Assessing potential impacts of shale gas development on shallow aquifers through upward fluid migration: A multi-disciplinary approach applied to the Utica Shale in Eastern Canada

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Abstract

Potential impacts of shale gas development on shallow aquifers has raised concerns, especially regarding groundwater contamination. The intermediate zone separating shallow aquifers from shale gas reservoirs plays a critical role in aquifer vulnerability to fluid upflow, but the assessment of such vulnerability is challenging due to the general paucity of data in this intermediate zone. The ultimate goal of the project reported here was to develop a holistic multi-geoscience methodology to assess potential impacts of unconventional hydrocarbon development on fresh-water aquifers related to upward migration through natural pathways. The study area is located in the St. Lawrence Lowlands (southern Quebec, Canada), where limited oil and gas exploration and no shale gas production have taken place. A large set of data was collected over a ~500 km^2 area near a horizontal shale gas exploration well completed and fracked into the Utica Shale at a depth of ≈2 km. To investigate the intermediate zone integrity, this project integrated research results from multiple sources in order to obtain a better understanding of the system hydrodynamics, including geology, hydrogeology, deep and shallow geophysics, soil, rock and groundwater geochemistry, and geomechanics. The combined interpretation of the multi-disciplinary dataset demonstrates that there is no evidence of, and a very limited potential for, upward fluid migration from the Utica Shale reservoir to the shallow aquifer. Microbial and thermogenic methane in groundwater of this region appear to come from the shallow, organic-rich, fractured sedimentary rocks making up the regional aquifer. Nonetheless, diluted brines present in a few shallow wells close to and downstream of a normal fault revealed that some upward groundwater migration occurs, but only over a few hundred meters from the surface based on the isotopic signature of methane. The

methodology developed should help support regulations related to shale gas development aiming to protect groundwater.

Keywords: shale gas, intermediate zone, aquifer vulnerability, upward fluid migration

1 Introduction

1.1 Context

The current pace of economic development is placing increasing demands on the exploitation of natural resources such as unconventional hydrocarbons, minerals and geothermal energy. These deep subsurface industrial activities are complex and may have significant environmental impacts, notably on freshwater aquifers used for water supply. So far, shallow aquifer vulnerability related to activities carried out at great depths (including hydraulic fracturing) has been the object of a limited number of studies, mainly because 1) the possibility of upward fluid migration through natural preferential pathways is perceived by some as being highly improbable and is still the object of an ongoing scientific debate (Vengosh et al., 2014; Birdsell et al., 2015; Lefebvre, 2017) and 2) very little information is available on the intermediate zone, located below shallow aquifers and above the zone targeted by the hydrocarbon industry (see Figure 1). It is, however, generally accepted that a natural connection between shallow and deep formations may be possible if large-scale permeable discontinuities (such as fault zones) are present across this intermediate zone, constituting preferential migration pathways (Gassiat et al., 2013; Kissinger et al., 2013; Vengosh et al., 2014; Raynauld et al., 2016; Lefebvre, 2017). The expression of vertical fluid migration is difficult to directly visualize in continental settings. However, in marine environments, pockmarks and venting hydrocarbons in the water column are commonly observed and represent useful indicators of active upward fluid migration (Pinet et al., 2008).

One of the major societal concerns regarding the development of deep-seated unconventional oil and gas is the risk of shallow groundwater contamination due to upward migration of post-fracturing flowback fluids and hydrocarbons from deep geological formations (Vengosh et al., 2014; Vidic et al., 2013; BAPE 2014; CCA 2014; EPA 2016). This concern is even stronger in regions where aquifers represent the main water supply and where hydrocarbon development has not yet taken place (Raynauld et al., 2016). As a result, these perceived environmental issues have sometimes led to moratoria on hydraulic fracturing, such as in eastern Canada and northeastern USA, while in other areas (mostly where oil and gas production has been on-going for decades), concerns and regulatory efforts focussed on how to minimize or mitigate impacts (Esterhuyse, 2017).

A number of studies have been conducted to examine the potential for fluid migration from deep unconventional reservoirs to surficial freshwater aquifers. Although leakage from faulty casings appears to be the main mechanism by which gas could migrate from depth (Dusseault and Jackson; 2014; Nowamooz et al., 2014; Soeder et al., 2014; Ryan et al., 2015; Sherwood et al., 2016; Lefebvre, 2017), several authors have stressed the need for a better understanding and representation of potential preferential flow pathways (Gassiat et al., 2013; Kissinger et al., 2013; Birdsell et al., 2015; Reagan et al., 2015; Grasby et al., 2016; Raynauld et al., 2016). Most of these studies have assessed the processes and conditions that could lead to upward fluid migration using numerical modelling based on generic conditions that may not represent the specific context of a given area.

On the other hand, many studies looking for indications of impacts on shallow aquifers have relied almost exclusively on geology and groundwater geochemistry and isotopes (e.g., Osborn et al., 2011; Warner et al., 2012; Jackson et al., 2013a; Molofsky et al., 2013; Darrah et al., 2014; McIntosh et al., 2014; McMahon et al., 2014; Moritz et al., 2015; Siegel et al., 2015; Humez et al., 2016; Nicot et al., 2017a, 2017b & 2017c). Early studies had suggested a link between groundwater quality and the distance to shale gas wells (Osborn et al., 2011; Jackson et al., 2013a), which had led to a scientific debate about the occurrence or not of such groundwater degradation. More recent studies in regions with or without shale gas exploitation have concluded that hydraulic fracturing and shale gas exploitation cannot be directly related to groundwater quality degradation (Siegel et al., 2015; Humez et al., 2016; LeDoux et al., 2016; Sherwood et al., 2016; Harkness et al., 2017; McMahon et al., 2017; Nicot et al., 2017a, 2017b & 2017c). Harkness et al. (2017) have stressed that controversies related to the potential for contamination from hydraulic fracturing have likely stemmed in large part from the lack of predrilling datasets (including a comprehensive suite of geochemical tracers), emphasizing the need to develop an understanding of the critical factors controlling the presence of elevated levels of hydrocarbon gas and salts in many aquifers located above shale gas reservoirs. McMahon et al. (2017) also warned that shale gas exploitation is a recent activity compared to the long residence time of groundwater, which may imply that today’s absence of detectable impact does not mean that no impact would be observed in the long term.

While organic and inorganic groundwater geochemistry (including isotopes) represents a powerful tool, it relies on phenomena that are already happening and cannot be used only by themselves to fully understand, predict or eventually avoid potential contaminations from deep units. Therefore, groundwater geochemistry and isotopes should be complemented with other direct and indirect data to obtain a better knowledge and understanding of the intermediate zone, including its fracturing, its hydrogeological properties, its ability to transfer fluids and thus, its integrity. Industry-driven rock geochemistry, geomechanical and geophysical characterization have almost exclusively focussed on the deep hydrocarbon reservoir in an attempt to characterize and improve hydrocarbon recovery, thereby largely ignoring the intermediate zone. The need for research on the role of natural fractures and faults as pathways for methane and other fluids to reach shallow aquifers has been stressed by Ryan et al. (2015).

In spite of a significant recent increase in research on potential impacts of deep resource exploitation to shallow groundwater quality, there is currently no scientific consensus on how to assess these impacts based on the specific context of a basin or shale gas exploitation area. This highlights the importance of collecting scientific data to increase knowledge, inform debates and provide tools to help regulators protect and manage groundwater resources in areas where unconventional hydrocarbon resources are exploited.

In an effort in this regard, a holistic integrated study aiming to assess potential impacts of shale gas activities on shallow aquifers in the St. Lawrence Lowlands (southern Quebec, Canada) was initiated in 2012. This region only experienced limited exploration activities with no commercial production. The purpose of this study was to investigate a 500 km^2 area and evaluate whether its geological context presents a risk of aquifer contamination prior to any potential large-scale drilling and fracturing activities. It was deemed critical to assess aquifer vulnerability to fluid upflow related to deep subsurface unconventional hydrocarbon exploitation in this frontier play, because groundwater is the local major source of water supply and strong societal protests against hydraulic fracturing had occurred. Since few data were available on the intermediate zone, in addition to conventional geological data our approach for this project involved the use of multi-source indirect data from different disciplines such as geophysics, geomechanics, hydrogeology, and rock and groundwater geochemistry. Extensive fieldwork was carried out including shallow well drilling (30 to 150 m deep), sampling of groundwater, soil, core and drill cuttings, shallow seismic surveys, borehole logging in shallow wells, airborne electromagnetic survey, structural surveys, hydraulic tests in sediments and bedrock, and groundwater monitoring. Multiple electric logs from shale gas wells as well as deep seismic profiles were also re-evaluated.

This paper presents an overview of the entire project and shows how diverse data types were integrated to assess the potential for fluid upflow from the unconventional shale gas reservoir. Sections 3 to 6 of this paper thus present an overview of the work carried out in each discipline, including its objective, fieldwork, methods used to interpret the acquired data and main results. Readers are referred to peer-reviewed papers on these specific studies to obtain more details. The understanding of the system hydrodynamics gained from the integration of results is discussed in section 7. A synthesis of other work that did not lead to useful results is provided in the Supplementary information.

The anticipated outcomes of this work were two-fold: 1) assess potential impacts of shale gas development on shallow aquifers through the identification of possible natural connections between deep and shallow geologic formations in the Saint-Édouard study area and 2) contribute to the development of a methodology that could be used to assess aquifer vulnerability relative to deep subsurface activities, which could serve as a basis for regulatory frameworks aiming to minimize the potential impacts to aquifers. To our knowledge, this is the first time that such an aquifer vulnerability assessment is made on the basis of specific local and regional conditions.

1.2 Description of the study area

The study area is located in the St. Lawrence Lowlands in southern Quebec (Canada), in the Saint-Édouard region, on the south shore of the St. Lawrence River, about 65 km southwest of Quebec City (Figures 2 and 3). The study area overlies the Upper Ordovician Utica Shale, which was explored from 2007 to 2010 to assess its potential for shale gas production. Talisman Energy (now Repsol Oil & Gas Canada Inc.) drilled two wells in this area: one vertical, A267, and one horizontal, A275 (Figure 3). The horizontal well was the most promising one drilled in the Utica Shale in southern Quebec with post-fracking initial gas production of 340 000 m^3/day (12 Mcf/d), which stabilized to 170 000 m^3/day (6 Mcf/d) after 25 days (Thériault, 2012).

The Utica Shale is part of the St. Lawrence Platform and extends over more than 10 000 km^2 (Figure 2). Only 28 shale gas exploration wells were drilled over this vast area, of which 18 were hydraulically fractured, before the *de facto* shale gas exploration moratorium came into force in 2010 (Lavoie et al., 2014). Due to this limited number of wells, the Utica Shale is considered a frontier play and, therefore, the St. Lawrence Platform is generally viewed as a "virgin" area with respect to hydraulic fracturing.

The topography of the ~500 km^2 study area shown on Figure 3 is relatively flat, having an elevation of nearly 90 m at the foot of the Appalachian piedmont, where well F2 was drilled (1.5 km south of the Talisman A267 and A275 wells), to ~30 m close to the St. Lawrence River. A ~20 m escarpment borders the south shore of the St. Lawrence River. Mean monthly temperatures vary between -11˚C (in January) and 19˚C (in July). Annual total precipitation is ~1170 mm, with about 23% falling as snow. Most residential wells in this area are open holes in the naturally fractured sedimentary rocks and have a steel casing across surficial sediments. Well depths vary from 20 to 80 m (average of ~50 m) and the wells are either under confined or semi-conditions (Ladevèze et al., 2016; Ladevèze, 2017), which allow them to be better protected against potential contamination from the surface (such as spills) than unconfined aquifers. The water table is relatively shallow in these low-permeability shales, ranging from 8.5 m deep to flowing artesian. The piezometric map (Ladevèze et al., 2016) shows that groundwater generally follows the topography, flowing from the Appalachians towards the St. Lawrence River.

2 Geological context

2.1 Regional geological context

In southern Quebec, a Cambrian – Ordovician sedimentary succession is preserved in the St. Lawrence Platform (Fig. 2) The succession records the evolution of the rift to passive margin and the final foreland basin events developed at the southern margin of the Paleozoic Laurentia craton (Lavoie, 2008; Lavoie et al., 2012).

In the upper 2 km depth of the study area, black organic-rich mudstones (Utica Shale and Sainte-Rosalie Group) are capped by shallowing-upward flysch (Lorraine Group) (Lavoie, 2008). The Utica Shale is dominated by calcareous black shales with minor siltstone and is overpressured (BAPE 2010; Chatellier et al., 2013).

The St. Lawrence Platform strata are sub-horizontal and locally affected by mesoscopic open folds. High-angle generally SE-dipping normal faults displace in a staircase fashion the Precambrian basement and the St. Lawrence platform succession up to the Utica Shale with increased thickness of the sedimentary package on the downthrown fault block (Castonguay et al., 2010; Séjourné et al., 2013) (Fig. 2). These faults are exposed along the NW edge of the St. Lawrence platform (Fig. 3) and extend to the SE below the Appalachian tectonic wedge. Previous geological interpretations (Konstantinovskaya et al., 2009; Castonguay et al., 2010) suggested that normal faults do not extend in flysch and molasse units (Lorraine, Sainte-Rosalie and Queenston groups). The St. Lawrence Platform is bounded to the southeast by a deformed zone where Middle to Upper Ordovician platform units are imbricated in several compressive thrust panels; this zone is known as the parautochthonous domain (St-Julien et al., 1983; Castonguay et al., 2010). The Aston Fault marks the contact between the largely undeformed autochthonous St. Lawrence Platform and the parautochthonous domain (Figs. 2 and 3). The contact of the parautochthonous domain with the Appalachians corresponds to a regional low-angle thrust fault known as the Logan's Line, which marks the position of the westernmost reach of allochthonous units (St-Julien et al., 1983; Castonguay et al., 2010) (Figs. 2 and 3). East of the Logan's Line, Paleozoic rocks of the Appalachians are steeply-dipping and involved in overturned NW-verging folds (Clark and Globensky, 1973).

2.2 Saint-Edouard geological setting

Surficial sediments in the Saint-Édouard area are usually thin (< 10 m) and made up of reworked tills and near-shore sediments of the former Champlain Sea, except in a few areas where fine-grained marine sediments have accumulated in local lows of the paleo-topography (Légaré-Couture et al., 2018; Rivard et al., 2018a).

Outcrops are sparse in this region, with the best sections exposed along the cliff on the shore of the St. Lawrence River and along small creeks (Clark and Globensky, 1973). The surface geology of the area consists of Upper Ordovician clastic-dominated units of the Lorraine Group (Nicolet and Pontgravé formations) and of the Sainte-Rosalie Group (Lotbinière and Les Fonds formations) (Fig. 3). The Lotbinière, Nicolet and Pontgravé formations are part of the St. Lawrence Platform (Fig. 3), whereas the Les Fonds Formation belongs to the parautochthonous domain (Fig. 3). The Lotbinière, Les Fonds and Nicolet formations consist of more or less calcareous mudstone and siltstone; these three units display variable content of organic matter and were shown to be in the oil window (Lavoie et al., 2016).

