significantly contribute to the reduction of concentrations of components emitted into groundwater by the landfill. Compared to a conservative tracer such as chloride, reductions in concentrations will even be more important for components that are either retarded or degraded during their transport. However, since aquifer conditions are partly anaerobic along the cross-section modeled, there are some concerns about the fate of the ammonium plume, which could be remobilized and migrate at high concentrations. Still, ammonium will undergo the same effect of dispersive reduction in concentration as chloride, which will limit its concentration. Moreover, the landfill source will eventually decrease in concentration whereas the model did not account for this effect. The numerical model shows that the interbedding of fine layers with coarser sediments in the aquifer limits groundwater flow and causes relatively long groundwater residence time in the deepest portion of the granular aquifer (40-60 years). Moreover, the leachate plume is strongly diluted by dispersion during its migration; which favours its attenuation before emerging in the surrounding streams.

7.4 Effect of geochemical heterogeneity on attenuation processes

7.4.1 Leachate attenuation paths

Investigation of site-specific geochemical conditions is required to gain an understanding of the processes that control natural attenuation at a landfill. Jankowski and Beck (2000) demonstrated that hydraulic conductivity and groundwater chemistry vary in relation to changes in depositional environments and also that chemical reactions, resulting from water-sediment interaction, produce significant changes in groundwater chemistry, which is a function of mineralogy, lithology and residence time in the aquifer. Moreover, McMahon and Chapelle (2008) stated that geological factors such as sediment origin and depositional environment can influence the availability of natural electron acceptors and donors in the subsurface. Therefore, to initially assess the impact of sediment heterogeneity on groundwater geochemical signatures and evolution in the study area, carbonate dissolution in groundwater, pH-pe conditions, attenuation of major ions and saturation indices (SI) were further investigated through graphical methods.

Carbonate minerals are ubiquitous in sediments and the degree of carbonate weathering is largely controlled by the availability of CO_2, while limited by the saturation of calcite in groundwater (Clark 2012). Tremblay et al. (2013) investigated this issue in two distinct recharge zones in the study area, finding that the dissolution of carbonate minerals is highly related to the material in place and its heterogeneity. Figure 7.8 shows the evolution of Ca^{2+} concentration as a function of pH for the HCA groundwater groups found in the study area (HCA subgroups symbols in Table 7.2). The relation between Ca^{2+} concentration and pH is controlled by calcite dissolution (Clark 2012), and it depends on whether groundwater dissolves calcite under an open or a closed system. Figure 7.8 shows that HCA groups related to natural groundwater have different ranges of Ca-pH conditions. The graph also differentiates leachate-impacted groundwater from natural conditions. Calcium is known to have generally high concentrations and moves with the plume front in leachate emitted by landfills (Christensen 2001). Therefore, the graph also confirms that the HCA groups correctly distinguish anthropogenic and natural conditions in the aquifer. Also, the amount of calcite dissolved is controlled by the partial pressure of CO_2 of the system (pCO_2). In recharge areas, as groundwater below the water table consumes carbonic acid through weathering, the pCO_2 drops and carbonate alkalinity increases (Clark 2012). Poles for groundwater origin are well defined on Figure 7.8. The figure shows that calcium concentration and pH remain similar in recharge groundwater (yellow and blue symbols below a pH of 6). Leachate groundwater (red symbols) plots in the neutral pH range but shows high concentrations of calcium. The two inferred leachate migration paths are recognizable on this graph: one towards lower pH (oxidizing path) and the other towards higher pH (anoxic path), while the calcium concentration decreases for both paths. Finally, the purple symbols are representative of groundwater from the sedimentary rock aquifer that emerges in the granular aquifer and mixes with neutral natural water (geochemical inferences of upflow were confirmed by vertical hydraulic gradients in upflow areas).

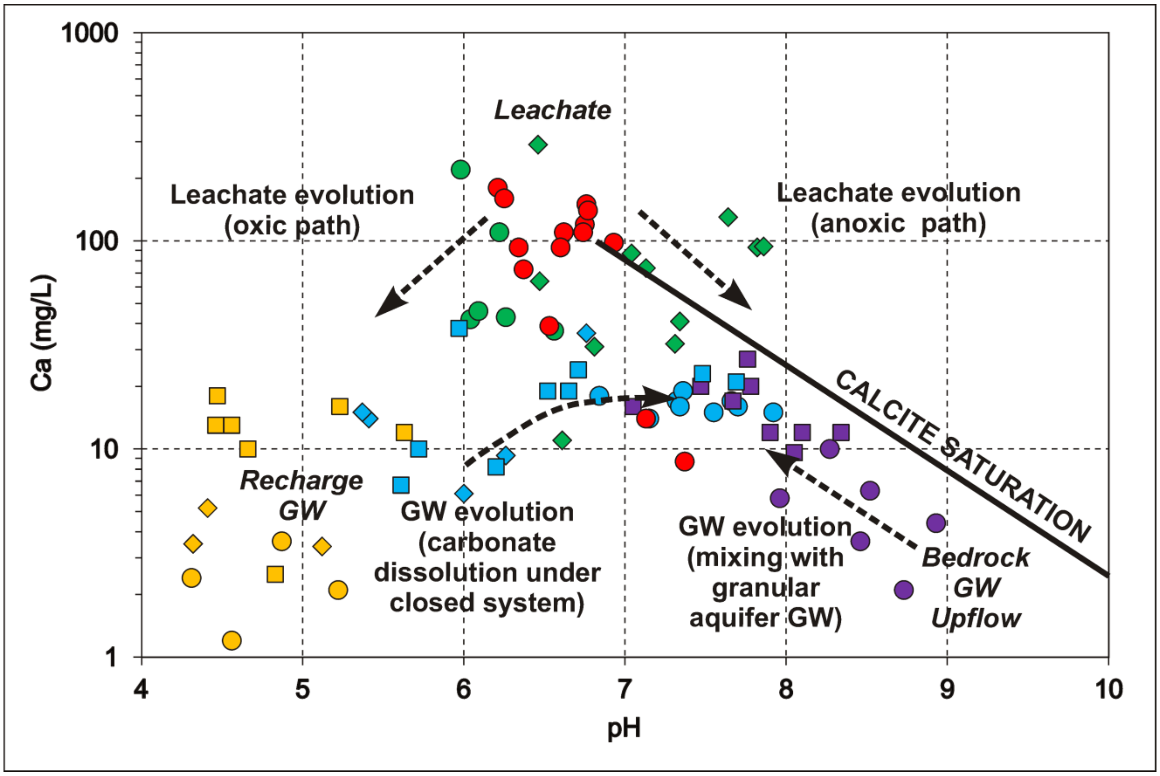

Figure 7.8 : Calcium concentrations versus pH

Carbonate cycle within the system under study: carbonate dissolution through natural groundwater evolution in relation to calcium ions (adapted from Clark 2012). Symbols used to distinguish groundwater groups are explained in Table 7.2

To better define the conditions of the two leachate attenuation paths, Figure 7.9 illustrates pH-pe conditions for leachate (red symbols) and leachate-impacted (green symbols) groundwater in the aquifer system (HCA subgroups symbols in Table 7.2). Figure 7.9 shows that pH-pe conditions identify two distinct leachate attenuation paths with different evolution of redox conditions. The occurrence of these attenuation paths depends on the hydrogeochemical conditions of groundwater in which leachate migrates: 1) “oxic path”: a leachate-impacted groundwater subgroup originating from the mixing of leachate with oxic and acidic groundwater in a relatively homogeneous geological setting within 100 m from the former waste pile; 2) “anoxic path”: a leachate-impacted groundwater subgroup related to leachate mixing with neutral semi-anoxic water in a heterogeneous geological setting farther than 100 m from the former waste pile. As a result, the attenuation reactions that will take place will be related to the type of natural groundwater found along a particular flowpath. Therefore the leachate-impacted groundwater down gradient from the landfill will also be related to the natural groundwater conditions that are highly variable at the subwatershed scale (Figure 7.4).

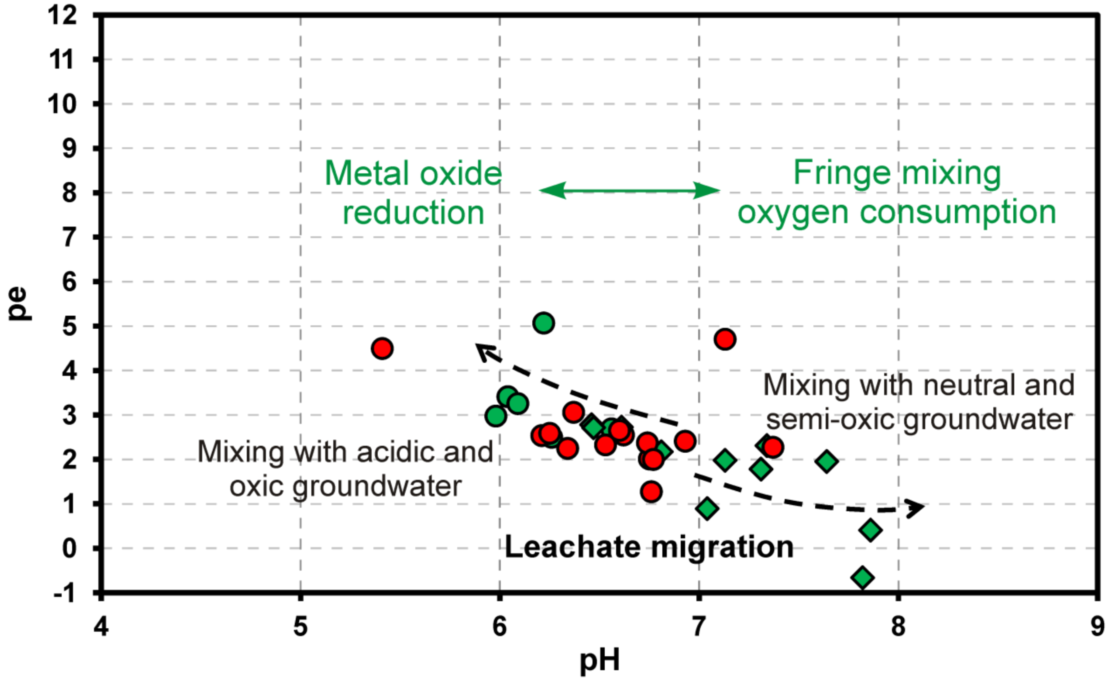

Figure 7.9 : Leachate and leachate-impacted groundwater pH-pe diagram
pH-pe conditions control the evolution of groundwater and the speciation of numerous elements as well as the leachate plume attenuation which occurs along two distinct paths under different geochemical conditions. Table 7.2 provides the legend of symbols

7.4.2 Attenuation processes and their effects

The groundwater inorganic geochemistry also provides indications about redox-sensitive mineral species. At the study site, it was found that redox conditions control some of the attenuation reactions but also natural groundwater geochemistry (Tremblay 2013; chapter 6). The series of graphs in Figure 7.10 show the relations between the concentrations of different major ions for the HCA water groups found in the study area (Table 7.2 provides a legend of the HCA group symbols).

