

Article

Impacts of Salinity on Saint-Augustin Lake, Canada: Remediation Measures at Watershed Scale

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Academic Editors: Joan M. Brehm and Brian W. Eisenhauer

Received: 3 May 2016; Accepted: 5 July 2016; Published: 11 July 2016

Abstract: Winter road network management is a source of anthropogenic salinity in the Saint-Augustin Lake watershed (Quebec City, QC, Canada). To prevent the potential impact caused by road runoff involving de-icing salts (NaCl) and trace metals (Cd and Pb) on the watershed, a full-scale treatment chain system (including a detention basin, a filtering bed, and a constructed wetland) was built. Average Cl and Na concentrations in groundwater were higher in wells affected by road network (125 mg/L Cl and 64 mg/L Na) than in control wells (13 mg/L Cl and 33 mg/L Na) suggesting a contamination by de-icing salts. The monitoring of influent and effluent surface water in the treatment system has shown a seasonal dependence in NaCl concentrations and electrical conductivity values, being the highest in summer, linked with the lower precipitation and higher temperature. Concentration ranges were as follows: 114–846 mg/L Na and 158–1757 mg/L Cl (summer) > 61–559 mg/L Na and 63–799 mg/L Cl (spring and autumn). The treatment system removal efficiency was significant, however with seasonal variations: 16%–20% Cl, 3%–25% Na, 7%–10% Cd and 7%–36% Pb. The treatment system has shown an interesting potential to mitigate the impact of anthropogenic salinity at watershed scale with higher expected performances in the subsequent years of operation.

Keywords: de-icing salt; trace metals; surface water; groundwater; watershed; remediation; ecoengineering

1. Introduction

Over the past decades, road networks have been globally extended to support urban expansion. As such, road runoff has become an important cause of urban water quality deterioration [1]. Particulate matters, nutrients, hydrocarbons, and trace metals are part of the pollutants found in road runoff [2]. Additionally, northern countries with cold climate need to use large amounts of de-icing salts during winter on the entire road network in order to ensure safe driving conditions. In Canada, four million tons of salts per year to de-ice roads are used [3]. The Ministry of Transport of Quebec (MTQ) uses approximately 1.5 million tons of de-icing salts each year [4]. De-icing salts are essentially composed of sodium chloride (NaCl) but can also be a complex mixture of calcium (Ca), magnesium (Mg), and potassium (K) ferrocyanide. Abrasive materials, such as gravel, are also used during snowstorms or freezing-rain events.

Winter practices are directly implicated in the origin of the anthropogenic salinity in watershed water (surface and groundwater). About 50% of the salt applied to paved road surfaces is transported via surface runoff, with the remaining 50% entering the subsurface as aquifer recharge [5]. Salinity

has a significant impact on water ecosystems, producing toxicity to benthic and other freshwater organisms and affecting invertebrate reproduction [6]. As reported by Novotny and Stefan [7], de-icing salts can modify lake stratification and indirectly aggravate lake eutrophication processes by extending anoxia conditions and allowing the release of phosphates from organic sediment. The increase of salt concentration in groundwater is a concern for drinking water [8]. A high proportion of salts can also be accumulated in roadside soils and vegetation [9], which can affect soil physicochemical properties, biogeochemical cycles, and soil ecology. In addition, road runoff includes toxic trace metals such as Cd and Pb [10]. Indeed de-icing salts have a strong ability to increase trace metal mobility [11]. In addition, long-term use of de-icing salts can induce problems associated with chloride-induced corrosion of automobiles and road infrastructure (highway components, steel reinforcement bars, and concrete) and accelerates pavement deterioration due to freezing and thawing cycles [12]. Despite de-icing salts negative impact on the environment in the short and long term [13,14], at present it is difficult to stop or reduce its use for safety reasons. Therefore, strategies are aimed at preventing the potential impact caused from road runoff by reducing pollutant concentration. Nowadays, there are various strategies including constructed wetlands, oil and grit separators, and storm-water ponds, which are normally part of a treatment chain system [15]. However, few of these systems target the removal of de-icing salts from road runoff.