The area is characterized by several regional faults, which could hypothetically act as potential natural or enhanced migration pathways for deep fluids. The St. Lawrence Platform is cut by the Rivière Jacques-Cartier (RJC) normal fault system (Fig. 3), which limits the Lotbinière Formation to the southeast. The RJC fault is a lateral equivalent of the major Yamaska Fault (Fig. 2) and has significant vertical throw (see below). The Aston and Logan NW-verging thrust faults in the southeastern part of the study area limit the parautochthonous domain where the Les Fonds Formation is present. Between the RJC and Aston faults, the Upper Ordovician succession consists of the Nicolet Formation (Fig. 3).

The Upper Ordovician Utica Shale is considered hosting a significant volume of hydrocarbons (Dietrich et al., 2011; Chen et al., 2014 & 2017). Based on seismic (Lavoie et al., 2016) and hydrocarbon exploration well data (Thériault, 2012), the Utica Shale is present at variable depths in the study area: it was intercepted by wells at relatively shallow depths in the northern part of the area (271 m to 580 m) and at greater depths in the southern part (1857 m in the Saint-Édouard Talisman well). In all the wells, the Utica Shale is either stratigraphically overlain by the Nicolet Formation or structurally overlain by the Les Fonds Formation. The characteristics of each formation is provided in Lavoie et al. (2016).

3 Seismic reflection

Objective

2-D seismic surveys were used to investigate whether faults in the Saint-Édouard region reach the surface or not, as the latter could form primary control on fluid migration pathways and may act as hydraulic conduits connecting the shallow and deep subsurface (Caine et al., 1996; Faulkner et al., 2010; Bense et al., 2013). Seismic data consisted in one set of industry data originally collected in the early 1970s and reprocessed by Talisman Energy during the late 2000s, and two sets of near-surface data collected specifically for this project. The three seismic data sets aimed at being complementary by providing seismic images with different vertical resolutions of various depth intervals from the unconventional reservoir up to the surface.

In-house reprocessing of the industry data aimed at enhancing the resolution of the upper ~2 km bedrock reflections. However, acquisition parameters for the industry survey were set to image targets approximately located 1800 to 2800 m below the surface. Therefore, the first ~500 m were poorly imaged and difficult to interpret. New shallow seismic surveys were carried out to obtain key information on the near surface extensions of faults to complement the available industry survey. Acquired seismic lines thus included areas of the moderately dipping Logan’s Line and Aston thrust fault, and the steeply dipping RJC normal fault (Figures 2 and 3). At the regional scale, these structural features are relatively well documented in the deep subsurface (Konstantinovskaya et al., 2009; Castonguay et al., 2010; Thériault, 2012), but their potential near-surface extension remains equivocal in the Saint-Édouard area. Shallow seismic surveys also aimed to help in positioning observation wells in or near faulted zones, so as to compare the permeability and groundwater quality of these zones with those of surrounding areas.

Fieldwork and methods

Shallow seismic surveys were performed using two types of vibrating sources: the Minivib (20-310 Hz) typically used for groundwater studies in unconsolidated sediments and the Envirovib (20-110 Hz) tentatively used in this project to fill the imaging gap between 50 and 500 m. The Minivib was operated in the horizontal inline-mode (H1) and P and S arrivals were recorded using a 37.5 m long landstreamer equipped with 48 3-Component stations with a 0.75-m spacing (Pugin et al., 2009). The Envirovib was operated in vertical-mode and P-arrivals were recorded using one 480 m-long landstreamer including 96 vertical-component stations with a 5-m spacing. In the Saint-Édouard area, a total of 18 km and 15 km of shallow seismic data were collected respectively with Minivib and the Envirovib in 2013 and 2014.

Results

Results from the shallow seismic surveys were heavily impacted by the high velocity contrasts between the free surface (P-wave velocity of 300 m/s) or the thin Quaternary cover (P-wave velocity of 800 to 1700 m/s) and Paleozoic rocks (St. Lawrence Lowlands and Appalachians; P-wave velocity of 3800 to 4200 m/s). This high velocity contrast located at or almost at the free surface converts most of the seismic energy into surface waves, impairing shallow bedrock reflection imaging from low energy and high-frequency seismic sources. Despite these difficult geological conditions for seismic acquisition, the contact between the thin veneer of unconsolidated sediments and the bedrock was adequately imaged with the Minivib using P and S-waves recorded on the vertical and H1 components of the station. All data parametrization attempts and filtering processes did not allow the Envirovib survey to be interpreted with confidence as this source simply lacked the energy to go beyond the strong velocity contrast that characterizes the first interface.

Therefore, given the specific near surface geological conditions, the shallow seismic surveys failed to locate the surface extension of faults imaged by the deep seismic survey.

A new subsurface interpretation of the area, below ~500 m, was made possible through the reprocessing of the industry deep seismic profile (Fig. 4a). The end-product confirmed the overall architecture of the Paleozoic successions and imaged the geometry of the St. Lawrence Platform and parautochthonous domain in greater details compared to most publicly-available vintage seismic lines (Lavoie et al., 2016). Several fault splays belonging to the RJC fault zone could be traced with confidence, some of which possibly extending to the near surface. These faults could represent potential fluid migration pathways, as shown by red arrows in Figure 4b. The geometry of the Appalachians SE of the Logan's Line remains poorly resolved by seismic data, due to structural complexity (Lavoie et al., 2016).

Based on the interpretation of the deep seismic (and due to a lack of good-quality data from the shallow seismic surface), four observation wells were drilled and five residential wells were sampled on both sides of the mapped normal fault potentially extending to the surface to obtain hydrogeological and geochemical information (see sections 5 and 6).

4 Geomechanics

Objective

A geomechanical study was carried out using petrophysical wireline logs from shale gas exploration wells in the Saint-Édouard area. The objective was to estimate geomechanical properties for the Utica Shale and the overlying intermediate zone, so as to identify the presence or absence of barriers to the propagation of hydraulic fractures towards the surface. To calibrate these values, laboratory geomechanical tests of core samples from the Lorraine Group and Utica Shale from the shale gas well had been planned. Unfortunately, these laboratory tests did not provide reliable results. Therefore, they are only briefly described in the Supplementary information section and discussed in Séjourné (2016).

Data and Methods

Publically available LAS digital files were integrated into the software Petra®, while DLIS files provided by Talisman Energy, containing all recorded data during logging campaigns, were used to verify the integrity and comprehensiveness of the LAS files and to occasionally complement them. Available data included gamma ray, caliper, density porosity, neutron porosity, resistivity curves, photoelectric factor, P and S waves, as well as bulk density and its correction curve. For a few wells, spectral gamma ray and mineralogy logs were also available.

Three shale gas exploration wells in or close to the study area had a complete borehole logging dataset and were thus used for this work: Saint-Édouard, Leclercville and Fortierville (Séjourné, 2016 & 2017). Acoustic logs (P and S waves) were used to derive elastic properties such as Poisson's ratios and Young's modulus. An acoustic brittleness index was derived from these two properties and a mineralogical brittleness index was derived from the mineralogy logs to better understand the observed variations between the

Utica Shale and its sedimentary cover. Subsequently, five additional conventional hydrocarbon exploration wells (drilled between 1972 and 1979) for which S-waves were not available were also used to extend the spatial coverage (Séjourné, 2017). To this purpose, a synthetic S-wave was generated for these wells and synthetic elastic modules were then calculated, allowing the estimation of the geomechanical properties.

Results

Values for Young's modulus and Poisson's ratios show that a large mechanical contrast exists between the brittle calcareous Utica Shale and more ductile clayey shales of the Lorraine Group. This relationship is best exemplified by the brittleness indexes shown in Figure 5, where a sharp contrast is observed at the contact between the upper part of the Utica Shale and the base of the Lorraine Group. Figure 5 also illustrates the good agreement between results obtained from acoustic and mineralogy logs. During hydraulic fracturing, the propagation of induced fractures within the more brittle Utica Shale would be severely limited or even stopped by these overlying ductile units. The geomechanical study thus strongly suggests that units of the intermediate zone (Lorraine Group) represent an efficient barrier to the propagation of hydraulic fractures, thus providing a good protection for the shallow groundwater aquifers. Nonetheless, these geomechanical property values remain indicative and qualitative, and should be taken with caution in absence of calibration with laboratory tests (see the Supplementary information section and Séjourné, 2017).

5 Hydrogeology

Hydrogeological fieldwork included the drilling of 15 observation wells, borehole logging of these wells, hydraulic tests in wells and permeability tests in surficial deposits.

5.1 Well drilling

Objective

Drilling of 15 observation wells into the regional fractured rock aquifer provided cuttings or core samples and groundwater samples in key locations. These wells provided information about stratigraphy, hydraulic conductivity and natural fractures. Locations of observation wells were strategically chosen to gain information, first on specific areas to improve spatial and formation coverage and second in the vicinity of mapped fault zones, whose positions are uncertain.

Fieldwork

Of the fifteen observation wells that were drilled, seven were hammer drilled (F5, F6 and F10 to F14) and eight were diamond drilled (F1 to F4, F7, F8, F20 and F21). Characteristics of these wells are provided in Ladevèze et al. (2016) and Table S-1 of the Supplementary information and their location is shown in Figure 3. The diamond-drilled wells provided core samples for stratigraphic assessment and organic geochemistry. Observation wells have a depth range of 30 to 147 m and they are all open in bedrock. Each well was instrumented with a datalogger to record water levels and. three of these wells were also equipped with barometers.

5.2 Fracture characterization

Objective

The main purposes of acquiring data on natural fractures were to characterize the geometry of the fracture network affecting the sedimentary succession in the Saint-Édouard region and to assess the continuity of structural features from deep to shallow units. Information on fractures were derived from bedrock outcrops, shallow observation wells through borehole geophysical logging, and three shale gas exploration wells using Formation Micro Imager (FMI) logs for depths exceeding 560 m. Since this project aims to assess the potential for upward fluid migration from the shale gas reservoir to surficial aquifers, the study of fracture networks was a key component.

Fieldwork

Shallow borehole logging was carried out in 11 observation wells. This work aimed to collect data on fractures (orientation, dip and aperture), bedrock lithology, compressional (P) and shear (S) wave velocities in the bedrock, fluid temperature and electrical conductivity, as well as identify zones of fracture flow. The suite of tools consisted of natural gamma-ray and guard resistivity, optical and acoustic televiewers, full waveform sonic logs, fluid temperature, fluid conductivity, and heat-pulse flowmeter. Since ambient flow (i.e. natural upward/downward gradients in borehole fluid) was not observed in any well, an impeller (Grundfos Redi-Flo 2) pump was used to induce upward flow in boreholes for the heat pulse flowmeter testing. Data on fractures (orientation, density, spacing, cross-cutting relationships) were also collected from 15 bedrock outcrops, mainly located close to the St. Lawrence River (Ladevèze, 2017; Ladevèze et al., 2018a).

Methods

Fractures identified using borehole geophysical logging were classified as open, closed or broken zones. Open and broken features were further classified as "flowing" based on flow indications. . P- and S-wave velocities were computed from full waveforms using a semblance processing routine to obtain slowness (μs/m) of P and S waves, and calculate velocity logs (details are available in Crow and Ladevèze, 2015).

FMI images were used to examine fracture densities, fracture types (open or closed) and orientations. Estimates of apertures from these images were provided by Talisman Energy. Imaged vertical sections for the three shale gas wells varied from 560 m to 2320 m deep, but were mostly located in the lower part of the Lorraine Group and in the underlying Utica Shale and (below 1500 m). Images of horizontal sections for each shale gas well, drilled into the Utica Shale, over lengths varying from 920 to 1000 m, were also available.

Chronology of fractures was based on intersection relationships and through fold test in order to calculate the fracture attitudes prior to folding events. Fracture spacing was also estimated to characterize the spatial organization of these structures using variograms.

Results

Four fracture sets were defined: three sub-vertical sets (FS1, FS2 and FS3) and one sub-horizontal, corresponding to bedding plane fractures (BPF). FS1 and FS2 are orthogonal to each other and to the bedding planes. They can be found everywhere throughout the shallow and deep intervals, however in shale units they seem to be clustered in “corridors”, while in siltstone beds they are more uniformly spatially distributed. Fractures from FS3 are more sparsely distributed and were mainly observed in the Utica Shale. BPF could only be observed at shallow depths, but are assumed to be present deeper, based on other studies examining shale successions (Gale et al., 2007; Gale et al., 2015; Wang and Gale, 2016). Based on the similarities between the shallow and deep fracture datasets and also based on the regional geologic and tectonic history, it was assumed that the fracture network characterization could also be applied to the intermediate zone (Ladevèze et al., 2018a). Figure 6 presents conceptual models of the fracture network for the entire sedimentary succession of the Saint-Édouard area.

Fracture data obtained from the shallow and deep intervals revealed that most fractures are open in the shallow rock aquifer, while very few fractures are open at depth. Most of these shallow fractures are present within the first 60 m of the rock, and especially the upper 30 m (Ladevèze et al., 2018b). At depth, fractures from only one fracture set (FS1) were found to be quite commonly open in the shale gas reservoir (21% of all FS1 fractures), and thus, by extension, very likely in the intermediate zone. FMI images also showed that the calcareous and brittle Utica Shale is more densely fractured than the Lorraine Group units (Séjourné, 2017). Denser fracture networks were, however, observed in the close vicinity of some of the thrust faults, with fractures generally oriented in the same direction as the FS1 fracture set. Implications of their presence on groundwater flow are discussed in section 5.3.

5.3 Hydraulic properties, groundwater flow and confinement conditions

Objective

Hydraulic conductivities (*K*) of shallow and deep rock formations were estimated to better understand the system hydrodynamics and to define hydrogeological conditions, as well as the role of fractures in groundwater flow. For the shallow intervals, *K* values were acquired through *in situ* hydraulic tests and theoretical equations were used for the deep interval. The goal for the deep interval assessment was to propose a semi-quantitative estimate of hydraulic properties based on available data and to develop a conceptual model of the fracture network to make a preliminary assessment of potential upward fluid migration through these structural features.

Fieldwork

In situ hydraulic tests were carried out in shallow observation wells using slug tests. However, the least permeable wells (10^{-8} m/s or less, see Table S-1 in Appendix) were instead pumped for a short period using a low-yield impeller pump (Grunfos Rediflo2) to avoid very large drawdowns that would have taken weeks to recover (see details in Ladevèze et al., 2016).