Figure 7.10a illustrates the relations between sodium and chloride concentrations. This graph allows the definition of background concentrations for natural groundwater conditions since chloride is a conservative tracer present in high concentration in leachate (limit of natural

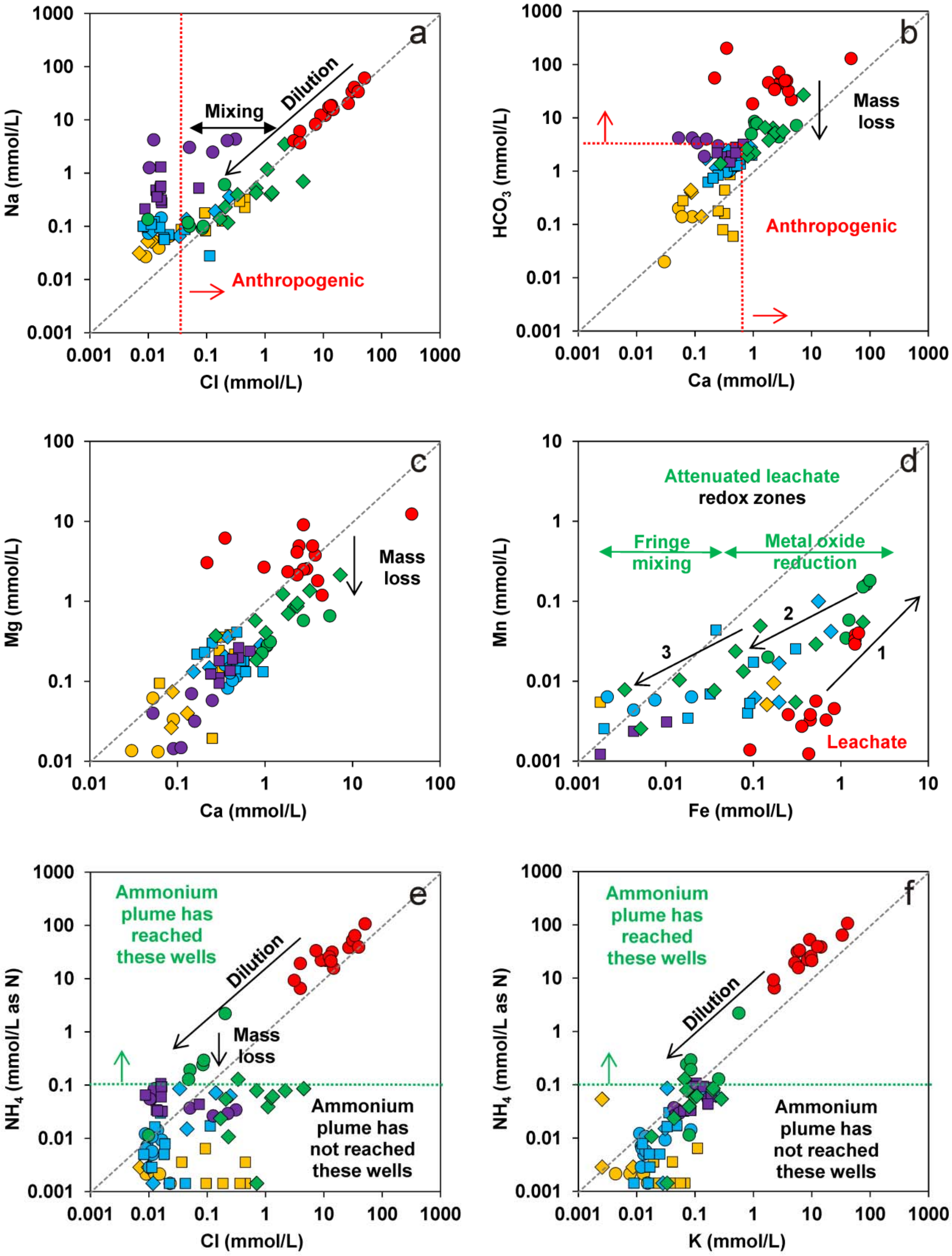

Figure 7.10 : Graphs showing the attenuation of major ions

Natural versus leachate plume conditions illustrated with groundwater chemistry for major ions. Graphs also show some of the attenuation processes along different flowpaths. Legend for the groundwater groups is shown in Table 7.2

conditions indicated by dotted red line on the graph). Although Tremblay (2013; chapter 6) showed that natural groundwater has chloride concentrations below 1 mg/l, a few samples outside the extent of the leachate plume were affected by agricultural activities and showed values around 10 mg/l. Other processes such as dilution and mixing between the natural groundwater groups and leachate (red symbols) are clearly identifiable on the graph. Within the alkaline group (purple symbols), for instance, sodium concentrations remain high because this groundwater type originating from the underlying sedimentary rock aquifer has naturally high sodium concentrations. However, Figure 7.10a shows that chloride concentrations are higher than background natural values due to mixing with leachate-impacted groundwater at the fringe of the plume. This occurs where the northern extent of the plume reaches alkaline groundwater (purple zone on Figure 7.4).

Bicarbonate (alkalinity) and carbon dioxide pressure are usually high in leachate due to the composition of waste and dissolution processes present in landfills (Figure 7.8; Christensen 1994). Alkalinity and calcium are thus both good tracers for the leachate plume when natural background concentrations are known, as calcium migrates with the front of the plume. Figure 7.10b illustrates the relationships between alkalinity and calcium (HCO_3 vs Ca). This graph identifies leachate-impacted groundwater from groundwater characterized by natural conditions (limit indicated by the red dotted line). The figure also shows that alkalinity decreases at a faster rate than calcium as the plume migrates (red and green symbols). Figure 7.10c shows the relations between Ca and magnesium (Mg) concentrations. In the literature, Mg is said to follow a similar trend as Ca since there are not many reactions involving Mg. However, in this study Mg is rather seen to be removed from the system slightly faster than Ca, as indicated by the lower ratio of Mg/Ca in leachate-impacted groundwater compared to leachate (Figure 7.10c).

The manganese (Mn) versus iron (Fe) graph (Figure 7.10d) illustrates the effects of redox zones related to the attenuation of leachate (green symbols are leachate-impacted groundwater and red circles represent leachate close to the source). Green circles are leachate-impacted groundwater sampled within 100 m of the landfill in a homogeneous geological environment where groundwater is acidic and oxic. On this graph, arrow 1 shows that when leachate migrates and mixes with this type of groundwater, additional metals originating from the aquifer sediments are released. Fe and Mn are examples of metals that dissolve under such conditions, which leads to increases in their concentration. The green diamond symbols represent groundwater sampled farther than 100 m from the landfill. Along the flow path represented by these samples, leachate mixes with neutral and semi-anoxic groundwater. The pH is then too high to release more metals in solution, so arrows 2 and 3 in Figure 7.10d indicate that concentrations of Mn and Fe decrease to background values along these paths.

Figures 7.10e and 7.10f, respectively, show the relations between ammonium (NH_4) versus Cl and the relation between NH_4 versus potassium (K). Cl and K can be used as tracers to help indicate plume degradation. Ammonium was found to be the dominant nitrogen species in the landfill leachate plume with concentration over 1000 mg/L below the landfill. However, its lateral extent was found to be relatively smaller than that of the Cl ion that is conservative. In the literature, it was demonstrated that ammonium behaves similarly to the potassium ion as they both have high affinities for cation exchange sites in sediments (Cozzarelli et al. 2011; van Breukelen 2003; Christensen et al. 2001). Moreover, it was demonstrated that ammonium can be retarded from 2.5 to 6 times in relation to the leachate plume chloride extent (Cozzarelli et al. 2010; Lorah et al. 2009; Böhlke et al. 2006; Heaton et al. 2005). In this study, only the groundwater samples represented by green round symbols plotted above natural concentration values for the study zone. Once again, these symbols are representative of leachate groundwater that mixes with oxic and acidic natural groundwater that are located within 100 m from the landfill. However, for samples located farther than 100 m from the waste pile, ammonium concentrations were similar to natural background values. From this interpretation, the extent for the ammonium plume can be inferred. This is important, since the NH_4 versus Cl graph can be used to infer plume scale nitrogen transport in relation to the extent of the Cl plume. It can be concluded from Figure 7.10e and 7.10f that no concentration of ammonium above natural concentration are found farther than 100 m from the waste pile and that the relatively constant ratios between NH_4 and Cl as well as NH_4 and K indicate that NH_4 concentrations are reduced due to leachate dilution, with some mass loss in leachate-impacted groundwater that has lower ratios.