The 1970s construction of a road section of the Felix-Leclerc Highway A40 near Quebec City (Canada) has greatly contributed to water quality degradation in the Saint-Augustin Lake watershed. Several studies show that Saint-Augustin Lake water quality is being compromised by the increased human settlements around the watershed [16–18]. The proximity of the Highway A40 to Saint-Augustin Lake has induced surface water, groundwater, and sediment pollution by road de-icing salts and trace metals. Indeed, different water and sediment quality changes in Saint-Augustin Lake were observed after the Highway A40 construction: (i) high electrical conductivity (0.7–1.3 dS/m) in surface water was recorded with a positive relation with Na and Cl ions [10]; (ii) saltwater algae species are now present in the lake [17]; and (iii) high trace metal concentrations were measured in sediments [19]. Moreover, after the construction of the Highway A40, high electrical conductivity values and NaCl concentrations in groundwater were reported [20]. In order to mitigate the impacts of de-icing salts on water quality in the Saint-Augustin Lake watershed, a full-scale chain system to treat road runoff from Highway A40 was constructed. The treatment chain system was built in 2011 and includes a detention basin, an active filtering bed and an adapted constructed wetland.

This work concerns a field research project that includes the design of the system, its construction and monitoring program to evaluate groundwater and surface water quality in the Saint-Augustin Lake watershed, as well as system performance in the first and second years of operation. Specific discussion is made on the temporal variation patterns of NaCl and trace metals (Cd and Pb), and other water quality indicators (temperature, pH, electrical conductivity, and suspended solids).

2. Materials and Methods

2.1. Site Description

The water quality monitoring was conducted from spring 2012 to autumn 2013 in Saint-Augustin Lake watershed (flood plain of St. Laurent River, Saint-Augustin-de-Desmaures, QC, Canada), located in the municipal limit of Saint-Augustin-de-Desmaures (40 m altitude). The Saint-Augustin Lake watershed is located about 15 km in the west of Quebec City (Canada) and has an area of 7.64 km² (Figure 1). Saint-Augustin Lake has a maximum dimension of 2.1 km in length, 0.3 km in width [21] and a depth average of 3.5 m [19]. The water renewal rate is estimated at 6 months and it is mainly driven by groundwater inflow [10]. Average annual temperature is 5.1 °C and total annual precipitation (rainfall + water equivalent of the total snow) is 1109 mm/year for the 1998–2012 period (Station Jean Lesage, Quebec City, QC, Canada) [22]. The Saint-Augustin Lake watershed is included in two geological formations of the Paleozoic. The north-northwest of the watershed, including the lake is in the St.

Lawrence Platform formation, upper and middle Ordovician. This formation is characterized by a rock composed of Mudrock, slate, dolomite and sandstone. The south part of the watershed is in the Appalachians Province, Ordovician to lower Silurian characterized by a rock composed of shale.