Water-level and barometric measurements were recorded every 15 minutes using pressure transducers (dataloggers). The purposes of acquiring these data series were 1) to help define confinement conditions and 2) to estimate aquifer recharge for wells under unconfined conditions.

Methods

Hydraulic conductivities (*K*) for the shallow observation wells were estimated from slug tests using two interpretation methods (Bouwer and Rice, 1976; Hyder et al., 1994). Both methods provided similar results. For the deep interval, hydraulic properties (porosity and hydraulic conductivity) were estimated based on fracture apertures, fracture spacing and density (see representative elementary volumes in Fig. 8) integrated in the cubic law assuming laminar flow between two parallel plates (Snow, 1968; Bear, 1993). Porosity was estimated as a percentage of the fracture volume in a quasi-impermeable matrix, based on available fracture characteristics and estimates of inter-cluster (or inter-corridor) distance.

Results

Hydraulic conductivities (*K*) of the shallow interval, which is the active groundwater flow zone (upper 60 m of the rock aquifer), range between $2x10^{-9}$ and $1x10^{-5}$ m/s. The geometrical mean is higher in the autochthonous domain ($1.8x10^{-6}$ m/s) than in the other two domains ($2.3x10^{-7}$ m/s). In addition, the range of values in the parautochthonous and allochthonous domains is wider than in the autochthonous domain. Restricted to the upper part of the Lorraine Group, siltstone beds are less than 10 cm thick and they are present in meter- to decameter-thick intervals; they are densely fractured (see Figure 6). Hydraulic tests have not indicated different permeabilities close to known (mapped) faults compared to those of the adjacent domain. Since open fractures in the shallow aquifer can locally display some large apertures, the fracture network significantly contributes to the total porosity of the rock mass (which is approximately 8%).

As very few fractures are open at depth and generally belong to a single set (FS1, see section 5.2), these fractures should not increase significantly the shale porosity and permeability, and hence should not favour fluid circulation because the possibility for fracture interconnection is very limited. Moreover, clay-gouge was observed in cores of shale gas wells drilled in the thrust fault zone (Ladevèze, 2017); therefore, given the shale - siltstone dominated succession in the entire study area, the presence of clay-gouge in the undrilled normal fault zone is strongly suspected due to the presence of potential secondary inverse-compressive fault-anticlines suggested from the reprocessed seismic line over the trace of the RJC fault (Lavoie et al., 2016). Therefore, flow across fault zones is considered very unlikely in this region. Within the deep intervals (intermediate zone and Utica Shale), very low hydraulic conductivities (*K*) in the order of 10^{-12}-10^{-18} m/s were estimated with the cubic law. These values are in agreement with values found in the literature for the Lorraine Group and Utica Shale (BAPE 2010; Séjourné et al., 2013).

While estimated hydraulic conductivities are very low, two features could possibly locally increase permeability in the two fault zones of the study area. In the thrust fault zone, open fractures were found to be more frequent in the damage zone surrounding the fault planes. However, they are also mainly in the same direction as fractures from FS1, hence limiting possible fracture interconnections. In the normal fault zone, siltstone beds, which are more permeable than shale strata and appear to be relatively frequent in the upper part of the Lorraine Group (Lavoie et al., 2016), were inferred to have been dragged into the core of the normal fault (Ladevèze, 2018b), an assumption based on data from abandoned (old) hydrocarbon exploration well logs, various geological descriptions and geochemical findings (see section 6.1). This possible increase in permeability could not, however, be quantified, as no hydraulic data are available at depth in this region. Pressure gradient data for the shallow intermediate zone is not available either, although it is well known that the deeper Utica Shale is overpressured (Chatellier et al., 2013; Séjourné et al., 2013; Konstantinovskaya et al., 2009). Nevertheless, the possibility that these fault zones act as large-scale flow pathways seems extremely unlikely (Ladevèze et al., 2018b).

6 Geochemistry

The groundwater and rock geochemistry study mainly focussed on the characterization of hydrocarbons (concentration and isotopic composition) from observation wells. These results were compared to the organic geochemistry data from the deep Utica Shale.

6.1 Groundwater geochemistry

Objective

As the main objective of the project was to investigate the potential for upward fluid migration from the shale gas reservoir to freshwater aquifers, it was crucial to sample and analyze shallow groundwater to see if it contained thermogenic gas and, if so, to determine its provenance. Isotopic geochemistry is a valuable tool to investigate the origin (microbial or thermogenic) of methane and its possible relation with the shale gas reservoir. Therefore, a multi-isotope approach was used, which included both stable and radioactive isotopes.

Fieldwork and analyses

Groundwater was sampled from 14 of the 15 observation wells (well F14, located in the thrust fault zone and likely in gouge, remained dry throughout the duration of the project), as well as in 30 residential wells in order to obtain a better spatial coverage. Among the 44 sampled wells, seven were sampled regularly over a period of up to 2.5 years at intervals between one to four months (see section 6.3). Groundwater was sampled at specific targeted depths in observation wells, where borehole geophysics had previously located flowing fractures.

Observation wells were sampled using a low-flow yield according to the EPA guidelines (Puls and Barcelona, 1996) using a submersible pump (either an impeller or a bladder pump), or HydraSleeve bags for the deepest well (F21). These sampling techniques provided similar results when tested systematically in 10 wells over three sampling campaigns (Rivard et al., 2018b). Residential wells were also sampled at a low yield from an outdoor tap, prior to any treatment. Samples for alkane concentrations and isotopic composition were collected under a head of water (method described in Bordeleau et al., 2018a, 2018b & 2018c), which minimizes degassing but does not prevent it completely when methane is effervescent (Molofsky et al., 2016). Hence, when downhole concentrations are above the methane saturation point at atmospheric pressure, measured concentrations are expected to be underestimated compared to true downhole concentrations.

Groundwater samples were collected and analysed for a wide variety of compounds including major and minor ions and trace metals, alkane concentrations (C_1 to C_3), VOCs, stable isotope composition of water (δ^2H and $\delta^{18}O$) and of methane, ethane and propane (C_1 to C_3-$\delta^{13}C$ and -δ^2H; when concentrations allowed), dissolved inorganic carbon (DIC-

$\delta^{13}C$), ^{222}Rn, tritium (^{3}H), as well as radiocarbon (^{14}C) in methane and DIC, and finally ^{36}Cl. Some of these analyses are quite atypical for this type of project (e.g., ^{36}Cl, CH_4-^{14}C) and they provided important information on groundwater age and hydrocarbon source. Analytical methods used in the different laboratories are described in details in Bordeleau et al. (2018a, 2018b & 2018c).

Results

Groundwater, which is generally potable in the region (based on metal, anion and VOC results), was classified into four water types according to the dominant cation and anion(s) in the samples. They are, in order of geochemical evolution: Ca-HCO_3, Na-HCO_3, Na-HCO_3-Cl and Na-Cl (Figure 7). There is no particular pattern in the spatial distribution of water types in the study area; instead, transitions occur vertically, and hence the major ion geochemistry is mostly related to sampling depth. Chemical evolution first occurs through ion exchange with the aquifer matrix, and additional salinity is gained through mixing with residual water of the Late Quaternary Champlain Sea water, which invaded this area between 13,000 and 11,000 years ago (Occhietti and Richard, 2003). While the bulk of the water in all water types is consistent with a meteoric origin (based on H_2O-$\delta^{2}H$ and -$\delta^{18}O$ values), the contribution of Champlain Sea residual water to salinity in the Na-HCO_3-Cl and Na-Cl water types throughout the study area is confirmed by a chloride to bromide (Cl/Br) molar ratio close to the seawater ratio of 639 (Hounslow, 1995). Another source of salinity that was identified in a few wells is formation brines, as even small amounts of these saline brines cause a high salinity in the samples, accompanied by a Cl/Br ratio well below that of seawater.

Samples with clear indication of deep formation brines were found to be located mainly close to (F5, F7, Zone 10R, Zone 11R2) or downstream (F1, F20, INRS-447) of the normal fault zone (Figure 7), suggesting that it acts as a discharge zone for deep regional groundwater flow (Bordeleau et al., 2018a, and section 7.1). Analyses of radioisotopes in samples from the bottom of wells F7 and F20 confirmed the very old apparent (bulk) age of this water, in the order of nearly 2 million years (based on ^{36}Cl values), with no contribution (F7) or very little contribution (F20) of modern recharge (based on ^{3}H and DIC-^{14}C values). Groundwater radon analyses (^{222}Rn) did not show any abnormally high values in the study area (the highest being 24 Bq/L), but the highest values were found close to the normal fault zone or downstream of it (Bordeleau et al., 2018c, and Figure S1 of the Supplementary Information section).

Dissolved methane was found to be ubiquitous in the study area, being detected in 96% of the sampled wells. The median concentration for all visited wells was 4.9 mg/L, with values in individual samples ranging from below detection limit (<0.006 mg/L) to 82 mg/L (Bordeleau et al., 2018a). The large spatial variations cannot be explained by differences in bedrock geology (Figure 7), but are strongly correlated to the water type and by extension, to the sampling depth and groundwater age (Bordeleau et al., 2018a). Higher methane concentrations were found in Na-rich evolved water, an observation in agreement with many other studies (Molofsky et al., 2013 and 2016; Darrah et al., 2014; Moritz et al., 2015; LeDoux et al., 2016; Siegel et al., 2015). A few wells also had significant concentrations of ethane and propane, which is an indication of the presence of thermogenic gas, since microbes can only produce very small amounts of these two molecules (Tazaz et al., 2013).

The origin of the natural gas found in groundwater can be ascertained from various geochemical graphs. The commonly-used Bernard graph (Bernard et al., 1976) showing the dryness ratio ($C_1/[C_2+C_3]$) versus methane carbon isotope values is presented in Figure 8. All individual samples collected in all wells are represented; for those with no ethane and propane, a dryness ratio cannot be computed, but was assigned an arbitrary value of 100 000 (at the top of the graphs, in the "unquantified ratios" box). Results from most samples fall outside of the traditional domains for microbial and thermogenic gas (Figure 8-A). This could be due to mixing between microbial and thermogenic gas, or to biogeochemical processes affecting the isotopic composition of microbial gas.

Mixing with thermogenic gas was confirmed in 15% of the wells, based on the presence of ethane and/or propane (Figure 8-B). These wells are not clustered in a specific area or located specifically along fault zones (Figure 7, see red circles). However, the only groundwater sample that falls within the 'thermogenic' domain on Figure 8 (shown with a red arrow), was collected in a well located along the RJC fault. The CH_4-$\delta^{13}C$ value of this sample corresponds to deep formation gas from the Lorraine Group, but not to the Utica Shale (values published in Chatellier et al., 2013). This residential well was sampled several times afterward, but all subsequent samples contained only microbial gas.

For samples without ethane or propane, other processes must be invoked to explain ambiguous CH_4-$\delta^{13}C$ values. The process that is most often suspected is oxidation, which causes an increase in both CH_4-$\delta^{13}C$ and -δ^2H values. Considering the CH_4-δ^2H values in our samples, and based on published isotopic fractionation factors for methane oxidation, this process does not appear to be significant in the Saint-Édouard area, except for Ca-HCO_3 type samples with very low methane concentrations and relatively high CH_4-$\delta^{13}C$

values (pink diamonds on Figure 8-A). Moreover, comparison of CH_4-δ^2H and H_2O-δ^2H values confirmed that methane isotopic values in most samples are consistent with regular (non-oxidized) microbial methane formed via the CO_2 reduction pathway using the local groundwater (Bordeleau et al., 2018b).

Another process that may explain ambiguous isotopic values is late-stage methanogenesis, which is the result of kinetic isotope effects occurring during microbial transformation of substrates (mostly CO_2 in this region) into methane, in an isolated groundwater reservoir where the carbon (reactant) pool has substantially been used up and is not replenished by fresh DIC from active recharge (Whiticar et al., 1999). Gas produced through this process is characterized by high dryness ratios typical of microbial methane, relatively high CH_4-$\delta^{13}C$ values resembling thermogenic gas, and higher than expected DIC-$\delta^{13}C$ values. An in-depth interpretation of these geochemical parameters (Bordeleau et al., 2018b) revealed that many samples contained late-stage microbial gas (Figure 8-C). Noteworthy, several of these samples also contain some thermogenic gas (Figure 8-B, C). Analyses of methane radiocarbon (CH_4-^{14}C), and comparison with DIC-^{14}C values in the aquifer, confirmed that the late-stage microbial gas was produced in the distant past (^{14}C-free), while the regular microbial produced at shallower depths in the aquifer tends to be more recent (^{14}C-bearing).

Finally, stable isotopic composition of ethane and propane, when available, did not provide additional information on the possibility of upward migration, as the values measured in groundwater, shallow bedrock gas and deep formation gas (Lorraine and Utica shales) span similar ranges (Bordeleau et al., 2018b).

6.2 Rock geochemistry

Objective

As 15% of the sampled wells contained dissolved thermogenic methane, it was necessary to determine the origin of that methane. If methane had been found to originate from the Utica Shale, it would have implied the presence of natural fluid migration pathways from the shale gas reservoir to the shallow groundwater. Therefore, initial organic geochemical analyses were carried out on shallow core samples to document gas characteristics in the upper 150 m, mainly composed of black shale units. The purpose of these analyses was mainly to provide the concentrations and isotopic ($\delta^{13}C$ and $\delta^{2}H$) compositions of alkanes (C_1-C_3) that were shown to be present in shallow bedrock and compare these values to those of dissolved natural gas in groundwater.

Fieldwork

Drill cuttings and core samples were collected during the drilling campaigns. Both were stored in Isojars® (Isotech Laboratories, Champlain, IL) containing ultrapure water and a bactericide during the 2014 campaign. A triplicate of Isojars® were collected approximately every 10 to 15 m, leading to a total number of samples per well varying from 3 to 10. Gas composition was analysed for 39 samples. Some core samples were also preserved in a double-layer vacuum plastic bag for pore-water analyses and some were disinfected and stored in aluminium foil, then frozen, for phospholipid fatty acid (PLFA) analyses. Analyses of the last two types of samples did not provide the expected results; they are thus only briefly discussed in the Supplementary information section.

Methods

Drill cuttings and core samples preserved in the Isojar® were initially analysed by Rock-Eval to assess the presence of pore-space free and adsorbed hydrocarbons as well as their thermal maturation. The latter was also evaluated through organic petrography of the samples. Gas extracted from drill cuttings and core samples (accumulated in the Isojar® headspace) was analysed for both alkane concentrations and isotopic composition. Therefore, the complete set of analyses included: Rock-Eval pyrolysis, petrographic observation of organic matter reflectance, alkane concentrations (C_1 to C_3 or C_1 to C_5 depending on the laboratory), alkane isotopic composition (C_1 to C_3) for $\delta^{13}C$ and sometimes δ^2H (depending on the laboratory), and ^{14}C of methane. Methods used in laboratories for the different analyses are described in Lavoie et al. (2016) and Bordeleau et al. (2018b).