Stable isotope analysis of ^{15}N can also be useful to identify the attenuation mechanisms of ammonium (Cozzarelli et al. 2010; Lorah et al. 2009; Böhlke et al. 2006; Heaton et al. 2005). Figure 7.11 provides complementary information on the fate of NH_4 using the isotope ^{15}N versus

NH_4 concentrations. Cozzarelli et al. (2011) demonstrated that, for the landfill located in Norman, Oklahoma, samples characterized by increases in $\delta^{15}N$ and decreases in NH_4 concentrations are representative of NH_4 oxidation reactions (nitrification) while samples with constant or decreasing $\delta^{15}N$ and decreasing NH_4 concentration are representative of ion exchange or mixing with background NH_4 concentrations. Moreover, Cozzarelli et al. (2011) demonstrated that $\delta^{15}N$ values greater than 7‰ occurs only in shallow groundwater, where the upper boundary of the leachate plume is near the water table. In this study, the figure shows a gap in ammonium concentration along flowpaths, as no samples with ammonium concentrations between 10 and 50 mg/l were collected. The lack of detailed data makes it difficult to identify or quantify

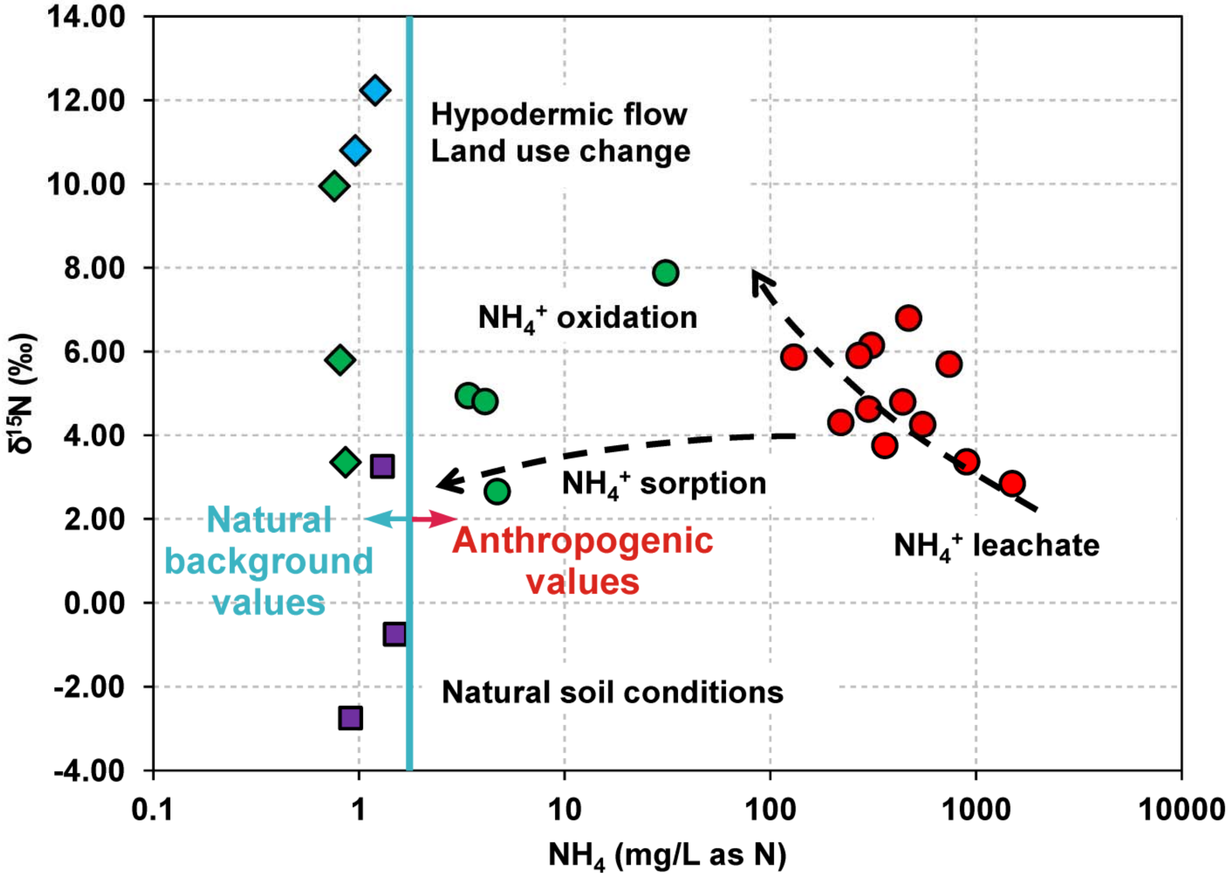

Figure 7.11 : ^{15}N isotopes providing evidence of NH_4 attenuation processes

There is a gap in ammonium concentrations along flowpaths, as no samples for concentrations between 50 to less than 10 mg/l were obtained. The lack of continuous data limits the identification or quantification of ammonium attenuation processes. Groundwater group symbols are shown in Table 7.2

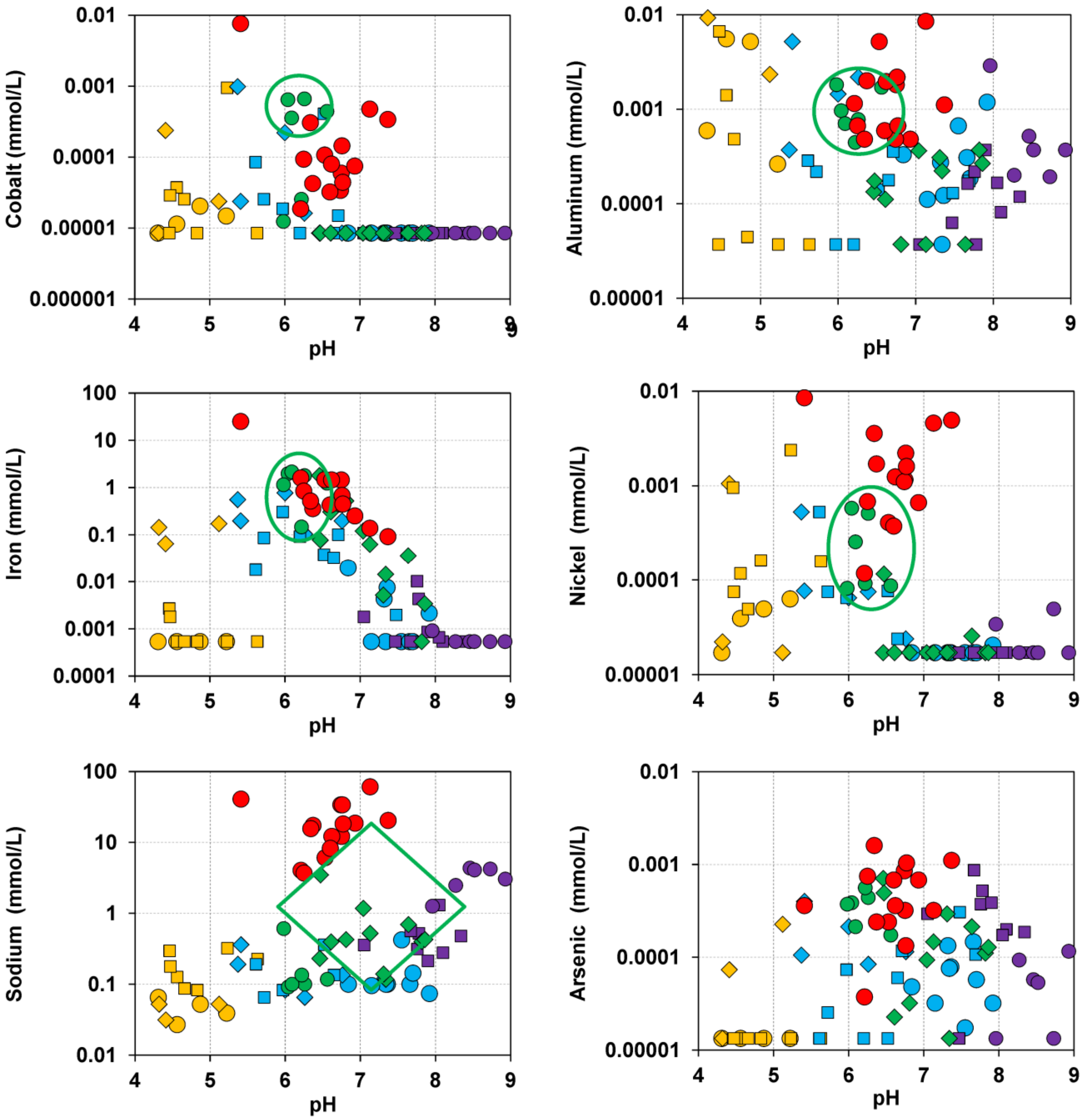

Figure 7.12 : Graphs showing attenuation of metals vs pH

As leachate (red symbols) migrates through distinct environments, different attenuation mechanisms take place. Leachate mixing with acidic groundwater in coarser sands (yellow symbols) puts in solution more metals than background values (green circles). Leachate mixing with neutral groundwater (blue symbols) in heterogeneous sediments has higher concentrations of sodium (green diamond). Arsenic is not only found in leachate but has elevated natural concentrations where groundwater from the underlying sedimentary aquifer emerges (purple symbols). Groundwater groups symbols in Table 7.2

ammonium processes. There is no presence of nitrate in wells where leachate is attenuated, although the concentration of ammonium decreases. Anammox reactions documented in the literature show a strong enrichment in δ15N for ammonium where reactions by anammox take place (Robertson et al. 2012), but this is not observed in the study area. Ammonium attenuation is difficult to explain with the available data since NH_4 concentrations decrease while nitrate or nitrite concentrations remain undetected along leachate migration paths.

Figure 7.4 illustrates the various geochemical contexts within which the leachate plume migrates as well as the present-day plume extent, as indicated by chloride concentrations. As the plume migrates through these distinct geochemical and geological environments, different elements are found in groundwater. Figure 7.12 illustrates the difference in groundwater composition between the flowpaths in relation to metal concentrations, as a function of pH. As the leachate plume migrates through distinct geological environments, different attenuation mechanisms take place. The figure illustrates that, as leachate mixes with acidic groundwater flowing in coarser sands, the presence of leachate leads to the release of more metals than background values (green circles on the graphs). In contrast, when the leachate mixes with neutral groundwater located in heterogeneous aquifer environments (green diamond), metal concentrations are significantly reduced, whereas it leads to higher concentrations of sodium. The figure also shows that arsenic is not only found in leachate but has natural elevated values in groundwater originating from the sedimentary rock aquifer.

In a leachate plume setting, precipitation of FeS, FeS_2 and $FeCO_3$ may deplete groundwater from sulfur or iron and precipitation of $MnCO_3$ may deplete groundwater from Mn (Christensen et al. 1994). Other studies on leachate plumes emitted by landfills demonstrated that groundwater affected by leachate is often surpersaturated with regards to calcite ($CaCO_3$) and siderite ($FeCO_3$). Since iron-reduction kinetics is important for assessing long-term potential for aquifer attenuation (van Breukelen 2003; Keldsen 2002), and siderite and calcite are key minerals that precipitate during leachate migration, saturation indices (SI) of groundwater in relation to iron-bearing minerals, manganese-bearing minerals and calcite should be further investigated. Figure 7.13 shows saturation indices of interest in leachate plume attenuation (goethite, siderite and calcite). Saturation indices along the modeled cross-section (Figures 7.2 and 7.3) show that the leachate plume is saturated with regards to goethite and siderite. As the leachate plume attenuates, the calcite SI increases on either side of the landfill and goes back to natural background values farther along the way. Calcium and iron concentrations are also shown in Figure 7.13 with regards to chloride, which is a conservative tracer. Away from the landfill, iron concentrations are seen to be reduced much faster than chloride, indicating that attenuation reactions take iron out of the groundwater, whereas calcium behaves in a similar way as chloride, thus showing that it is an indicator of leachate provenance at this site and is not involved in significant attenuation reactions.

Even though SI may indicate that a mineral will tend to dissolve or precipitate, the reaction may not occur because of kinetic constraints and other factors, such as : solid solution complicating the mineral composition, chemical formula of the minerals that may increase the magnitude of SI, errors in analyses, unreliable thermodynamic data, etc. (Zhu and Anderson 2002). Furthermore, the SI does not provide information about reaction rates or the mass of the mineral involved (Zhu and Anderson 2002). Therefore, this analysis only provides indications of potential mineral reactions in relation to leachate attenuation in the aquifer system. Finally, it is difficult to identify proper saturation indices specific to a site for geochemical interpretation since the elements found in large concentrations in leachate also have elevated SI values for all the minerals containing these same specific elements. Therefore, a detailed description of the chemistry of the aquifer material can help define the metal species that can participate in reactions.

7.4.3 Conceptual model of leachate attenuation

Leachate composition varies significantly amongst landfills depending on waste composition, waste age, and landfilling technology (Kjedsen 2002). Leachate from landfills is typically strongly reduced, contains high amounts of organic compounds and would therefore have a great capacity for donating electrons during reactions. These electrons must be accepted by aquifer material and aqueous species. The investigation conducted in this study on the effects of hydrogeological and geochemical heterogeneity on natural attenuation of a leachate plume provides insights on the mechanisms and processes taking place at the sub-watershed scale. Figure 7.4 provides the distributed geochemical conditions at the site and Figure 7.3 shows a detailed conceptual model of the heterogeneous hydrogeological conditions.

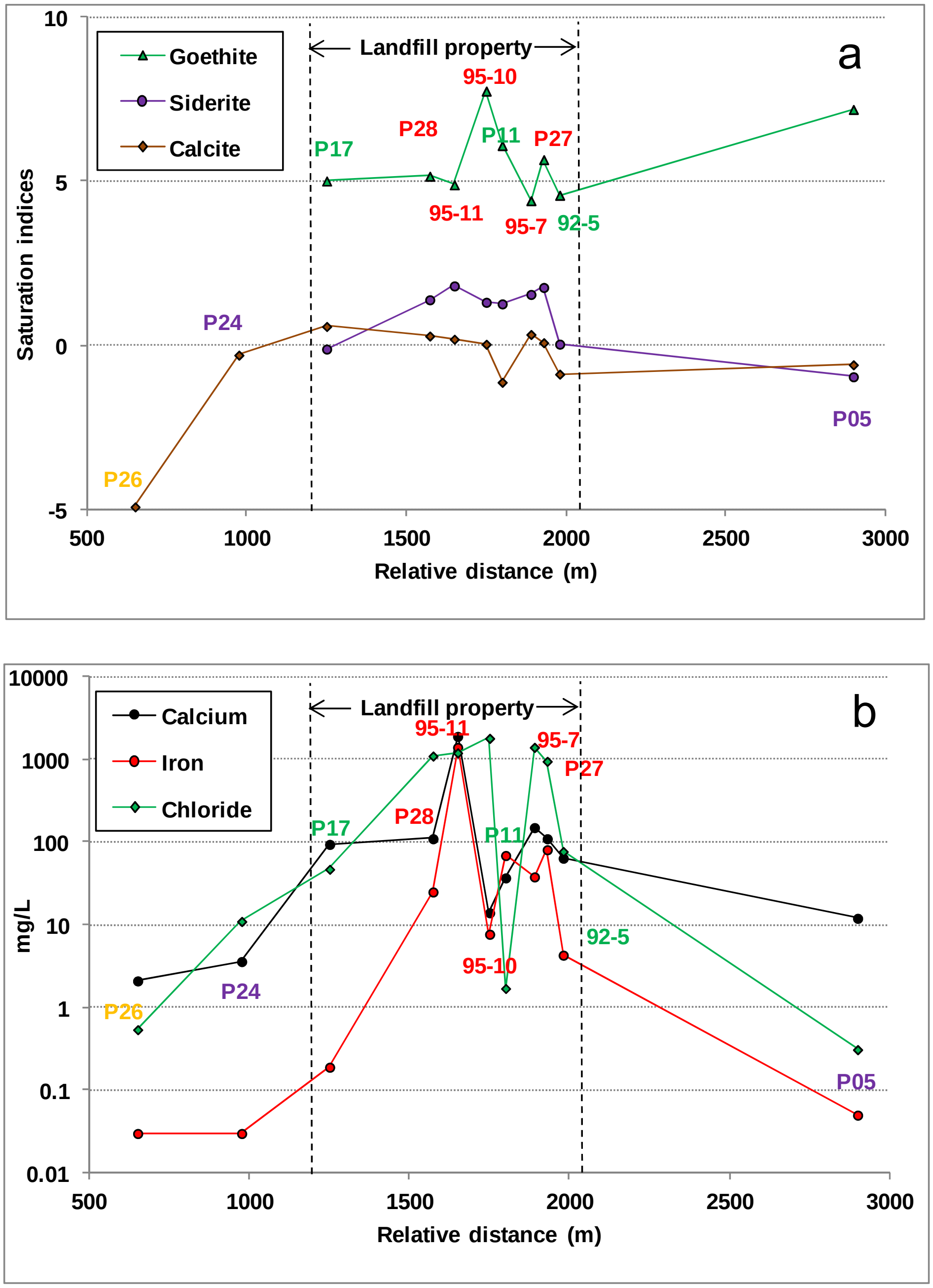

Figure 7.13 : Geochemical conditions along part of cross-section A-A' (Figure 7.3) a) Saturation indices (SI) showing that the leachate plume is oversaturated relative to goethite and siderite. As the leachate plume attenuates, the calcite SI increases on either side of the landfill and goes back to natural values farther. b) Calcium and iron concentrations are illustrated in relation to chloride to indicate the extent of the leachate plume (background [Cl] is below 10 mg/l)

The leachate plume extent was primarily defined with bulk electrical resistivity data of groundwater and from direct push soundings with CPT/SMR in the first phase of the characterization program. Zones where electrical conductivities were higher when compared to natural background values for the study zones were thus identified (Tremblay et al. 2013). Afterward, groundwater geochemical data collected from multiple observation wells confirmed the composition of the contamination source and the main leachate plume migration paths. Graphical analysis of geochemical data also allowed the identification of two distinct attenuation paths (acid and oxic versus neutral and suboxic), which has implications for leachate migration and attenuation. The overall field work sequence in three phases at the sub-watershed scale was thus essential to establish the variability of natural geochemical conditions and to understand the implications of leachate mixing with different groundwater types. The results from the second phase of the characterization revealed that the plume reaches the stream to the south of the landfill with higher concentrations than elsewhere and that ammonium concentrations are present at that stream. However, ammonium does not reach the observation wells at the northern limit of the landfill property. Instead, higher concentrations of calcium can be observed at that location. These concentrations can be the result of dissolution of calcite by CO_2 in the leachate produced by the reductive dissolution of primarily Fe oxide (Christensen et al. 2001). In comparison, concentrations of chloride are clearly reduced in both north and south leachate migration directions. Below the decommissioned waste pile, elevated concentrations were found down to 11-15 m depths. This observation corroborates with the $^3H/^3He$ groundwater age measured at depth (40 years) at an analogous location and the age of the waste site (39 years). No concentration of organic compounds or heavy metals were recorded in groundwater farther than 50 m from the waste pile in the northern direction, however, phenol concentrations above provincial standards were recorded all the way to the stream in the eastern portion of the landfill property (southern direction).

This final phase of the investigation with geochemical tracers provided indications of prevailing attenuation processes. Table 7.4 describes the leachate plume attenuation processes depending on geochemical conditions. The samples collected in wells located within 100 m of the former waste pile in a homogeneous geological environment revealed acidic and oxic groundwater. Other groundwater samples were collected in wells located farther than 100 m from the former waste pile where leachate mixes with neutral and semi-anoxic groundwater along its migration paths. The pH is too high to release more metals into solution so concentrations decrease to background values along these paths. Finally, from the data collected, the ammonium plume has not reached any well located in heterogeneous settings at more than 100 m, but has reached the wells located within 100 m in homogeneous environments. The heterogeneous environment provides better potential adsorption sites for ammonium and the plume is therefore retarded along this path. As the leachate plume migrates through distinct geological environments, different attenuation mechanisms thus take place.