Results

All rock samples from the Les Fonds, Lotbinière and Nicolet formations showed organic matter typical of Type II kerogen, the three units are thermally mature and have reached oil window conditions at the time of maximum burial. The Lotbinière and Les Fonds formations have total organic carbon (TOC) content of a fair hydrocarbon source rock (TOC>0.5%), whereas the Nicolet Formation has a lower TOC content and is considered as a poor hydrocarbon source rock (Lavoie et al., 2016).

All shallow bedrock samples contained gas hydrocarbons, but with locally variable composition and concentration that were not always linked to specific bedrock units. Over the depth range covered by samples, a downward increase in the concentration of alkanes ($C_1+C_2+C_3$) was observed, often along with a decrease in the gas dryness ratio ($C_1/[C_2+C_3]$)

(Lavoie et al., 2016). In fact, samples with dryness ratios <100 (thermogenic gas) were found along the whole depth range, but samples with ratios between 100 and 1000 (mixed gas) were restricted to the top 50 m of bedrock. Likewise, samples with CH_4-$\delta^{13}C$ values corresponding to thermogenic gas (>-50‰) or mixed gas (-60 to -50‰) were found over the whole depth range, while samples with microbial values (<-60‰) were generally restricted to the top 15 m of bedrock. This suggests that in the top part of the fractured rock aquifer, the thermogenic gas that was originally present in the bedrock pores has escaped and/or was affected by microbial degradation, while ongoing *in situ* methanogenesis in this active part of the aquifer adds microbial gas.

Isotopic results and dryness ratios for shallow bedrock samples appear on Figure 8 (grey diamonds), along with deep formation gas (>600 m) from the Lorraine and Utica shales (black diamonds) published by Chatellier et al. (2013). The CH_4-$\delta^{13}C$ ratios of shallow bedrock gas have limited overlap with those of the deep formation gas, which typically show isotopically-enriched values. The presence of unambiguously thermogenic gas in some shallow bedrock samples, and the mix of microbial and thermogenic gas in others, is confirmed by a Whiticar (1999) plot of CH_4-$\delta^{13}C$ versus CH_4-$\delta^{2}H$ values (see Lavoie et al., 2016). As mentioned in section 6.1, the $\delta^{13}C$ values of C_2 and C_3 alkanes are indistinguishable from values for the deep formation gas samples (Lavoie et al., 2016).

The CH_4-$\delta^{13}C$ ratios of shallow rock samples overlap with groundwater results for a large part, but more thermogenic gas was found in rock (Figure 8), confirming that thermogenic gas is trapped in rock pores, while recent microbial gas is constantly forming and dissolving in groundwater. A comparison of CH_4-$\delta^{13}C$ and –$\delta^{2}H$ values for shallow bedrock and groundwater was also made on a well by well basis, showing that values were very similar for a given well and sampling depth (Bordeleau et al., 2018b). Based on these findings, the likely source of thermogenic gas in groundwater appears to be the shallow fractured rock aquifer itself (which is mainly composed of organic-rich black shale), rather than the deep Utica Shale (Lavoie et al., 2016).

6.3 Groundwater monitoring

Objective

The objective of monitoring dissolved methane concentration and its isotopic composition was to document natural variations in wells with different characteristics. Knowledge of these temporal variations for a given area and even for a given well prior to any shale gas development is critical, mainly to distinguish natural fluctuations from anthropogenic impacts stemming from deep industrial activities. Until now, few studies have documented such variations (e.g.: Coleman and McElreath, 2012; Gorody, 2012; Humez et al., 2015; Sherwood et al., 2016; Smith et al., 2016; Currell et al., 2017, Botner et al., 2018) and they have relied on only a few wells and/or on a few sampling events.

Fieldwork and methods

Monitoring was carried out over more than two years in six wells, including four observation wells (F1 to F4) and two residential wells (INRS-447 and Zone 9R). Well F21, which was drilled later in the project, was subsequently added to the monitoring program and was sampled over 15 months. The selected wells reflect the diversity of wells present in this study area (e.g.: high and low methane concentrations, observation and residential wells, shallower and deeper wells, purely microbial gas and mixed gas). Groundwater was sampled for alkane (C_1-C_3) concentrations, methane isotopic composition (CH_4-$\delta^{13}C$ and –δ^2H), and DIC-$\delta^{13}C$ analyses. Groundwater sampling was carried out as described in section 6.1.

Results

Results showed that dissolved methane concentrations can fluctuate greatly even in the absence of industrial activities, depending on the sources of gas and microbial activity. Some examples from monitoring wells are presented in Figure9. For the Saint-Édouard area and for the monitored period, methane concentrations varied from 2.5 to 6 times the lowest recorded values for a given well (with sampling depth and sampling technique remaining the same), which is well above the uncertainty expected for sampling, handling and analysis (Rivard et al., 2018a). The gas dryness ratio (not shown here) also varied significantly over time for a given well, but in general it did not affect the interpretation, as values tended to remain within the same "class" (<100 for thermogenic gas, >1000 for microbial gas).

In contrast, isotopic values (CH_4-$\delta^{13}C$ and -δ^2H) were generally very stable over time, with variations remaining within the uncertainty expected for sampling, handling and analysis (Rivard et al., 2018a). However, the two wells with the lowest methane concentrations (Zone 9R and F3) showed significant variations over time (Figure 9). Changes in isotopic composition due to either mixing of gas sources in varying proportions, or to post-genetic processes affecting methane, are likely to have a more noticeable effect when methane concentrations are small. In well F3, the variations were attributed to oxidation (Rivard et al., 2018a) while in Zone 9R, the main factor is the detection of thermogenic gas with a relatively deep formation gas signature in the first sample of the series (Bordeleau et al., 2018b). DIC-$\delta^{13}C$ values were also found to vary significantly over time (not shown here), especially in wells where methane concentrations were high and where late-stage methanogenesis was predominant (Rivard et al., 2018a).

Therefore, while isotopic values of methane are usually a good and stable indicator of gas origin in a well, significant variations may occur in some wells and thus monitoring for both concentration and isotopic values should be carried out for a sufficient period ahead of any shale gas activities to establish natural variability. Moreover, monitoring of DIC-$\delta^{13}C$ may prove very helpful in interpreting the origin of methane, especially when CH_4-$\delta^{13}C$ values are ambiguous. Most importantly, interpretation of gas origin based on a single sample from a well could be erroneous if that sample is a punctual anomaly, as was observed on a few occasions in this study. These results could therefore have important implications on regulatory or voluntary procedures aiming to define the natural baseline presence of methane prior to shale gas activities.

7 Importance of multi-source and multi-discipline data

The complex scientific issues associated with the study of potential upward fluid migration from hydrocarbon reservoirs to shallow aquifers ideally require multisource data and multidisciplinary expertise. Within the Saint-Édouard project, the approach used integrated data from and expertise in structural geology, stratigraphy, hydrogeology, geophysics, geomechanics and organic/inorganic geochemistry, which together indicated key elements to better understand the relations between shallow and deep earth systems. This multidisciplinary approach involved many research scientists and required intensive fieldwork and numerous laboratory analyses. Moreover, the access to industry data appears to be essential for this type of project, since the cost of data from dedicated deep wells would be prohibitive. The combination of these data allowed an assessment of the integrity of the intermediate zone (presence or absence of fluid migration pathways) and characterization of natural gas in shallow groundwater.

7.1 Integrity of the intermediate zone

At first, the geological interpretation from industry deep seismic data provided indications that fluid pathways linking the shale gas reservoir to shallow aquifers could possibly be present, especially in the vicinity of the RJC normal fault (Lavoie et al., 2016). Unfortunately, the shallow seismic survey, which was expected to provide key information on the presence or absence of faults in the near surface (and if present, their precise location), did not work well in this region. However, the HTEM survey (see Supplementary information section) provided further indications of the presence of an anomalous feature in the first few hundred meters, in the vicinity of this normal fault. Although firm conclusions could not be drawn, ^{222}Rn in soil gas and groundwater (see also Supplementary Information section) indicated possibilities of active upward migration in fault zones, where slightly higher values were found compared to areas away from faults (Bordeleau et al., 2018c). Therefore, these early surveys justified the need to drill observation wells in the vicinity of fault zones to provide more focused data close to these geological features.

On the other hand, initial geological and hydrogeological data provided indications against the presence of a fluid migration pathway along the two regional fault zones. The piezometric map seemed to indicate that groundwater flow was merely occurring from the Appalachian uplands all the way towards the St. Lawrence River, following topography. Therefore, the St. Lawrence River had initially been assumed to correspond to the regional groundwater discharge zone. Also, previous publications on the geological context of this area had reported lines of evidence of fault sealing based on overpressured conditions and low water saturation in the Utica Shale, as well as unbalanced fluid pressure on both sides of the Yamaska fault, which extends northeast into the RJC fault present in our study area (Chatellier et al., 2013; Séjourné et al., 2013; Konstantinovskaya et al., 2009). Furthermore, clay gouge was observed in cores drilled in the parautochthonous domain to the south and is suspected to be present in the RJC normal fault due to the presence of similar shale units to the north and because clay gouge was also observed in at least two deep wells drilled into the Yamaska fault zone south-west to the study area (Séjourné et al., 2013), supporting a lack of significant circulation across the fault.

The integration of geophysical (shallow and deep borehole logging), geomechanical, geological (structure and stratigraphy) and hydrogeological data allowed the characterization of the fracture network throughout the entire sedimentary succession and provided a first estimate of hydraulic properties for both the shallow and deep intervals and, by extension, for the intermediate zone (Ladevèze et al., 2018b). Open natural fractures in the reservoir (and thus in the intermediate zone) are mainly from one set (FS1).

Due to their single orientation, these open fractures are thus poorly interconnected, which does not favour permeability nor flow towards the surface, even if a higher open fracture density was observed in the vicinity of thrust faults. Ladevèze et al. (2018b) concluded that the possibility for the fracture network or fault zones to act as large-scale flow pathways was very unlikely. Furthermore, the geomechanical stratigraphy (Séjourné, 2017) provided clear evidence against any significant upward extension of hydraulically-induced fractures from the brittle Utica Shale into the more ductile Lorraine Group. These findings are in agreement with the reported overpressured conditions of the Utica Shale (Chatellier et al., 2013) that indicate a lack of significant natural fluid connectivity with the surface.

Groundwater geochemical data provided evidence that upward migration of saline fluids is occurring in the normal fault zone over a distance in the order of a couple hundreds of meters (likely 200 to 500 m), but not from as deep as the Utica Shale. Also, the first sample from well Zone 9R, which had a thermogenic signature, appeared to have a CH_4-$\delta^{13}C$ value that is consistent with data from deep shales of the Lorraine Group and not of the Utica Shale (Bordeleau et al., 2018b). While geological and hydrogeological data alone would not have detected a fluid migration pathway along the normal fault, geology did provide a working hypothesis to explain the local increase of permeability compared to the regional-dominant shale host rock in this area (entrainment of porous and more fractured siltstone beds into the fault core in the upper part of the intermediate zone), which would allow the upward migration of deeper brines.

Based on the ensemble of data just mentioned, Figure 10 presents the conceptual model for regional groundwater flow occurring in shallow aquifers and the upper portion of the intermediate zone. This conceptual model infers that some of the water recharging in the

Appalachians would circulate towards the northwest at a certain depth in the intermediate zone (between 200 and 500 m), where it would gain salinity while flowing within the intermediate zone. In the thrust fault zone of the parautochtonous domain, it is believed that a small portion of the flow may be able to pass through at different depths in the more or less tectonized and fractured thrust slices. This water would then discharge upward in the more porous and fractured damage zone of the RJC normal fault, where siltstone beds have likely been dragged. Numerical simulations carried out by Janos et al. (2018) showed the hydraulic plausibility of this inferred impact of the RJC fault on regional groundwater flow. Once groundwater reaches the upper, more fractured part of the aquifer, it may cross the core of the fault and flow downstream (to the northwest), following the general flow direction towards the St. Lawrence River. The airborne (HTEM) survey, fracture data from the borehole logging, and geochemical profiles confirmed that the active groundwater flow zone is in the order of 60 m deep, below which the intermediate zone begins.

A summary of the arguments in favor or against the presence of migration pathways, obtained from each scientific discipline, is presented in Supplementary Table S-2.

7.2 Methane baseline study

One of the main objectives of this study was to provide a baseline characterization of methane and higher alkanes in shallow groundwater of the Saint-Édouard area, before any shale gas exploitation occurred. In this respect, the key element of this project was the combination of geochemical data from: 1) the industry (concerning deep-seated fluids from the Lorraine Group and Utica Shale), 2) shallow groundwater from both residential and dedicated observation wells, and 3) shallow bedrock cores and cuttings from observation wells. A wide suite of geochemical, and particularly isotopic parameters provided indications about the source (microbial versus thermogenic) and origin (shallow depths or deep units) of dissolved methane in the aquifer, as well as the processes affecting methane. The processes that affect microbial methane in this region (late-stage methanogenesis, mixing with thermogenic gas and, to a lesser extent, oxidation) originally resulted in ambiguous interpretation of thermogenic versus microbial origins, but the use of multiple lines of evidence helped shed light on the complex history of methane production in the study area.

This multi-isotope approach led to the conclusion that both microbial and thermogenic methane comes from shallow fractured rock, which is mainly composed of organic-rich black shales. In Late Ordovician, the latter units were tectonically and stratigraphically buried under at least 4 km of Paleozoic strata, which provided conditions that allowed the production of thermogenic methane (Dietrich et al., 2011), before erosion resulted in the modern landscape. Today, shallow units also have the necessary reducing conditions (oxygen- and sulfate-free) to support microbial methanogenesis. Analyses of shallow bedrock cores and cuttings, as well as groundwater monitoring over time, demonstrated that thermogenic gas and old microbial gas produced in the distant past, remain trapped in shale pores, and are rapidly released during and shortly after well drilling, as well as very slowly afterwards (Lavoie et al., 2016; Rivard et al., 2018a). In addition, recent microbial gas is formed in groundwater, which may locally undergo oxidation.

7.3 Knowledge gaps and recommendations for future studies

The combination of data from multiple disciplines allowed the development of geological and hydrogeological conceptual models integrating all depths of interest (shallow aquifer, intermediate zone and deep industry-targeted shale), and thus provided a full evaluation of the possibility for upward fluid migration from the Utica Shale to the shallow aquifer. As the intermediate zone was very poorly characterized, the interpretation of the intermediate zone integrity relied on indirect data. To address the remaining unknowns to carry out a quantitative assessment of the potential environmental risks of upward migration, drilling of research observation wells in the intermediate zone would be necessary, as these wells would provide key information, such as the depth to which upward migration occurs and actual permeabilities, especially near fault zones. Deep monitoring wells are indeed considered essential for the development of fundamental geoscience by several authors (Jackson et al., 2013b; Soeder, 2015; Ryan et al., 2015). Despite these recommendations, these expensive wells are still too few.