Tableau 7.4 : Attenuation of the leachate plume depending on geochemical conditions

Geochemical Context	Spatial Location	Main processes
Strongly reducing and neutral conditions in waste pile	Within or close to the waste pile	Dolomite dissolution, calcite solution, calcite precipitation Ions exchange, ions precipitation, adsorption, sorption, iron and sulfate reduction
Acidic, oxic and homogenous geology, wells are located within 100 m from the waste pile	Along the groundwater divide line, mostly on the eastern section of the former landfill	Ions exchange, iron precipitation, calcite solution Re-dissolved metal available in the sediments such as: Co, Cu, Al, Ni, Fe, Mn. Groundwater has shorter residence time
Neutral, semioxic and heterogenous, waste are located over 100 m from the waste pile	On either sides of the waste pile from the center of the former landfill extending to west	Ions exchange, ions precipitation, sorption, iron reduction, calcite precipitation pH too high to dissolve metals from the sediments.

The numerical model based on the detailed conceptual model reproduced the hydrogeological conditions and was verified with hydraulic and geochemical data. The numerical simulations showed that the concentration of pollutants should not increase above the provincial standards within the next 35 years. The simulations also demonstrate that although ammonium concentrations can be remobilised, the dilution effect observed on the chloride plume should be sufficient to significantly decrease the ammonium concentrations, even without attenuation processes acting on ammonium.

The natural attenuation efficiency can be evaluated on the basis of the general hydrogeological understanding gained at the site investigated. The geological setting of the site seems to provide efficient natural attenuation. However, temporal monitoring should be implemented to follow potentially evolving future conditions, such as climatic conditions or land use.

7.5 CONCLUSIONS

A 2D numerical model and geochemical tracers were used to evaluate, respectively, the effects of hydrogeological and geochemical heterogeneity on the natural attenuation of a leachate plume emitted by an unlined decommissioned landfill in a shallow granular aquifer. The 2D numerical flow and mass transport model was developed on the basis of a detailed conceptual model that is representative of aquifer heterogeneities with regards to hydrostratigraphy and groundwater geochemistry. This conceptual model and its related data used for the development and history-matching of the numerical model were derived from the integration of multi-source field data provided by a previous characterization (Tremblay et al. 2013). This characterization, carried out in three phases over the 12 km^2 sub-watershed that encompass the landfill and surrounding streams, was essential to define site-specific conditions and understand the variability of those conditions.

The selected cross-section passed through the landfill along groundwater flow paths. Significant efforts were dedicated to the verification of numerical simulation results with hydraulic head data but also with geochemical data (groundwater age and geochemical tracers).The mass transport numerical model predictions for the conservative ion chloride were verified with geochemical data (groundwater age and geochemical tracers). Results from the numerical model showed that the interbedding of fine material with coarser sediments in the aquifer limits groundwater flow and induced relatively long groundwater residence time in the deepest portion of the granular aquifer (40-60 years). Moreover, the leachate plume is strongly diluted along its migration path; which favours its attenuation before emerging in the surrounding streams.

Geochemical data were a key component in assessing the effect of heterogeneities on migration and attenuation of the landfill leachate plume. Geochemical data were used to constrain the numerical model by verifying the simulated leachate plume migration paths and by constraining the hydraulic properties needed to reproduce groundwater residence times and chloride plume extent. The hydrogeophysical characterization data provided the level of details required to reassess the conceptual and numerical models as constrained with geochemical data. Furthermore, graphical interpretations of geochemical data provided insight on the natural environment of the semi-confined aquifer in the sub-watershed and into the geochemical processes controlling the plume evolution. Graphical analysis of geochemical data allowed the identification of two distinct leachate attenuation paths in relation to the geochemical conditions under which the leachate migrates and attenuates. It was possible to identify the attenuation mechanisms (dilution, and mass loss through sorption, and oxidation induced precipitation) of the leachate through the rapid reduction of major ion and metal concentrations that presently prevent the leachate plume concentrations from exceeding standards beyond the landfill property limits.

The detailed knowledge of aquifer conditions allowed a better evaluation of the environmental risk on the receptors (streams). The geochemical data in combination with the 2D numerical modeling provided a better understanding of groundwater flow and natural attenuation mechanisms of the leachate plume at the former landfill. This improved knowledge of the site conditions will lead to a better assessment of the environmental risks to groundwater and surface water receptors. Finally, the applied integrated methodology and the tools used to characterize the aquifer and the leachate plume can be used to assess MNA at other sites located in complex geological settings. The results obtained from this approach provided the level of detail required to define the main attenuation processes, which are rather site-specific, in an efficient manner.

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CARACTÉRISATION INTÉGRÉE D’UN AQUIFÈRE
GRANULAIRE POUR L’ÉVALUATION DES PROCESSUS
GÉOCHIMIQUES INFLUENÇANT L’ATTÉNUATION
NATURELLE D’UN PANACHE DE LIXIVIAT

Partie 3

Synthèse, perspectives et conclusions

Chapitre 8

SYNTHÈSE, PERPECTIVE ET CONCLUSIONS

8.1 Sommaire de la thèse et relation entre les différents chapitres

8.1.1 Sommaire général

Ce projet doctoral avait comme objectif de définir l'efficacité des processus d'atténuation naturelle d'un ancien site d'enfouissement sanitaire tout en approfondissant des aspects scientifiques et techniques d'intérêt en hydrogéologie. Les objectifs scientifiques de la thèse étaient de développer une approche qui améliorait la compréhension des aquifères et qui faciliterait la modélisation de l'écoulement et du transport de masse en relation avec des problèmes de contamination. Le choix d'un site affecté par l'émission de lixiviat géré par atténuation naturelle permet le développement et la mise en œuvre de l'approche de caractérisation intégrée.

Les chapitres de mise en contexte de la thèse illustrent les aspects connus de la région d'étude avant le début du projet ainsi que les efforts déployés pour faire l'intégration de toutes les données historiques disponibles, afin de valider la cohérence des informations et de maximiser la compréhension du contexte et ainsi orienter les nouveaux travaux de caractérisation. L'approche globale développée, appliquée à une région d'étude de 12 km^2 entourant l'ancien site d'enfouissement sanitaire afin de définir l'efficacité de l'atténuation naturelle, est présentée en quatre étapes qui font l'objet d'articles scientifiques dans la thèse : 1) "Modèle conceptuel de la migration d'un panache de lixiviat dans un aquifère granulaire grâce à l'intégration des données d'une caractérisation détaillée"; 2) "Indicateurs directs et indirects de la distribution spatiale de la recharge et de l'émergence dans un aquifère granulaire superficiel affecté par un panache de lixiviat"; 3) " Hétérogénéité géochimique des sédiments d'un aquifère, de l'eau souterraine et du lixiviat d'un site d'enfouissement à l'échelle d'un sous-bassin versant"; et 4) "Effets de l'hétérogénéité hydrogéologique et géochimique sur l'atténuation naturelle d'un panache de lixiviat".

8.1.2 Article 1 : Développement de l’approche de caractérisation intégrée

Les modèles numériques doivent avoir les deux caractéristiques suivantes pour être utile à la compréhension du devenir et de la migration des panaches de contamination: 1) couvrir un territoire incluant les zones sources de contamination ainsi que les milieux récepteurs; et 2) être basés sur des modèles conceptuels représentatifs et tenant compte de l’hétérogénéité des aquifères. Malheureusement, les méthodes de caractérisation actuelles des aquifères ne fournissent pas les données requises pour développer ce type de modèle. Les objectifs de cette étude étaient de développer des procédures de caractérisation et d’intégration des données hydrogéologiques: 1) adapté à la définition régionale de l’hétérogénéité des aquifères (excédant quelques km^2) et 2) fournissant les données complémentaires requises pour supporter le développement de modèles numériques détaillés de l’écoulement de l’eau souterraine et du transport de masse.

Cette étude comprend la caractérisation multidisciplinaire d’un aquifère granulaire peu profond dans un sous-basin versant de 12 km^2 à l’intérieur duquel se trouve un ancien site d’enfouissement sanitaire qui émet un panache de lixiviat géré par atténuation naturelle (Figure 4.1). Utilisant des méthodes éprouvées, la séquence de caractérisation a été conçue pour optimiser chacune des méthodes de terrain en termes de localisation, d’échelle d’acquisition, de densité et de qualité (Figure 4.2). L’emphase a été mise sur l’acquisition de données hydrogéophysiques indirectes détaillées qui ont été intégrées avec des données hydrauliques et géochimiques de haute résolution (Figure 4.6). Le programme de travaux a fourni un important jeu de données de sources variées spatialement bien distribuées dans la région d’étude. L’intégration de ces données, grâce à la géostatistique, a permis de définir la géométrie de l’aquifère en 3D, l’hétérogénéité des matériaux granulaires de l’aquifère, les voies d’écoulement de l’eau souterraine, les contextes de recharge et d’émergence et l’étendue du panache de lixiviat (Figure 4.7). Cet article met l’emphase sur les premières étapes d’intégration qualitatives et géostatistiques des procédures développées, ce qui permet le développement d’un modèle conceptuel hydrogéologique détaillée (Figure 4.12).

Cette étape initiale est un prérequis à d’autres étapes d’intégration, telles que la prédiction d’hydrofacies et de valeurs de conductivité hydraulique (K), des simulations géostatistiques de K et l’étude des processus géochimiques et la modélisation numérique de l’écoulement du transport de masse. Ce niveau détaillé de caractérisation n'aurait pas été atteint dans le même laps de temps et avec les mêmes ressources en utilisant des méthodes conventionnelles. Cette méthodologie a été efficace en termes de coûts et de temps et elle peut être adaptée à tout type d'environnement hydrogéologique et d'échelle de région d'étude.

8.1.3 Article 2 : Investigation des zones de recharge et d'émergence

La compréhension des processus de recharge et d'émergence est essentielle pour la gestion environnementale et à l'évaluation des risques de pollution de l'eau souterraine ainsi que de l'eau de surface reliée hydrauliquement à un aquifère. Cette étude démontre l'utilité de mesurer plusieurs indicateurs superficiels afin de valider un modèle hydrogéologique conceptuel et pour clarifier les conditions limites d'un aquifère, particulièrement la recharge.