Also, the Council of Canadian Academies report (CCA, 2014) on environmental impacts of shale gas extraction in Canada stressed that appropriate environmental monitoring approaches have not yet been designed and that monitoring needs to engage independent experts to gain public trust. The development of a sound methodological approach for the assessment of shallow aquifer vulnerability to deep industrial activities is the prerequisite for many jurisdictions to get the social licence to safely develop a shale play. As a first step in this direction, and while we are conscious that the local geological and hydrogeological context has a major impact on the results, Table 1 provides recommendations for future projects. Although many of the scientific activities were carried out in an effort to see what could work best for this project, almost all of them ended up being rated high, as each provided pieces of information that, once put together, allowed a better understanding of the system. Table 1 can obviously be adapted according to the available equipment and expertise available to conduct a project. Finally, to provide a more hands-on example of the usefulness of each activity, Supplementary Table S-2 presents a summary of new knowledge generated by this research focussing on its significance for or against the possibility of large-scale migration in the Saint-Édouard area.

Table 1: List of recommended activities, along with their requirement, importance and potential contribution

Activity	Requirement	Importance / potential contribution
Re-processing and interpretation of deep seismic surveys	Have access to industry data.	High / Could provide preliminary information on potential upward migration, especially close to regional fault zones.
HTEM survey and interpretation	Have significant funds. Have a nearly undeformed geological setting: complex geology (e.g. folded rocks) will probably not allow good results. The transect should only cross a minimum number of municipalities and electromagnetic features (e.g., electric lines), otherwise data are unusable in these areas.	Moderate / Could provide a regional shallow survey of the fresh-saline water interface that may not be obtained otherwise. Other data providing indications about lithology or fluid content (e.g. geophysical logs) would allow the verification of the inversion and of its interpretation.
Shallow seismic survey	Have significant funds or have access to in-house equipment and expertise. Make preliminary tests in the study area to see if usable and conclusive to image the first 500 m.	High / Could complement the deep seismic survey to see if faults extend to the surface (or close to).

Geomechanical interpretation	Have access to borehole logging data and core samples from deep oil and gas wells and substantial budget if no expertise among stakeholders.	High / Could provide geomechanical properties that allows a better understanding of the behavior of induced fractures into the IZ in case of hydraulic fracturing and of specific units (e.g. shales of the Utica Shale and of the Lorraine Group in this case).
Drilling of observation wells	Have a substantial budget.	Very high / Main interest of having observation wells is to be able to perform all kinds of measurements (e.g. borehole logging, hydraulic tests, sampling at specific depths, geochemical profiling). Observation wells can be complemented by residential wells for a better spatial coverage and to see if they react similarly over time.
Characterization of the fracture network	Have access to borehole logging data from shallow and deep wells (and, if possible, intermediate depths)	High / Obtain information on fracture interconnection and permeability over the entire succession to assess risk of upward migration. Since few data are usually available in the IZ and this zone controls shallow aquifer vulnerability to deep activities, it is important to characterize intervals for which data are available and try to see if the results are applicable to the IZ.
Characterization of hydraulic properties	Complete a fracture network characterization from the gas reservoir to the surface based on shallow and deep	High / As the objective of the project is to study the potential for upward fluid migration, hydraulic

	well logging data and visited outcrops. Perform hydraulic tests in shallow wells. Have access to fracture aperture data from the industry or be able to estimate them from available data. Ideally, have access to pressure values at different depths to estimate the vertical hydraulic gradient and to hydraulic properties (e.g. drill-stem tests) from the industry.	properties within the different intervals, including the IZ, is also very important. Estimates from data apertures and frequency of open fractures provide best guests for porosity and permeability at depth if the latter are not already available. *In situ* hydraulic tests at depth would provide key data, necessary to carry out a quantitative assessment of shallow aquifer vulnerability to deep industrial activities.
Groundwater geochemistry	A moderate budget is needed for the baseline study, both for human resources and laboratory analyses. The latter would include, at the very least, major ions and metals, and alkane concentrations and -$\delta^{13}C$. To allow a thorough interpretation, alkane-$\delta^{2}H$ and DIC-$\delta^{13}C$ are also highly recommended. If possible, DIC-^{14}C, ^{3}H, and H_2O-$\delta^{2}H$ and –$\delta^{18}O$ are also recommended. Other, less traditional indicators have proven to be useful in this study, such as CH_4-^{14}C and ^{36}Cl. A good spatial coverage, including a finer distribution in the vicinity of regional fault zones, are ideal. A combination of observation wells that can be	Very High / GW geochemistry allows the indirect verification of possible upward fluid migration, given that sampled wells are adequately located.

	thoroughly investigated and residential wells that increase the density of spatial coverage is recommended.	
Rock geochemistry	Have access to drill cuttings or core samples (some placed in Isojars® for hydrocarbon composition analyses), and well preserved deep core samples or gas analyses from the industry for comparison.	Useful only if hydrocarbons are detected in GW. In this case, it rates High / Essential if one wants to make a comparison between GW and rock organic geochemistry to provide evidence that gas comes from the shallow aquifer itself, or from deep formations.
Soil geochemistry (see Supplementary Information section)	Useful only if hydrocarbons are present in GW. A thin, permeable surficial sediment cover may provide clearer indications of potential fluid migration.	Moderate / Could provide preliminary information on potential upward migration, especially close to regional fault zone.
Groundwater monitoring	Moderate budget and time is required for regular sampling campaigns. GW monitoring should be carried before, during and after shale gas (or any deep industrial) activities; it should include alkane concentrations and isotopic composition, as both can vary over time and these fluctuations could be misleading once oil and gas activities start. Monitoring of DIC-$\delta^{13}C$ can also provide important information	Very high / Monitoring data will provide reliable data in case a complaint is logged against the industry. As more data will be collected, this will help develop appropriate regulations for given areas. Such monitoring provides indications of the natural variability of methane concentrations and isotopic composition, which may help provide firmer indications

	on microbial processes affecting methane. Monitoring prior to any oil and gas activities should be carried out for at least a year, two years if possible.	of impacts on groundwater from industrial subsurface activities.
Use of downhole gas sensors (see SI section for work carried out)	Have a substantial budget or develop in-house sensors (or collaboration with people doing research in this field).	While these sensors would be necessary to collect shorter-term data on methane concentrations, it is probably better to wait until technology and methodology is ready.
Pore-water analyses from shallow and deep shale samples	Have access to this type of analyses (exclusively carried out in a few universities worldwide). Core samples must not be too tight (potential for these analyses is severely limited in tight shales).	Low / Could provide information on brines and hydrocarbons in pores at different depths.

8 Conclusion

Knowledge of potential impacts of deep unconventional reservoir development on shallow aquifers is a major issue for the hydrocarbon industry, mainly due to environmental concerns related to groundwater contamination, especially in regions where large-scale unconventional hydrocarbon activities have never taken place. This paper presented an overview of the work carried out during a 4-year multidisciplinary project on the assessment of shallow aquifer vulnerability to deep shale gas activities through upward fluid migration, in a region where only little exploration has occurred so far. The Saint-Édouard area, overlying the Utica Shale in eastern Canada (more specifically in the St. Lawrence Lowlands, province of Quebec), had been selected notably because the shale gas exploration well drilled in this locality was the most promising well in the St. Lawrence Lowlands, and importantly, it was possible to have access to data from the operator.

Potential links between deep geological units targeted by the industry and surficial aquifers are usually not well documented or understood. Aquifer protection thus strongly depends on a better geological and hydrogeological knowledge of these zones. As data have generally not been acquired in the past within the intermediate zone, scientific information must be gained through indirect data, ideally from multiple sources. In this project, extensive fieldwork was carried out and a wide range of data were collected and analysed including geological, geophysical, geomechanical, geochemical and hydrogeological data. These data were both interpreted individually and in combination with data from the other disciplines.

Results showed that groundwater in this area generally contains methane, sometimes in very high concentrations, and occasionally some ethane and propane. Of the 44 residential and observation wells that were sampled, 36% had methane concentrations above 7 mg/L, the alert threshold for the Department of Environment of Quebec. These concentrations, as well as the sources of methane and processes affecting it, were shown to be variable in space and/or time. Most methane in this region is of microbial origin, but thermogenic gas was found in 15% of the wells. Rock organic geochemistry showed that the shallow bedrock itself is the source of both types of gas found in groundwater. While evidence of local upward fluid migration of brines from the intermediate zone was found in the vicinity of a normal fault, aquifers of this area appear to be well protected against contamination from industrial activities carried out at depth, based on the acquired data and interpretation. Nonetheless, the interpretation inferred from the available data, combined to the lack of direct data in the intermediate zone, indicate that care should be taken should drilling of gas wells be resumed, and until new data specific to the fault zones are available, hydraulic fracturing should be avoided in their vicinity.

The specific results and conclusions presented here are valid only for the study area and should not be directly extended to other regions, even within the St. Lawrence Lowlands. Nonetheless, the methodology developed within this project and recommendations made for future similar projects should likely be useful to other regions where deep industrial activities are planned, and it is hoped that it will serve as a basis to help research scientists develop a more in-depth methodology. Overall, collaborative and integrated projects between the government, academia and industry are necessary to quantitatively assess impacts or risks for shallow fresh groundwater for such environmental studies.

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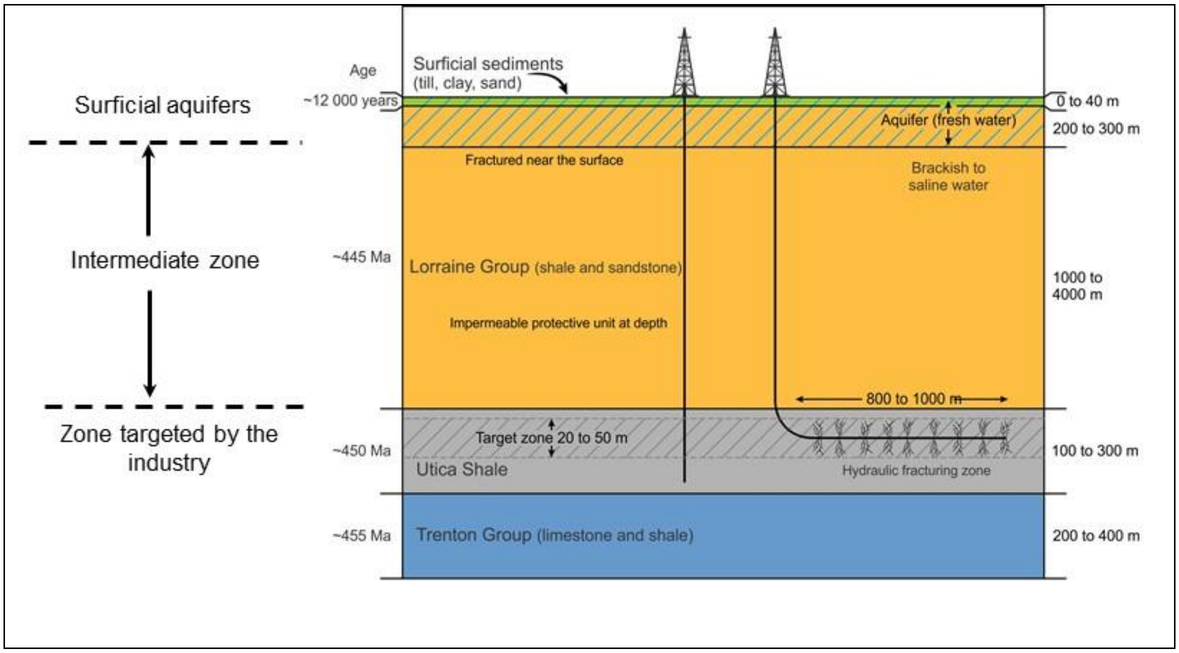

Figure 1. Schematic geological cross-section presenting the different zones referred to in this paper: shallow aquifers, intermediate zone and shale gas reservoir (in this case: the Utica Shale).

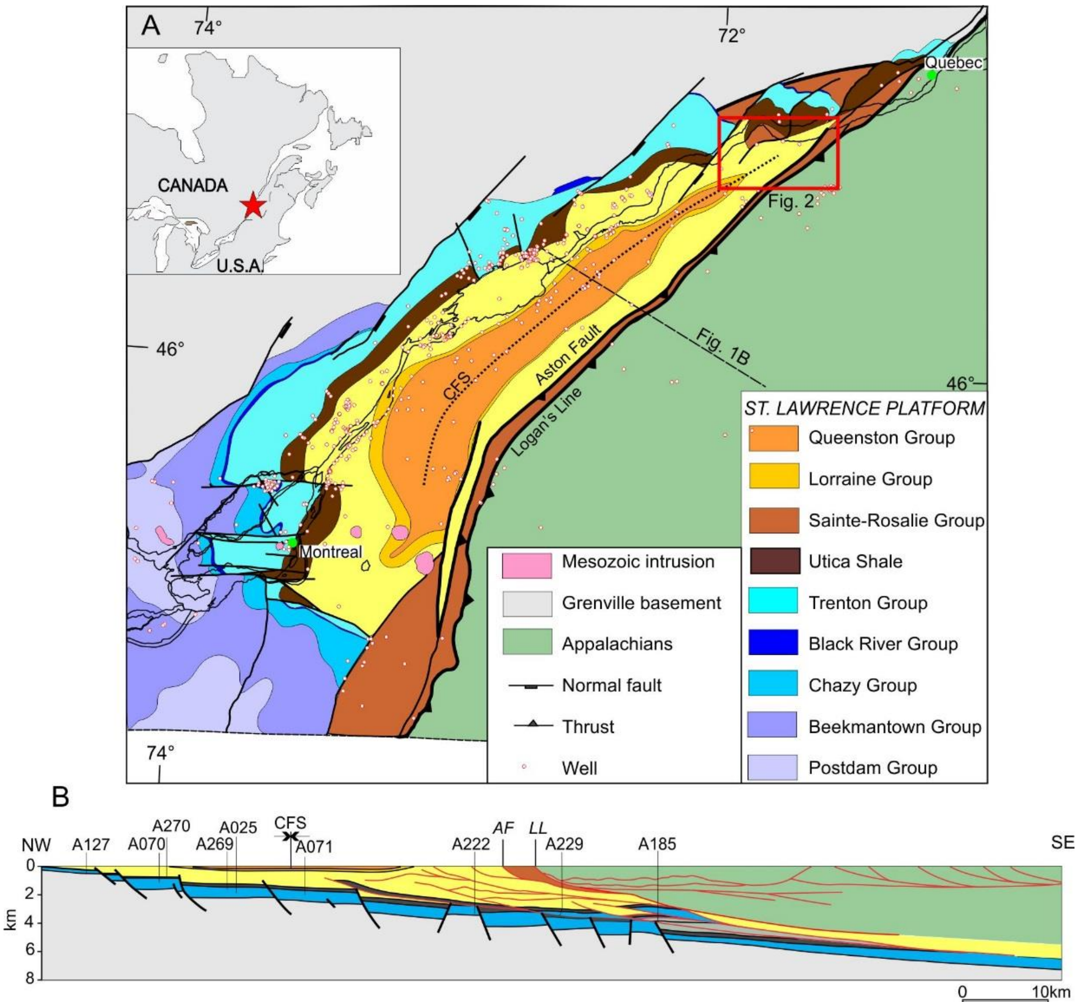

Figure 2. A) Geological map of the St. Lawrence Platform between Montreal and Quebec City, southern Quebec, Canada (modified from Globensky, 1987). The Saint-Édouard study area is shown with a red box. B) Cross-section presenting the general tectonostratigraphic framework (modified from Séjourné et al., 2013). CFS: Chambly-Fortierville Syncline; AF: Aston Fault; LL: Logan's Line.

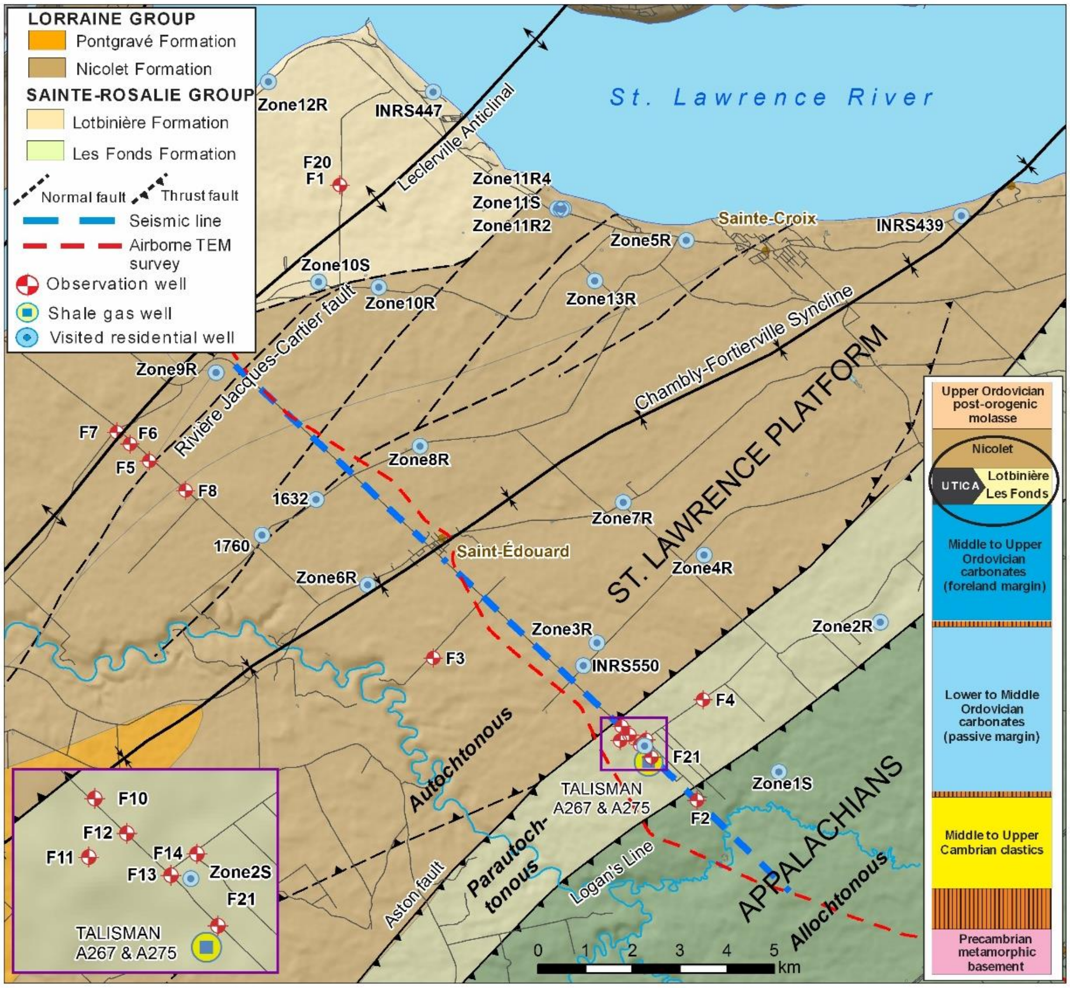

Figure 3. Location of the Saint-Édouard area (red box on Figure 2) and of the observation and residential wells, along with the stratigraphic column (adapted from Rivard et al., 2018b).

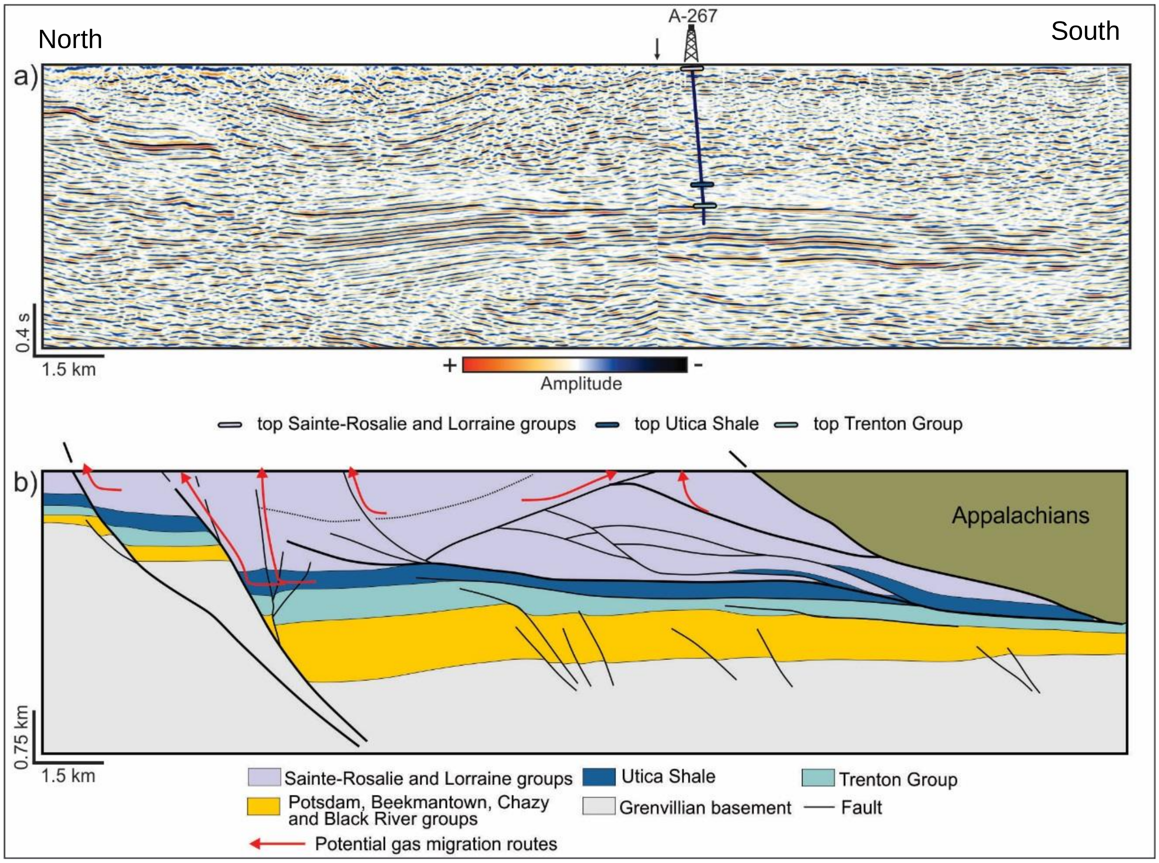

Figure 4. a) Uninterpreted industry seismic section crossing the Saint-Édouard wells (A-267 is the horizontal well) and b) geological interpretation of the seismic section shown in a) (modified from Lavoie et al., 2016). The arrow (at the top of the section on a) points at the location where two adjacent survey lines were merged together.

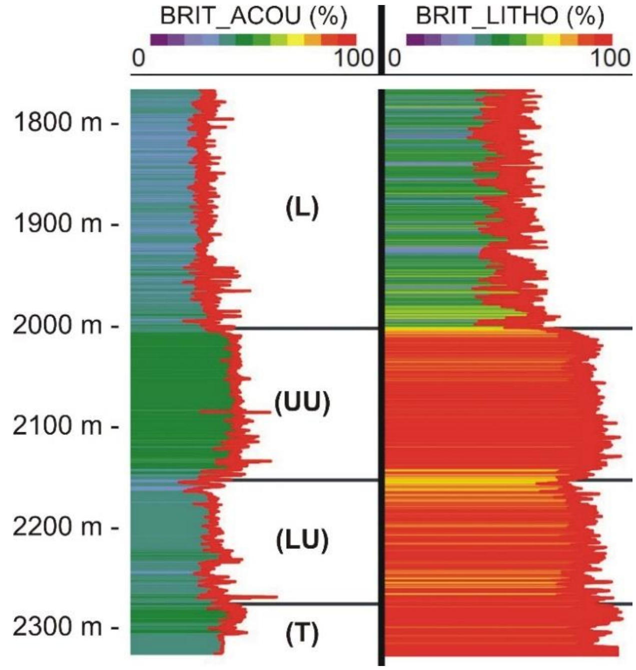

Figure 5. Geomechanical profile of the lower section of the Fortierville shale gas well illustrating the vertical variations in the acoustic brittleness index (BRIT_ACOU) and the mineralogical brittleness index (BRIT_LITHO) across the base of the Lorraine Groupe (L), the upper and the lower parts of the Utica Shale (respectively UU and LU) and limestones at the top of the Trenton Group (T). The higher the value of the index, the more brittle is the strata. Note that the two independently calculated indexes display similar trends, but with different absolute values, which is explained by the different nature of the physical properties considered (acoustic versus mineralogic). Modified from Séjourné (2017).

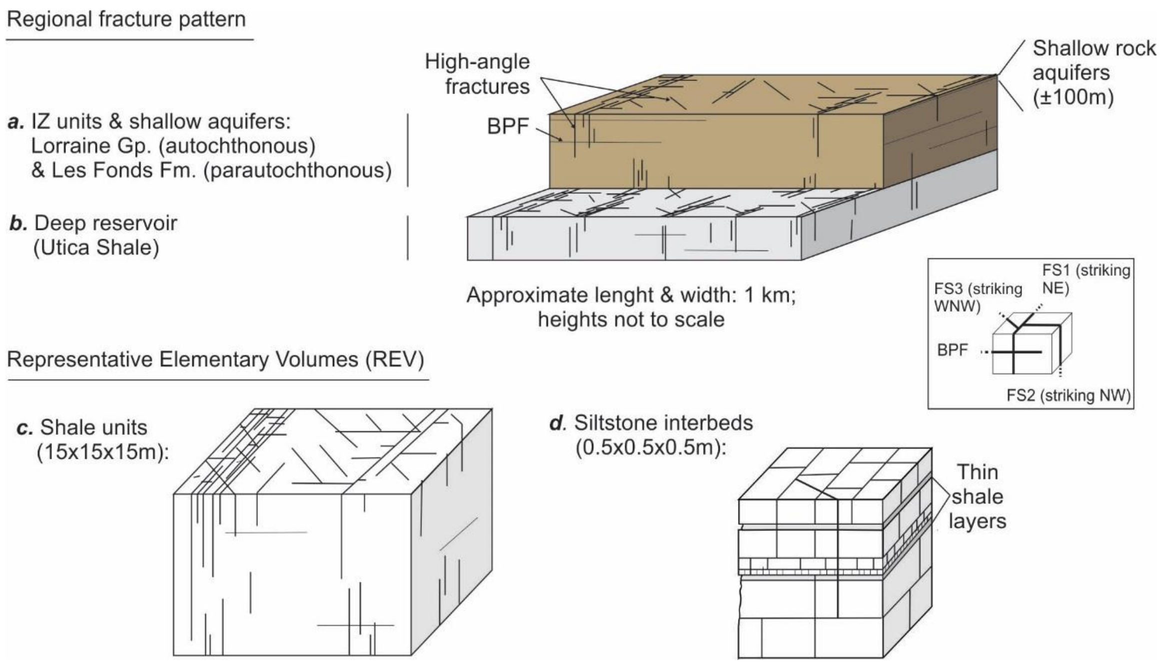

Figure 6. Conceptual models of the fracture network (modified from Ladevèze et al., 2018b). The regional fracture pattern is represented in **a**. for the shallow aquifers and intermediate zone (IZ) units; in **b**. for the deep reservoir. The fracture pattern is also represented using representative elementary volumes (REVs) at a much more local scale for: **c**. shale units and **d**. siltstone interbeds. BPF: Bedding parallel fracture.