L'étude porte sur un sous-bassin versant de 12 km^2 où un ancien site d'enfouissement repose sur un aquifère granulaire hétérogène peu profond. Les indicateurs considérés sont la topographie, de multiples propriétés physiques et géochimiques des sols, la géochimie de l'eau souterraine à faible profondeur et celle de l'eau de surface, les types de sédiments sous les cours d'eau, les charges hydrauliques mesurées dans des puits superficiels en bordure des cours d'eau et des mesures de débit dans les cours d'eau à l'étiage (Figure 5.2). Ces indicateurs sont combinés à des données isotopiques de l'eau souterraine (^{18}O, ^{2}H, ^{13}C & ^{14}C, ^{3}He/^{3}H), des profils physicochimiques dans les puits et des données hydrauliques de haute résolution dans les puits d'observation pour établir les taux de recharge, l'écoulement de l'eau souterraine et les zone d'émergence, ainsi que leur impact sur la migration et l'atténuation naturelle d'un panache de lixiviat. Les résultats démontrent une grande variabilité des patrons de recharge sur la petite zone investiguée (de près de 0 à 420 mm/an) (Figure 5.13). Cette variabilité est surtout influencée par la topographie, l'hétérogénéité de l'aquifère, la nature des matériaux présents en profondeur et les directions verticales d'écoulement de l'eau souterraine. Des zones locales de résurgence de lixiviat dans les cours d'eau ont été identifiées et le flux de masse a été évalué à partir de multiples traceurs (chlorure, sodium, potassium, ammonium, carbone organique dissous, etc.) (Figures 5.5 et 5.6).

Ces résultats vont supporter le développement d'un modèle numérique de l'écoulement et vont orienter des recommandations par rapport à l'utilisation du territoire et à la gestion du risque environnemental relié à l'ancien site d'enfouissement sanitaire.

8.1.4 Article 3 : Définition des conditions géochimiques de l'eau souterraine

Cette étude a pour but de décrire les conditions géochimiques qui pourraient influencer la migration et l'atténuation d'un panache de lixiviat dans un sous-basin versant de 12 km^2 à l'intérieur duquel est situé un ancien site d'enfouissement sanitaire sans base imperméable qui émet un panache de lixiviat dans un aquifère granulaire peu profond. L'atténuation naturelle d'un panache de lixiviat est influencée par la géochimie naturelle de l'eau souterraine dans laquelle le lixiviat est émis. Cette géochimie est étroitement liée aux propriétés physiques et chimiques des sédiments qui composent l'aquifère. Cette étude a pour but de décrire les conditions naturelles qui influencent l'évolution géochimique du panache de lixiviat et de délimiter l'étendue de la contamination dans l'eau souterraine.

Les travaux sont basés sur l'interprétation des données de géochimie du lixiviat et des eaux souterraines dans la région d'étude (Figure 6.1). De plus, considérant l'importance du lessivage des sols sur la géochimie de l'eau souterraine, cette étude propose un plan de travail pour caractériser, efficacement et à moindre coûts, les propriétés physiques et géochimiques des sédiments d'un aquifère (Figure 6.2). Un outil de fluorescence X portable a été utilisé pour analyser la géochimie des sédiments. La géochimie des eaux souterraine a aussi été caractérisée grâce à une gamme étendue de paramètres analytiques. La géochimie des sols et de l'eau souterraine a été interprétée avec les méthodes statistiques multivariées d'analyse par composante principale et de classification hiérarchique ainsi qu'avec d'autres méthodes graphiques (Figures 6.4 et 6.8).

Les données géochimiques et leur interprétation ont permis de 1) préciser le modèle conceptuel de l'écoulement de l'eau souterraine dans le système aquifère (Figure 6.13); 2) comprendre les processus géochimiques le long des voies d'écoulement (Figures 6.9 et 6.10); et 3) d'identifier la signature du panache de lixiviat dans l'eau naturelle et ses conditions d'atténuation naturelle (Figure 6.11).

Les propriétés physiques et géochimiques des sédiments peuvent être liées à leur historique de déposition et ces propriétés ont fourni des indications sur le rôle de la chimie des sédiments sur les types d'eau souterraine, particulièrement au niveau du cycle du carbone. Cette étude démontre qu'il existe une grande hétérogénéité au niveau des sédiments, ce qui influence aussi la variabilité des types d'eau souterraine que l'on retrouve dans l'aquifère. Cette étude démontre aussi que ces conditions sont influencées par l'émergence d'eau souterraine provenant de l'aquifère rocheux fracturé sous-jacent à l'aquifère granulaire. Même si la géochimie de l'eau est hétérogène, il a été possible de définir les conditions naturelles dans l'aquifère et de reconnaitre l'eau influencée par l'émission de lixiviat. Cette géochimie naturelle variée fait en sorte que l'atténuation du panache de lixiviat est différente selon les conditions oxydantes ou plus réductrices retrouvées le long d'une voie de migration. Ces résultats vont être intégrés à un modèle numérique de l'écoulement de l'eau souterraine et de la migration de lixiviat pour assurer la compréhension nécessaire à la gestion du risque environnemental de l'ancien site d'enfouissement sanitaire.

8.1.5 Article 4 : Évaluation des processus d'atténuation naturelle

Le panache de lixiviat émis par l'ancien site d'enfouissement sanitaire dans un aquifère granulaire hétérogène peu profond est géré par atténuation naturelle. Afin d'évaluer l'efficacité de ce mode de gestion, les travaux documentés dans les articles 1, 2 et 3 ont permis la caractérisation détaillée du sous-bassin versant de 12 km^2 entourant l'ancien site d'enfouissement. Ces travaux ont montré que les conditions hydrogéologiques et géochimiques sont hétérogènes dans l'aquifère (Figures 7.3 et 7.4). La présente étude avait donc pour objectif d'évaluer les effets de ces différents types d'hétérogénéité sur l'atténuation naturelle du panache de lixivat.

Les effets de l'hétérogénéité hydrogéologique ont été évalués à l'aide d'un modèle numérique 2D coupe passant à travers le site d'enfouissement le long des voies d'écoulement et de migration du lixiviat (Figure 7.5). Des efforts particuliers ont été dédiés à la vérification de la représentativité du modèle numérique à l'aide de données hydrauliques mais aussi de données géochimiques (âge de l'eau et traceurs géochimiques) (Figure 7.6). Le modèle numérique a montré que l'alternance de couches fines et grossières dans l'aquifère limitait l'écoulement et conduisait à des temps de résidence relativement long dans la partie inférieure de l'aquifère (40-

60 ans) (Figure 7.7). Le panache de lixiviat est aussi fortement dilué lors de sa longue migration qui favorise sa dégradation naturelle avant son émergence dans les cours d'eau (Figure 7.7). Les analyses graphiques des données géochimiques ont permis de d'identifier deux voies d'atténuation naturelle du lixiviat en fonction des conditions géochimiques de l'eau souterraine dans laquelle migre le lixiviat (Figures 7.8 et 7.9). Les mécanismes d'atténuation du panache de lixiviat ont pu être identifiés et ils contribuent à la réduction rapide des concentrations en ions majeurs et en métaux (Figures 7.10 et 7.12), de sorte que l'eau souterraine n'excède aucune norme en dehors des limites du site d'enfouissement (Figure 7.13). La connaissance détaillée des conditions de l'aquifère permet une meilleure évaluation du risque environnemental que représente le panache de lixiviat sur les milieux récepteurs.

8.1.6 Relations entre les étapes des travaux

La Figure 7.1 (article 4) montre que chacun des quatre articles de la thèse documente une étape des travaux contribuant, individuellement et dans leur ensemble, à l'atteinte des objectifs scientifiques de cette recherche doctorale. L'article 1 documente le développement et l'application d'une approche de caractérisation de l'hétérogénéité des aquifères. Cette étape représente l'assise des travaux de recherche car elle permet l'acquisition des données qui vont assurer la compréhension du système aquifère, représenté schématiquement par le modèle conceptuel hydrogéologique de la région d'étude. Ces travaux ont aussi permis de définir les conditions d'écoulement et l'étendue du panache de lixiviat. L'article 2 raffine le modèle conceptuel hydrogéologique en établissant les conditions et les zones de recharge et d'émergence dans la région d'étude. Les taux de recharge vont aussi être nécessaires au développement du modèle numérique documenté dans l'article 4. Dans le cadre hydrogéologique défini par les articles 1 et 2, l'article 3 documente l'analyse des données géochimiques sur les sédiments et l'eau souterraine. Cette analyse permet de définir les conditions géochimiques naturelles de l'eau souterraine qui se révèlent fortement hétérogènes et contrôlées par la nature chimique des sédiments à travers lesquels se font la recharge et l'écoulement de l'eau souterraine. Ces travaux permettent aussi d'identifier l'apport d'eau souterraine de l'aquifère rocheux régional ainsi que les eaux affectées par la présence de lixiviat. Enfin, l'article 4 fait une analyse plus détaillée de l'atténuation naturelle du lixiviat et de l'impact de l'hétérogénéité hydrogéologique et géochimique sur les processus d'atténuation.

8.2 Originalité et contributions scientifiques

Les pages d'introduction de chacun des articles (chapitres 4, 5, 6 et 7) décrivent en détail les aspects originaux et les contributions des articles de la thèse. Conséquemment, la présente section ne fait qu'un bref rappel des aspects jugés les plus importants de ces articles.