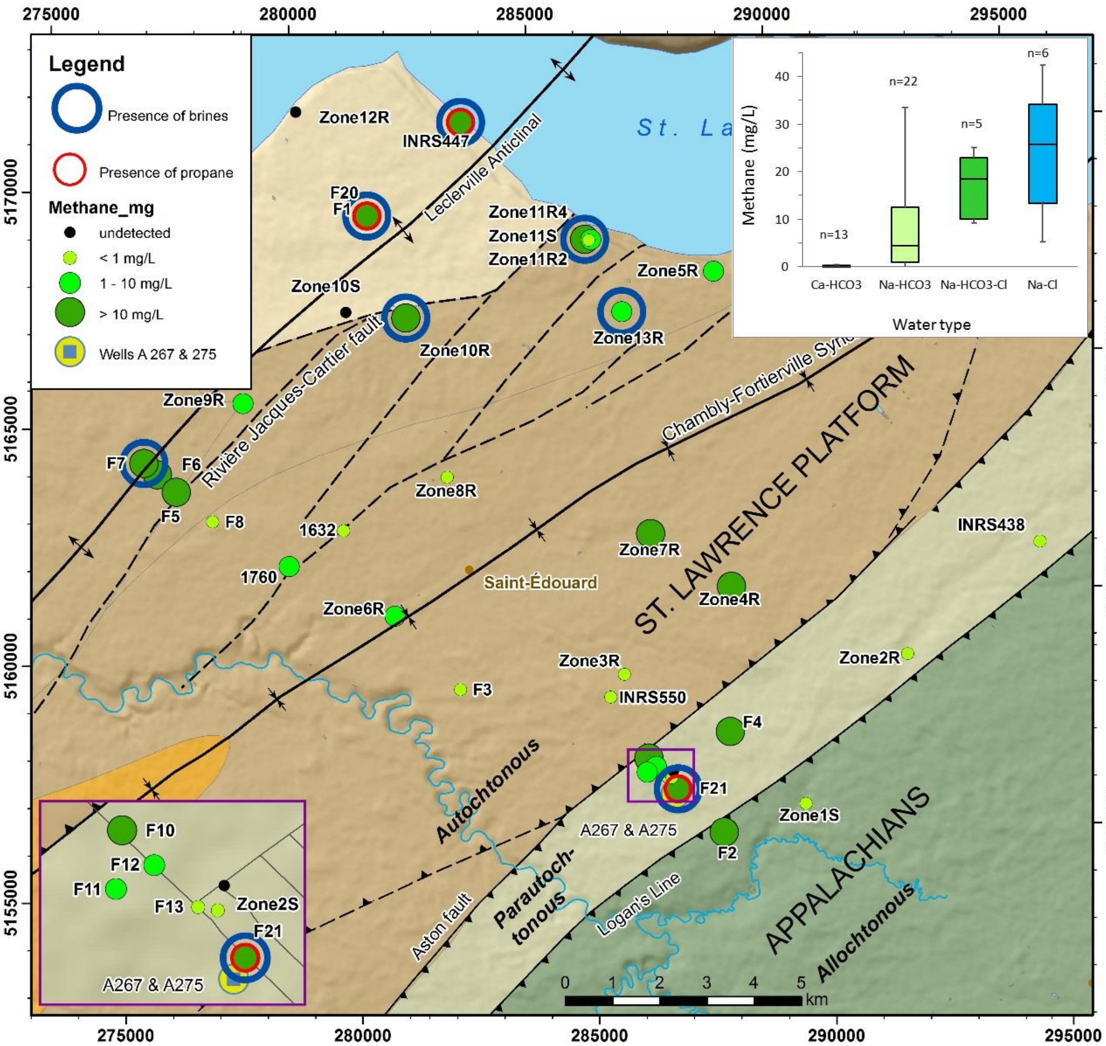

Figure 7. Spatial distribution of dissolved methane concentrations. Bedrock geology legend as in Figure 3. Inset: boxplot of methane concentrations as a function of water type.

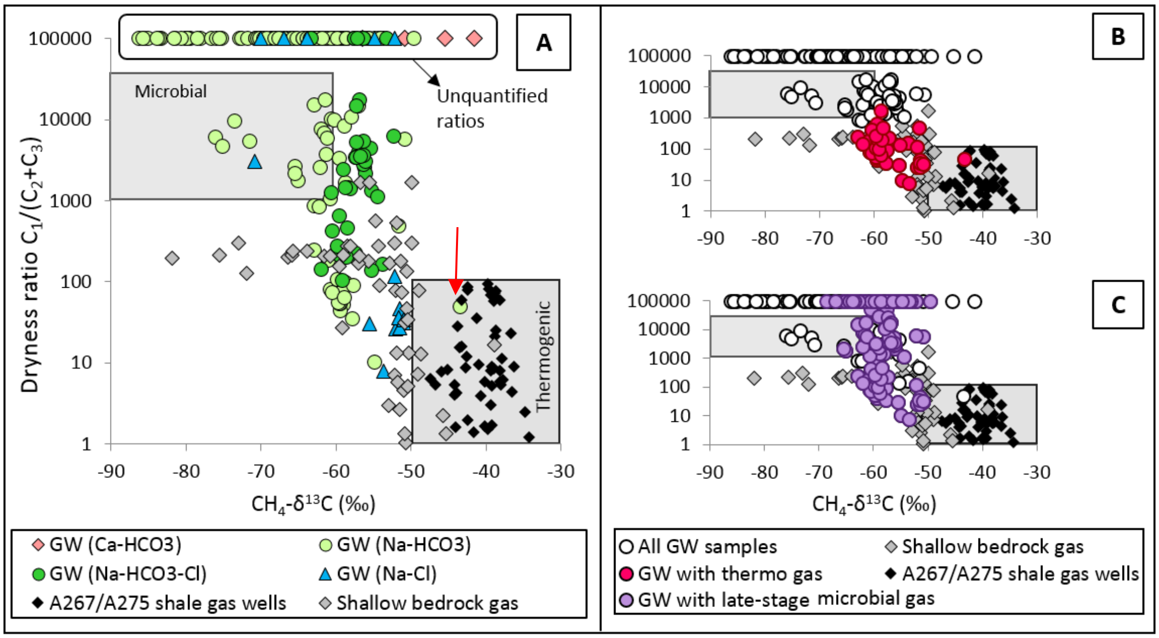

Figure 8. Bernard graphs showing the dryness ratio versus methane carbon isotopic composition obtained from shallow groundwater, shallow bedrock gas samples, and deep formation gas data published by Chatellier et al. (2013). A) Groundwater samples are classified according to their water type; B) emphasis on groundwater samples containing thermogenic gas and C) on groundwater samples containing late-stage microbial gas or microbial gas affected by oxidation (bottom). Gray boxes represent typical microbial and thermogenic gas domains.

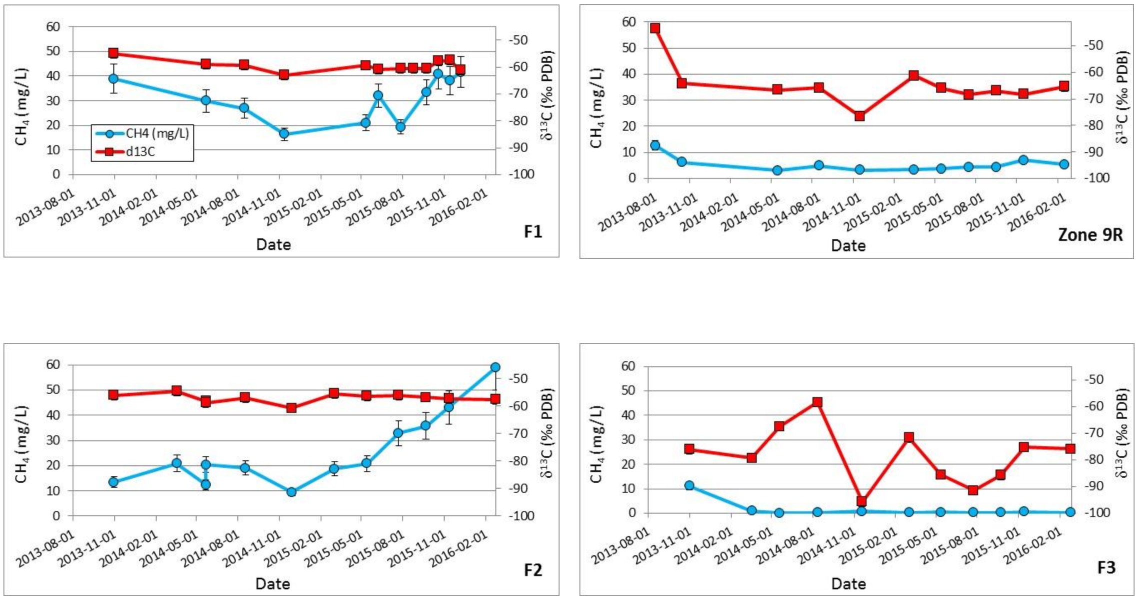

Figure 9. Examples of dissolved methane concentrations over time for four of the seven observation wells (blue), along with the methane carbon isotopic ($\delta^{13}C$) values (red). Uncertainty, represented by error bars, is ±15% of the values for concentrations and 1.7‰ for $\delta^{13}C$ (nearly impossible to see) (estimated in Rivard et al., 2018a).

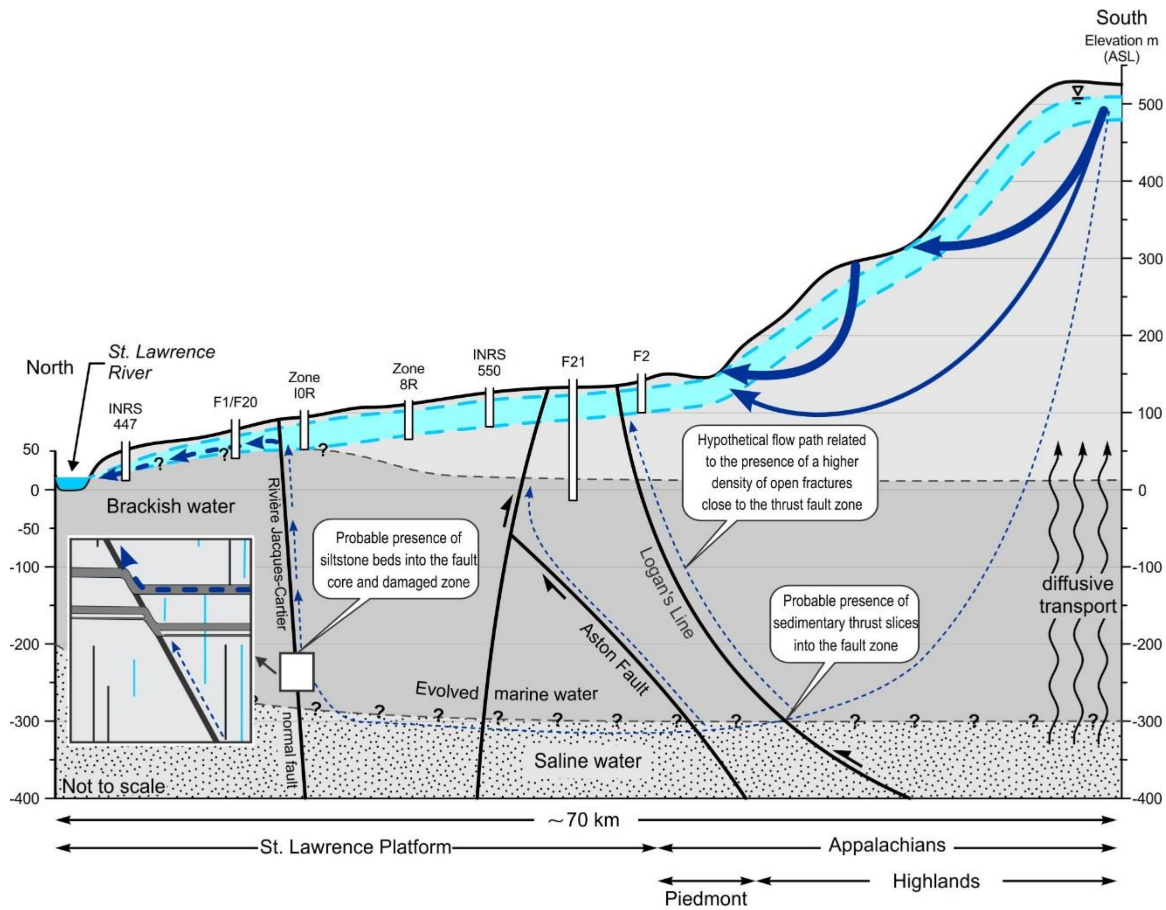

Figure 10. Conceptual model of the regional groundwater flow. Modified from Bordeleau et al. (2018a) and Ladevèze et al. (2018b). Groundwater flow is indicated using blue arrows whose size is related to the relative magnitude of groundwater flow.

Supplementary information

Additional work that proved less conclusive

Other analyses and fieldwork than those presented in the paper were attempted during the course of the project, but led to results that were not as useful as expected. Although this often occurs in research projects, as this paper is meant to provide an overview of the multidisciplinary work and multi-source data collection that was undertaken, it was decided to briefly present some of this work in this section. Nonetheless, these activities could be successful and beneficial in other contexts. The main activity that did not reach its objective is the shallow high resolution seismic survey that aimed to image the upper portion of the intermediate zone, especially in fault zones. However, this activity was discussed in section 3 and will not be addressed here.

Soil gas geochemistry

A survey of soil gas concentrations (CO_2, radon, methane, ethane, propane, butane) was planned early in the project to try to indirectly identify areas containing more hydrocarbons and radon in soils, which could have implied that upward fluid migration was actively occurring in the study area. The soil hydrocarbon gas survey was carried out over 247 stations, whereas radon gas was measured at 155 stations (Aznar and Malo, 2014).

Methane was found to be very abundant locally in soils, especially in the southeastern part of the study area. However, locations with high methane concentrations were neither correlated with locations having high ethane, propane and butane concentrations in soil gas, nor with high methane concentrations in groundwater. Therefore, it is believed that the major part of methane in soil gas is produced locally by microorganisms at shallow depths, and is not an indication of higher concentrations at somewhat greater depths in the bedrock aquifer. The highest concentrations of radon in soils were also detected in the southeastern part of the surveyed area, and do not correspond to the locations of higher radon concentrations in groundwater (Figure S1). However, it is interesting to note that the highest radon concentrations in groundwater were found along the normal fault (F7, Zone 9R) and to the north of it (F1), where presence of small amounts of deep formation brines was inferred from geochemical data. Unfortunately, no soil gas samples were collected in the vicinity of wells Zone 9R, F7 and F1.

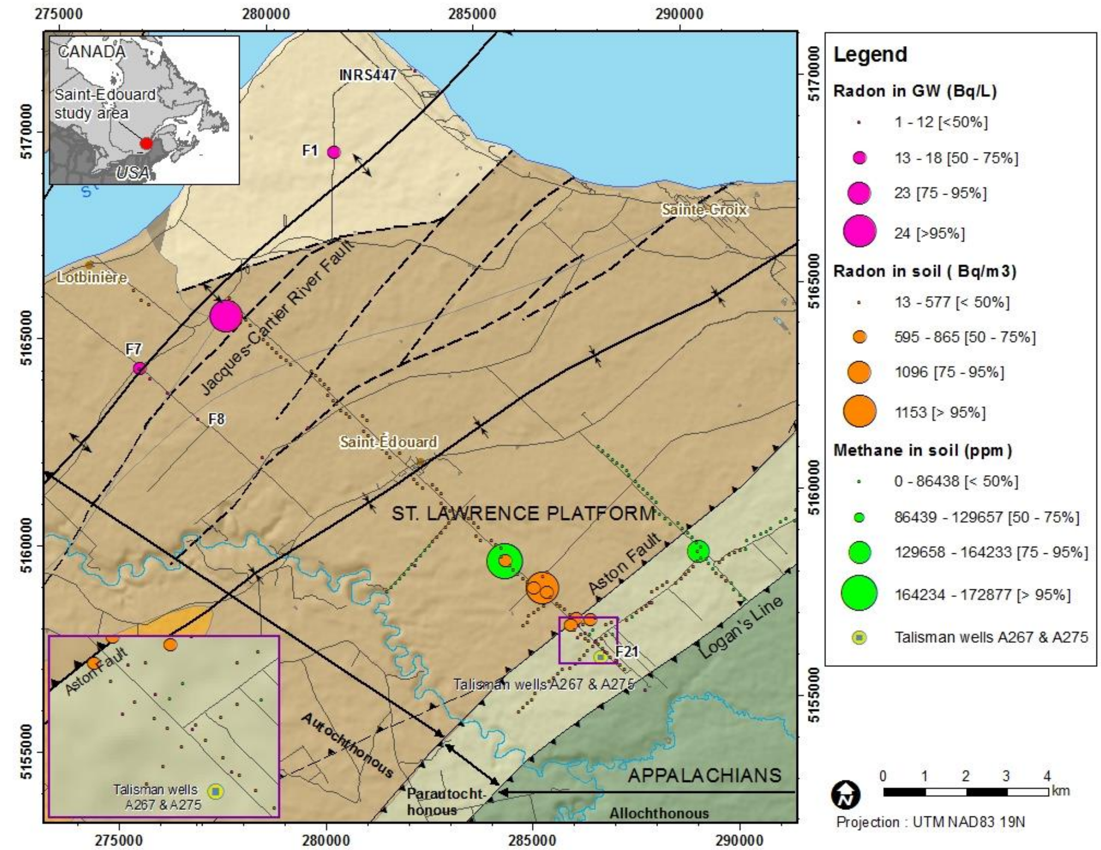

Figure S1. Spatial distribution of methane and radon (^{222}Rn) in soil, as well as radon (^{22}Rn) in groundwater of the Saint-Édouard area, using different percentile classes. Uranium values in rock represent mean values for the different sampled depths.