L'article 1 a porté sur le développement et l'application d'une approche de caractérisation de l'hétérogénéité des aquifères. Cet article fait une contribution importante au domaine de l'hydrogéologie ainsi qu'aux nombreuses spécialités pouvant contribuer à la compréhension des systèmes aquifères (géologie, géophysique, géostatistique, géochimie, etc). Ces travaux sur l'hétérogénéité des aquifères se démarque d'abord par l'échelle des travaux qui visent une région relativement grande et incluant la zone source et les milieux récepteurs. Une telle échelle est requise pour les modèles numériques servant à évaluer la contamination d'un site et son impact potentiel sur les récepteurs. Ces travaux sont aussi originaux par le rôle et la séquence des méthodes de caractérisation utilisées. Bradford et Babcock (2013) ont récemment exhorté les responsables des caractérisations environnementales de procéder d'une façon similaire à celle employé dans nos travaux, soit en faisant des levés d'abord indirects (géophysique ou CPT) avant de réaliser des forages et d'installer des puits d'observation. Ces travaux se distinguent aussi par l'utilisation de puits crépinés sur toute épaisseur installés par enfoncement. Bien qu'il y ait des limitations à l'usage de tels puits pour la caractérisation géochimique, ils ont permis d'établir les conditions hydrauliques de façon détaillée et efficace. Enfin, une approche a été développée pour prédire les concentrations en solides dissous totaux à partir de la conductivité électrique mesurée avec le piézocône (CPT/SMR), ce qui a permis une délimitation efficace de l'étendue du panache de lixiviat. L'article 1 fait ainsi de nombreuses contributions technologiques et méthodologiques, mais il discute aussi de l'importance de la caractérisation intégrée pour l'établissement de modèles hydrogéologiques conceptuels dont le rôle n'est pas toujours reconnu à leur juste valeur.

L'article 2 documente l'utilisation d'indicateurs superficiels pour identifier les zones (et le taux) de recharge et d'émergence de l'eau souterraine et son interaction avec les eaux de surface. Ces travaux se distinguent aussi par l'échelle d'étude, qui est relativement détaillée par rapport aux travaux sur la recharge qui se font souvent à l'échelle régionale. L'intégration de multiples méthodes d'évaluation de la recharge de même que les indicateurs superficiels ont permis la compréhension des mécanismes fins et du rôle des conditions du sol et de leurs effets sur les conditions géochimiques de l'eau souterraine peu profonde. Ces résultats, de grand intérêt, supportent donc l'utilisation plus généralisée des indicateurs employés lors de ces travaux, surtout qu'ils ne sont pas coûteux ou difficiles à acquérir. L'approche est aussi d'intérêt car elle a permis d'évaluer la distribution spatiale de la recharge ainsi que les interactions aquifère-cours d'eau qui représentent des sujets de recherche important en hydrogéologie. Ces approches peuvent aussi avoir une incidence sur l'évaluation du risque sur les récepteurs, particulièrement les cours d'eau.

L'article 3 document une étude des facteurs contrôlant les conditions géochimiques hétérogènes d'un aquifère et leur importance dans l'atténuation naturelle de contaminants. Ces travaux présente encore une fois le développement de méthodes originales de caractérisation; cette fois pour la chimie des sédiments de l'aquifère et des sols. Avec des méthodes multivariées conventionnelles, les données chimiques des sols et de l'eau souterraine ont pu être analysées, d'abord pour définir des regroupements ayant des caractéristiques similaires, et ensuite pour établir des relations entre les sols et les conditions géochimiques de l'eau souterraine. Ces travaux donnent un exemple intéressant d'établissement des conditions et des mécanismes géochimiques contrôlant l'évolution géochimiques naturelle de l'eau. Cette approche a aussi permis d'identifier les eaux affectées par le lixiviat et les voies de dégradation du lixiviat. Cette étude démontre aussi l'importance de tenir compte de l'hétérogénéité géochimique pour évaluer l'atténuation naturelle des contaminants émis dans les aquifères.

L'article 4 décrit l'évaluation de l'effet de l'hétérogénéité hydrogéologique et géochimique sur l'atténuation naturelle du lixiviat émis par le site d'enfouissement. Ces travaux se distinguent d'abord par l'utilisation complémentaire de la modélisation numérique et des données géochimiques pour évaluer les processus d'atténuation. Ces travaux font aussi l'utilisation d'une approche originale de calage du modèle numérique avec des données hydrauliques et chimiques, notamment l'âge de l'eau souterraine. Cette étude fait une démonstration probante des effets des conditions hydrogéologiques et géochimiques hétérogènes sur l'atténuation naturelle du lixiviat.

Globalement, ces travaux se distinguent par l'utilisation de méthodes variées, incluant travaux de terrain, méthodes statistiques, analyses de données géochimiques et modélisation numérique. Les apports des travaux se situent d'abord dans le développement de concepts, méthodes et approches permettant une meilleure compréhension des systèmes aquifères et des processus géochimiques. L'utilisation de méthodes variées (géologie, hydrogéologie, géophysique, géochimie, modélisation, etc.) de façon intégrée a aussi permis l'acquisition d'un ensemble de données exceptionnel qui permettra la réalisation de bien d'autres travaux de recherche. Enfin les travaux ont contribué à la compréhension d'un système hydrogéologique et géochimique complexe tout en abordant des questions de fond en hydrogéologie : modèles conceptuels, hétérogénéité hydrogéologique et géochimique, incertitude, atténuation naturelle.

8.3 Perspective

En hydrogéologie, il est d'intérêt de faire l'élaboration d'approches systématiques menant au développement de modèles conceptuels représentatifs. Dans le cas de travaux reliés à des sites contaminés, ces modèles conceptuels doivent être basés sur une large gamme de données de caractérisation, acquises à des échelles englobant les zones sources et les milieux récepteurs. De plus, l'intégration des données de sources variées devrait maximiser l'information dérivée des données directes et indirectes. Cette thèse avait pour objectif de faire une contribution à ces développements, ce qui permet de supporter la gestion de sites contaminés et l'évaluation du risque environnemental sur les milieux récepteurs associé à une source de pollution de l'eau souterraine.

Ces développements représentent un grand défi, car ils abordent deux questions élusives en hydrogéologie: 1) le développement de modèles conceptuels représentatifs, et 2) la quantification de l'incertitude (Bredehoeft et Konikow 2012; Sousa et al. 2012; Anderson et McCray 2011; Frind et al. 2006). La caractérisation de l'hétérogénéité (physique, chimique et biologique) de la sous-surface a un rôle crucial pour la protection et la réhabilitation des aquifères (Anderson et McCray 2011; Yang 2004). Même s'il est accepté que des données de terrain suffisamment denses et à une échelle appropriée soient essentielles à la compréhension d'un site contaminé dans la perspective de sa réhabilitation, certains peuvent argumenter sur la définition de "données de qualité" (Crumbling et al. 2003). Des données directes clairsemées ou le manque de sondages "in situ" avec des réponses disponibles en cours d'acquisition entravent la compréhension des systèmes hydrogéologiques (Yang et al. 2004). Cette étude démontre l'importance d'acquérir des données directes et indirectes, à une échelle géographique englobant les sources de contamination et les milieux récepteurs, en suivant une séquence d'acquisition pour assurer la localisation propice, la densité requise, la bonne échelle d'acquisition et la qualité des données de terrain. Cette étude démontre aussi que même si l'acquisition des données est faite selon les règles de l'art, il existe toujours des limites à chaque méthode de terrain, ce qui induit une part d'incertitude dans une étude. Le jugement professionnel sera toujours requis dans toutes les étapes d'une caractérisation, à partir de la planification de travaux, en passant par l'acquisition des données sur le terrain et leur traitements, et en allant jusqu'à l'interprétation des données. Il est essentiel de considérer la limite de chaque méthode de terrain et de connaitre la source des données pour en évaluer la validité et la précision avant d'en faire l'usage ou l'intégration. Finalement, l'acquisition professionnelle de données de terrain débute par la synthèse exhaustive des données déjà disponibles pour une zone d'étude pour ensuite choisir une combinaison adaptée de technique qui permettrait de capturer les aspects pertinents à investiguer de la sous-surface et aussi pour planifier l'acquisition à une échelle englobant les sources et les récepteurs.

Les plus grandes difficultés rencontrées lors de la réalisation de cette l'étude se retrouvent aux niveaux de l'intégration des sources variées de données ainsi que de la représentation et la communication des divers aspects investiguée les uns avec les autres. La mise en commun des réponses obtenues par les diverses méthodes pour arriver à définir avec cohérence la dynamique physique et géochimique d'un système aquifère complexe comporte de grands défis. Premièrement, l'erreur associée aux réponses obtenues par les diverses méthodes ne peut pas être prise en considération lors de l'intégration. L'échelle des unités de mesure varie et les données de sources variées sont incompatibles. De plus, il n'existe pas encore d'outils informatiques permettant d'intégrer efficacement tous les types de données qui ont été collectées au cours de cette étude et de quantifier de façon précise tous les processus impliqués. L'intégration requiert un effort immense d'interprétation et une compréhension intime du site à l'étude dans son ensemble de la part de l'utilisateur. Souvent les données sont seulement comparées en tenant compte des limitations de chacune des méthodes et l'interprétation finale de la zone d'étude repose sur les connaissances de l'utilisateur. Ensuite, une fois l'intégration complétée et les processus en place compris et cohérents avec les autres informations disponibles sur le système aquifère (recharge, hétérogénéité de l'aquifère, écoulement de l'eau souterraine, géochimie de l'eau, migration des contaminants), la représentation et la communication de tous ces aspects détaillés posent une difficulté additionnelle. De plus, sous quelle forme doit-on sauvegarder les diverses sources d'information et sur quelles bases, pour qu'elles puissent être utiles à d'autres utilisateurs et transférables aux gestionnaires du site contaminé. De plus, la documentation et la bonne organisation des données dans une base de données non seulement accroissent l'efficacité de leur analyse, mais est en fait essentielle pour permettre l'utilisation des données par de multiples collaborateurs (Cox et al. 2013). Ces questions ont été abordées, mais certainement pas résolues dans cette thèse. Cette thèse a fait des avancés au niveau de la caractérisation des aquifères, au niveau des efforts d'intégration et de gestion des sources variées de données qui une fois combinées aident à mieux comprendre les systèmes aquifères complexes, mais il reste encore beaucoup de travail à faire pour gérer les sources variées de données de manière efficace, les rendre disponibles, les intégrés et les représenter.

La pertinence socioéconomique de l'étude repose dans l'avancement de la compréhension des systèmes d'écoulement; en général et en relation avec la contamination des eaux souterraines. Il existe de plus en plus de sites contaminés et la compréhension de la dynamique d'écoulement de l'eau souterraine à l'intérieur et autour de ces sites est nécessaire à l'application de mesures de contrôle, de réhabilitation et de suivis efficaces. Les travaux de caractérisation réalisés par l'étudiante et l'intégration des sources variées de données contribueront aux développements et à l'utilisation d'approches novatrices en hydrogéologie. Ces méthodologies efficaces permettent de répondre plus rapidement aux questions soulevées par un site contaminé et guident la sélection de mesures de réhabilitation efficaces. La méthodologie employée a permis une détermination détaillée des conditions du site à l'étude et les connaissances ont été vulgarisées et transférées à l'organisme qui gère le site étudié (la RIGDCC), assurant une meilleure compréhension des enjeux environnementaux spécifiques au site et à la gestion éclairée de l'utilisation de la propriété en fonction des nouvelles connaissances.

8.4 Travaux futurs potentiels

Cette étude aborde des problématiques importantes en hydrogéologie par rapport aux développements de modèles conceptuels représentatifs pour guider des études plus spécifiques sur la recharge, la géochimie, le risque environnemental et la modélisation numérique. Les méthodes d'intégration emploient une variété d'approches, de façon qualitative et quantitative, notamment les coupes géologiques, l'intégration géostatistique par krigeage et la conversion des valeurs de résistivité des sondages par enfoncement en estimés de matière dissoute dans l'eau souterraine. Les méthodes d'intégration utilisées permettent de développer un modèle conceptuel du système d'écoulement, en définissant les conditions hydrogéologiques, en identifiant les zones de recharge et d'émergence et en délimitant l'étendue du panache de lixiviat. Ces éléments sont essentiels pour la compréhension du système d'écoulement et le développement de modèles numériques représentatifs. Les travaux de recherche documentés dans cette thèse ont permis de répondre aux principaux objectifs initiaux, et ont aussi apporté de nouvelles questions. Cette section présente les incertitudes ainsi que des travaux qui pourraient être complétés dans la région d'étude à partir, en grande partie, des données présentées dans cette thèse et en relation avec les objectifs établis.

8.4.1 Nouvelles connaissances pour mieux comprendre la région d'étude

La géométrie de l'aquifère, particulièrement son épaisseur, varie en fonction de la topographie de la surface du roc qui est très variable dans la région d'étude. La base de l'aquifère granulaire est le till qui repose sur le roc. Il s'agit d'un till très compact et silteux qui varie en épaisseur et qui est absent par endroit. L'amincissement du till a des implications importantes par rapport à l'écoulement de l'eau souterraine et les échanges avec l'aquifère de roc fracturé sous-jacent. Des matériaux fluvioglaciaires très perméables ont aussi été a été identifiés en profondeur à certaines localisations. Ces matériaux sont très grossiers et cela peut avoir une implication au niveau de l'écoulement. Par contre, leur continuité demeure toujours inconnue. Enfin, une autre unité aquitard a été identifiée en profondeur. Il s'agit d'un silt glaciomarin d'épaisseur variable et dont l'étendue et la continuité demeurent aussi inconnues. Les relations entre l'aquifère et les cours d'eau pourraient aussi être précisées avec les traceurs géochimiques. Une caractérisation complémentaire visant à donner des précisions sur ces questions permettrait de mieux comprendre les conditions d'écoulement dans l'aquifère de même que les échanges avec l'aquifère rocheux sous-jacent et les cours d'eau.

Bien que l'efficacité de l'atténuation naturelle du lixiviat ait été évaluée dans le cadre des présents travaux, cela ne représente pas une évaluation complète. Pour évaluer l'atténuation de façon exhaustive, il faudrait compléter les étapes requises de l'USEPA tout en développant d'autres aspects scientifiques (faire de la géochimie détaillée à des endroits d'intérêts). Les données géochimiques ont permis de comprendre la géochimie naturelle de la région d'étude et d'identifié deux types d'atténuation (réducteur ou oxydant) selon le contexte naturel. Par contre, les puits étaient trop éloignés les uns des autres pour pouvoir faire de la modélisation géochimique et comprendre en détail les mécanismes de dégradation et les quantifier. De plus un suivi de l'évolution de la source de la contamination serait requis. Donc, des travaux complémentaires permettraient de mieux caractériser et d'approfondir la compréhension des taux de dégradation des contaminants et particulièrement de l'ammonium. Ces travaux pourraient aussi permettre de valider les effets de la fluctuation de la nappe sur la dissolution du fer et de comprendre la source du soufre/sulfate grâce à un suivi temporel.

8.4.2 Développements scientifiques à partir des données acquises et des approches développées

Les données, connaissances, approches et concepts développés dans le cadre de cette étude doctorale pourraient permettre de poursuivre d'autres avancées scientifiques sur des thèmes connexes ou de faire des applications spécifiques. La présente section décrit certains des développements qui pourraient faire suite aux travaux doctoraux documentés dans cette thèse.

Les travaux de caractérisation hydrogéologique réalisés dans le cadre de cette recherche doctorale ont produit un jeu de données exceptionnel. Ces données ont été exploitées pour définir un modèle conceptuel et pour considérer l'hétérogénéité de la distribution des matériaux dans le modèle numérique 2D en coupe verticale. Cependant, ces données pourraient être exploitées de façon beaucoup plus approfondie dans le cadre du développement des approches de représentation de l'hétérogénéité des aquifères. Certains de ces développements ont été réalisés dans le cadre de projets de 2^e ou 3^e cycles parallèles : reconnaissance des hydrofaciès et prédiction des profils verticaux de conductivité hydraulique (K) à partir des sondages CPT/SMR (Paradis et al. 2008); prédiction de K à partir des levés de résistivité et des sondages CPT/SMR (Ruggeri et al. 2013b); simulation géostatistique des hydrofaciès et K dans un modèle 2D coupe (Brunet et al. 2012). Cependant, l'intégration complète des données n'a pas été faite afin de produire un modèle 3D de la distribution spatiale des hydrofaciès et K pour l'ensemble de la région d'étude. Un tel modèle géologique 3D permettrait de développer un modèle numérique 3D de l'écoulement et du transport de masse pour l'ensemble de la région d'étude pour mieux vérifier les effets de l'hétérogénéité sur l'atténuation naturelle des contaminants.

Aussi, les travaux réalisés dans le cadre de cette recherche doctorale ont été basés sur la définition des conditions hydrogéochimiques à partir d'une seule caractérisation dans le temps.

Ces résultats pourraient être utilisés pour établir un plan de suivi temporel de la géochimie de l'eau souterraine et de surface afin de préciser l'efficacité de l'atténuation comme mode de gestion du panache de lixiviat. Ce suivi serait développé en fonction des besoins spécifiques du site et pourrait cibler les endroits et les paramètres à surveiller, diminuant l'effort de surveillance et les coûts.

L'étude détaillée des conditions et taux de recharge et d'émergence dans la région d'étude offre aussi la possibilité de poursuivre des travaux sur ce thème. Encore une fois, si le suivi des niveaux d'eau était poursuivi sur une plus longue période, ces données pourraient être combinées au développement d'un modèle numérique sur l'ensemble de la région d'étude pour faire l'évaluation de l'effet potentiel qu'auraient des changements d'utilisation du territoire ou la fluctuation des précipitations causée par le changement climatique sur les patrons d'écoulement de l'eau souterraine. L'incidence de ces changements potentiels de l'écoulement sur la migration et l'atténuation du panache de lixiviat pourrait ensuite être faite.

Les travaux réalisés ont montré que les conditions géochimiques dans la région d'étude sont très hétérogènes. Une investigation géochimique plus détaillée pourrait être faite pour mieux comprendre les échanges entre les sédiments et l'eau souterraine qui contrôlent ces conditions géochimiques. Ces travaux pourraient porter à la fois sur les zones de recharge et sur les sédiments pour mieux comprendre les processus naturels de l'évolution de l'eau souterraine et les échanges avec la matrice de l'aquifère. Dans le cadre de tels travaux, le XRF portable pourrait être testé en mode "*soil*" sur les échantillons de sédiments pour sa capacité à évaluer les concentrations d'éléments traces en lien avec les mécanismes d'atténuation. Ceci serait une méthode rapide qui pourrait être appliqué in situ pour identifier les espèces chimiques immobilisées par les sédiments. La modélisation géochimique pourrait être utilisée pour quantifier les processus géochimiques qui sont impliqués dans l'évolution naturelle de l'eau souterraine de la région d'étude si des points de contrôle plus rapprochés étaient mis en place.

De même, au niveau des processus géochimiques impliqués dans l'atténuation naturelle du lixiviat, plus de travail de détails pourrait être fait le long des deux voies d'atténuation identifiée (oxydante et réductrice). Afin de quantifier les réactions d'atténuation et statuer sur l'avenir du panache et la capacité d'immobilisation des contaminants de l'aquifère, de la modélisation géochimique pourrait être effectuée avec les types d'eau souterraine naturelle pour mieux comprendre les réactions d'atténuation. À cette fin, il faudrait que les points de contrôle de la géochimie soient plus près du site et plus rapprochés les uns des autres. Ces travaux pourraient aussi mettre l'emphase sur le devenir de l'ammonium, dont les mécanismes d'atténuation ne sont toujours pas compris, notamment à savoir s'il se dégrade ou s'il est seulement retardé et pourrait éventuellement être remobilisé si les conditions géochimiques venaient à changer.

L'approche de caractérisation intégrée développée dans le cadre de ce projet se voulait d'application générale et adaptable à d'autres échelles ou contextes hydrogéologiques. D'ailleurs, cette approche a déjà été utilisée dans le cadre de l'étude hydrogéologique régionale d'une région de 9000 km^2 en Montérégie Est (Lefebvre et al. 2011). Il serait toutefois opportun de faire une nouvelle application de cette approche à un autre site contaminé pour affiner l'approche et préciser son potentiel d'adaptation et d'application à d'autres contextes ou à d'autres types de contaminants.

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CARACTÉRISATION INTÉGRÉE D’UN AQUIFÈRE GRANULAIRE POUR L’ÉVALUATION DES PROCESSUS GÉOCHIMIQUES INFLUENÇANT L’ATTÉNUATION NATURELLE D’UN PANACHE DE LIXIVIAT

Partie 4

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