HTEM survey

Since the shallow seismic survey could not help locate the surface extension of known fault zones, a 75 km-long helicopter-borne transient electromagnetic (HTEM) survey was flown, from a topographic high in the Appalachians to the St. Lawrence River. The helicopter flew at a mean altitude of 109 m above the ground with an average survey speed of 80 km/h. This survey was designed to locate fault zones and map out the freshwater-saltwater interface. Information on the depth of saline water is important, as it is much denser than freshwater and, hence, plays an important role in the system hydrodynamics.

The interpretation was carried out in a two-step procedure. First, a classical 1-D layered inversion using seven layers based on data from resistivity logs (see section 5.2) was used, but it did not provide results that seemed to fit with the current knowledge of the study area, particularly in the vicinity of the RJC fault. Second, HTEM data were inverted using laterally constrains (Auken et al., 2005) and Bayesian inversion using non-stationary Matérn matrix (Bouchedda et al., 2017). This allowed for a more laterally continuous and more accurate spatial distribution of the electrical resistivity of the ground. The second resistivity model tended to confirm that there is a compartmentalisation of groundwater flow on each side of the normal fault zone. Resistivity contrasts in the vicinity of the fault zone indeed suggest that the latter could constitute a barrier to flow, especially around the normal RJC fault. Evidence of such compartmentalisation has been provided by geochemical data (see section 6.1).

Geomechanical laboratory testing

Laboratory tests on core samples had been planned to calibrate the geomechanical parameters derived from acoustic and other petrophysical logs from shale gas wells. However, the only deep shale core samples that were available for physical testing consisted of one quarter of a 2.5 inches cores, which were, moreover, very friable. Therefore, it had been decided to strengthen these samples with a cement grout or gypsum that had similar geomechanical properties than this shale to run dynamic (acoustic) tests and static (uni- and tri-axial compression) tests in laboratory.

Results of the laboratory (dynamic) tests on these hybrid samples showed that most Poisson's ratios had values outside the plausible range (0-0.5) and thus had to be rejected, while the comparison for Young's modulus values with those obtained with well logs showed that laboratory test values were significantly lower. These low values are likely due to a loss of structural integrity of the core samples, which could have occurred during their *in situ* sampling, transport or handling in the laboratory or could be due to the inadequacy of cementation for these samples. It was thus concluded that these grouted core samples could not provide reliable values in this case. These laboratory tests, their results and comparison with those obtained from well logs are discussed in Séjourné (2016).

Downhole gas sensors

Efforts were also invested in downhole sensors for monitoring purposes, as it had been observed early in the project that methane concentrations could vary significantly over time in this region and it would have been interesting to have more information about short-term (at least daily) variations. Two types of sensors were installed in a few observation wells: the first type provided total dissolved gas pressure (TDGP), while the second type measured both TDGP and methane. TDGP sensors were meant to provide a proxy for dissolved methane concentrations, as methane is difficult to estimate using permeable membranes when concentrations are significant (above 1 mg/L with the probes that were used for this project). Downhole gas sensors were deployed in three wells to measure daily methane concentrations in groundwater.

The TDGP sensors did not provide reliable values, as groundwater in these wells is effervescent and is thus degassing; hence dissolved gas could escape through the free phase as the sensors had not been installed below a packer. Moreover, chemical analysis of the different dissolved gases (N_2, CO_2, O_2+Ar, and C_1-C_5 alkanes) over time in the selected wells showed that the proportions of individual gases may vary significantly; hence, TDGP is not a good proxy for methane concentrations in this region.

The methane sensor did not generate useful data because methane concentrations were quite high and as the permeable membrane inside becomes saturated, it takes several days, even weeks, to adapt to new conditions. Although downhole sensors appear to be promising tools for methane monitoring, additional technical development must be performed before they can be routinely used in a gas-rich environment.

Strontium isotopes

Stable strontium isotopic ratios ($^{87}Sr/^{86}Sr$) were analysed in 23 groundwater samples, seven (7) shallow bedrock (41-146 m) samples, and three (3) deep rock samples from the Lorraine (1745 m) and Utica (1883 and 2000 m) shales. These analyses were planned because strontium has proven to be a useful tracer for groundwater flow paths (Clark and Fritz, 1997).

$^{87}Sr/^{86}Sr$ ratios varied widely, ranging from 0.7091 to 0.7151 in groundwater and from 0.7085 to 0.7313 in rock samples. The $^{87}Sr/^{86}Sr$ ratios in groundwater follow the trends of their respective host rock formation, but with less radiogenic values, indicating that the isotopic signature of the shallow rock formations is imparted to the groundwater, along with another, less radiogenic Sr end-member. Potential candidates for this end-member include Utica Shale brines, Champlain Sea water, or overburden sediments. Brines from the intermediate zone are another potential candidate, but no data is available for rocks between 146 m and 1745 m. The measured $^{87}Sr/^{86}Sr$ ratios did not allow identifying the end-member, but mass balance calculations considering Sr concentrations advocate against Utica Shale brines. Hence, Sr isotopic analyses did not provide exclusive information and did not unequivocally allow ruling out the presence of Utica brines in shallow groundwater, but the results are still consistent with our interpretation of the hydrogeological system in this area (Bordeleau et al., 2018).

Pore-water from shallow and deep shale samples

As an upward flow of formation brines was identified in the vicinity of the RJC normal fault but the depth of the source is unknown, an attempt was made at analysing pore water in shale core samples from shallow (0-150 m) and deep (1900- 2500 m) wells, based on the work of Clark et al. (2013). The rationale was that some brine might still be trapped in the pore space of the rock samples, and its geochemical fingerprint could be compared to the brines found in groundwater (similar to what was done with the shallow bedrock alkanes and groundwater alkanes). Unfortunately, not enough pore-water could be extracted from these tight shale samples, and analyses of Br/Cl and $^{87}Sr/^{86}Sr$ ratios could not be performed.

Presence of phospholipid fatty acids (PLFAs)

Analyses of phospholipid fatty acids (PLFAs) from crushed shale samples from the Nicolet, Lotbinière and Les Fonds formations were attempted in order to identify biomarkers for *in situ* active microbial population. Distributions and stable carbon isotope ratios ($\delta^{13}C$) of PLFAs can provide valuable insight into sources and biogeochemical cycling of carbon (Ahad and Pakdel, 2013). However, despite repeated attempts to extract PLFAs using large amounts of crushed rock (between 0.5 to 1.0 kg), PLFA yields from these samples were consistently around the same trace levels as those determined in process blanks. Although no direct understanding into microbial carbon sources could be obtained, these results indicate very little active microbial carbon cycling within the shale, and thus point to an ancient or pre-existing source of methane in groundwater samples not associated with on-going microbial utilization of fossil carbon. Therefore, late-stage methanogenesis that was recognized in a large number of groundwater samples in this study area would likely be attributable to old methane generated in the distant geological past, and not recent methane produced by microbes using old carbon.

Presence of naphthenic acids (NAs)

Acid extractable organics (AEOs) in groundwater samples were analysed to determine the occurrences, distributions and sources of naphthenic acids (NAs) in the subsurface (Ahad et al., 2018). As classically defined by $C_nH_{2n+Z}O_2$, the most abundant NAs detected in the majority of groundwater samples' AEOs were straight-chain (Z = 0) or monounsaturated (Z = -2) C_{16} and C_{18} fatty acids. Several groundwater samples, however, contained significant proportions of potentially toxic alicyclic bicyclic NAs (i.e., Z = -4) in the C_{10}-C_{18} range. These compounds may have originated from migrated waters containing a different distribution of NAs, or are the product of *in situ* microbial alteration of shale organic matter and petroleum. Although concentrations of AEOs were very low (< 2.0 mg/L), the detection of these compounds in groundwater overlying an undeveloped unconventional hydrocarbon reservoir points to a natural background source. In light of these findings, routine screening for NAs in environmental samples from areas undergoing shale gas development may be warranted.

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Supplementary tables

Table S-1: Characteristics of the observation wells

Site	Drilling type	Drilling year	Total drilled depth (m)	Static water level (m below TOC)	Overburden thickness (m)	Sampling depth (m below TOC)	Conditions	K (m/s)	Geological formation
F1	Diamond	2013	50	1.86	2.44	7.5	Semi-confined	5.93×10^{-7}	Lotbinière
F2	Diamond	2013	52	2.76	6.10	21.5	Semi-confined	2.38×10^{-7}	Les Fonds
F3	Diamond	2013	50	1.68	20.12	22.7	Confined	3.97×10^{-7}	Nicolet
F4	Diamond	2013	60	8.87	40.84	54.0	Confined	3.02×10^{-6}	Les Fonds
F5	Hammer	2014	52	2.30	9.75	14.4	Confined	4.55×10^{-7}	Nicolet
F6	Hammer	2014	52	2.35	6.71	10.0	Confined	2.00×10^{-6}	Nicolet
F7	Diamond	2014	52	4.42	11.43	17.7	Semi-confined	1.10×10^{-5}	Nicolet
F8	Diamond	2014	52	0.72	1.43	20.2	Confined	3.12×10^{-6}	Nicolet

F10	Hammer	2014	30	3.84	15.85	23.8	Confined	$6.15x10^{-7}$	Les Fonds
F11	Hammer	2014	55	2.60	6.4	10.3	Semi-confined	$4.55x10^{-8}$	Les Fonds
F12	Hammer	2014	73	3.02	7.92	20.4	Confined	$1.94x10^{-7}$	Les Fonds
F13	Hammer	2014	61	2.13	1.83	7.7	Confined	$3.38x10^{-9}$	Les Fonds
F14	Hammer	2014	30	30.92	14.33	dry	Confined	$5.78x10^{-7}$	Les Fonds
F20	Diamond	2014	50	0.73	3.05	7.4	Semi-confined	$2.26x10^{-9}$	Lotbinière
F21	Diamond	2014	147	3.60	3.05	145	Confined	$5.93x10^{-7}$	Les Fonds

Notes:

1) Diamond: Diamond-drilled well with a 100 mm (4 in.) diameter; Hammer: Hammer-drilled well with a 152 mm (6 in.) diameter. TOC: top of casing.
2) During the 2013 drilling campaign, it was observed that pores in the shallow bedrock contained a lot of gas with the light components (C_1 to C_3) being mostly lost due to evaporative loss. Therefore, core samples from the 2014 drilling program were carefully preserved for different types of hydrocarbon analyses (see section 6.2). As the 2013 well F1 cores contained a significant volume of gas (but the light fraction was lost), well F20 was drilled next to it (6 m away) in 2014 to obtain data on the full spectrum of gas in the rock pores.

Table S-2: Information or evidence for or against upward migration

Discipline	**Evidence in favor of potential large-scale upward migration**	**Evidence against large-scale upward migration**
Geophysics: seismic reflection and VTEM surveys	Reprocessing and re-interpretation of deep old seismic surveys from the industry showed possibilities for upward migration to a certain depth. However, a clear reflector imaging in the upper ~1000 m of this survey was difficult and the near-surface seismic survey could not fill the gap due to the geological context that was not well suited for this method. The stochastic inversion of the VTEM survey revealed an anomalous feature that supports the assumption that the normal fault acts as a regional discharge zone by showing a drastic difference in saline water depths upstream and downstream of it.	
Geomechanics		Based on geomechanical properties such as Poisson's coefficient and Young's modulus, the IZ was found to represent an efficient barrier to the propagation of induced fractures during hydraulic fracturing, thus providing a good protection for the shallow GW aquifers.

Structural geology, borehole logging	Well-known regional fault zones are present in the study area: a normal fault zone in the northern part and a thrust – back-thrust fault system in the southern part. Data from deep shale gas wells showed that denser natural fracture networks can develop in the close vicinity of thrust faults. The presence of siltstone interbeds, which are more permeable than shale beds, have probably been dragged into the normal fault core in the upper part of the IZ, where they are frequently present. This would explain the presence of brines in shallow observation wells near the normal fault (see below).	The Utica Shale is known to be overpressured. This suggests that there is no "leak" from the gas reservoir. Density of the open natural fractures is very low at depth and the risk of connectivity is minimal. Open fractures at depth close to thrust faults are mostly parallel (in the same direction as fractures from FS1), greatly limiting possible interconnections and thus an increase in permeability. The IZ is composed of low *K* shale. This shale is clayey. Gouge is present in fault cores.
Hydrogeology	Although very few local data are available on the vertical hydraulic gradient, as the Utica Shale is overpressured, the overall gradient could make groundwater flow upward given that conditions (sufficient permeability and gradient) would allow it and counteract the opposite effect of high salinity on formation water density.	The presence of overpressure suggests that conditions for significant upward movement do not exist.

Geochemistry: groundwater, rocks, soils and GW monitoring	15% of the wells show a thermogenic component in groundwater. Presence of thermogenic gas and radon in soils. Groundwater geochemistry, and in particular geochemical profiles, showed that brines were present in a few shallow wells, mainly located close to or in the damage zone of the hanging wall of the normal fault. These brines with a higher Br/Cl ratio than sea water can only come from deeper units. Dissolved methane show very high fluctuations of methane concentrations, both spatially and temporally. Likewise, methane isotopic composition can show large variations over time.	Thermogenic methane found in GW does not have the same isotopic composition as methane in the Utica Shale. There is not more thermogenic methane in wells containing brines, indicating that the latter does not come from the Utica Shale. Both microbial and thermogenic gas appears to come from the shallow bedrock itself based on a comparison with gas extracted from shallow core samples.

Note: IZ: intermediate zone; GW: groundwater