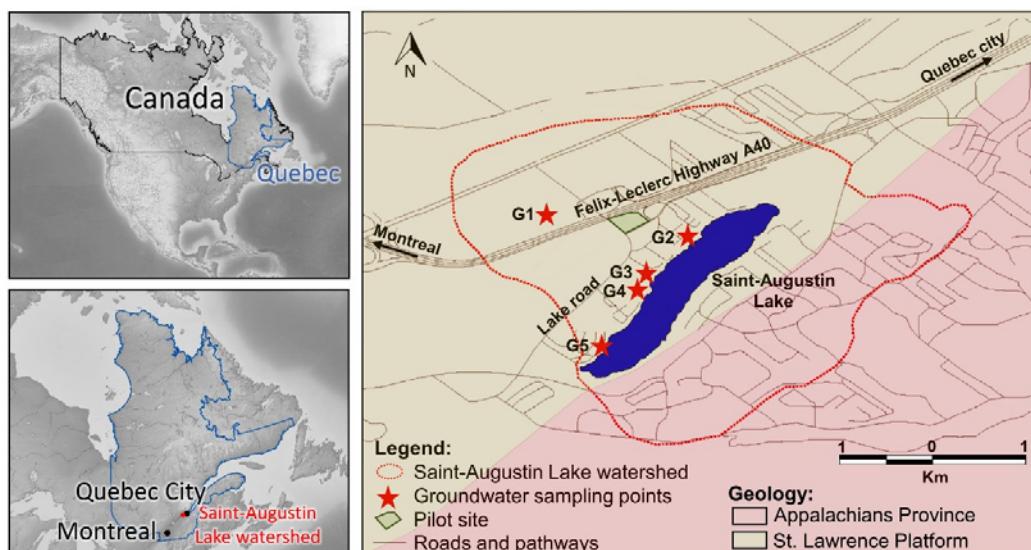


Figure 1. Groundwater sampling wells in Saint-Augustin Lake watershed (Quebec) and geological context. Red stars represent groundwater sampling wells: G1 (road control); G2 (residential control); and G3 to G5 (study wells affected by road and residential areas). Pilot site to runoff desalination is also indicated in the picture.

From 1960 onwards, population and land use have markedly changed. Conversion of forest to residential zones has become the dominant land use change. Currently, main land uses are agriculture, forestry, and residential, being 20%–25%, 20%, and 30% of the total area, respectively. In 1974, a section of Felix-Leclerc Highway A40 was constructed through part of the watershed (Figure 1) resulting in an increase in road traffic. Currently, the annual average daily traffic on this section is around 72,000 vehicles [23], which corresponds to a medium highway [2]. This construction was accompanied by increased population in the municipality [17]. In last decades, the electrical conductivity in water of Saint-Augustin Lake highly increased, coincident with the use of de-icing road salts on Highway A40 [10]. With the aim to deal with salt pollution in runoff water, in 2011 an environmental management solution (hereafter named pilot site) was constructed [24].

2.2. Pilot Site Characteristic

The pilot site is a treatment chain system composed of three units (Figure 2): (1) detention basin; (2) active filtering bed; and (3) free surface-flow adapted constructed wetland. The units were designed and constructed in 2011 in order to collect and treat approximately 25% of the Saint-Augustin Lake watershed's runoff water.

The pilot site was constructed within the limits of the Highway A40 ramp at the north of Saint-Augustin Lake. The site is compact and well integrated into the landscape with an area of 22,500 m². Runoff is collected in a highway drainage ditch and accumulated in the detention basin before being redirected to the active filtering bed and/or the adapted constructed wetland (Figure 3).

The detention basin plays a role in the homogenization of the water collected from the highway ditch and in the regulation of flow water to the adapted constructed wetland and the active filtering bed. The detention basin accumulates road runoff and removes sediments and particles. The detention basin surface area is 340 m² and the volume is 880 m³.

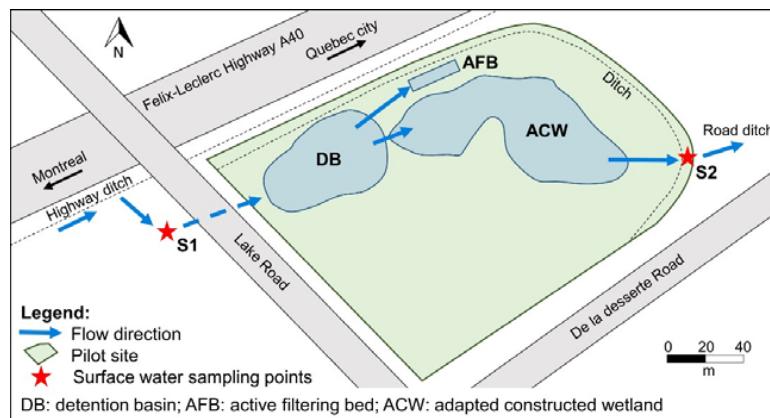


Figure 2. Surface water sampling points in the pilot site located in Saint-Augustin Lake watershed (Quebec). Red stars represent surface sampling points: S1 (influent water) and S2 (effluent water).

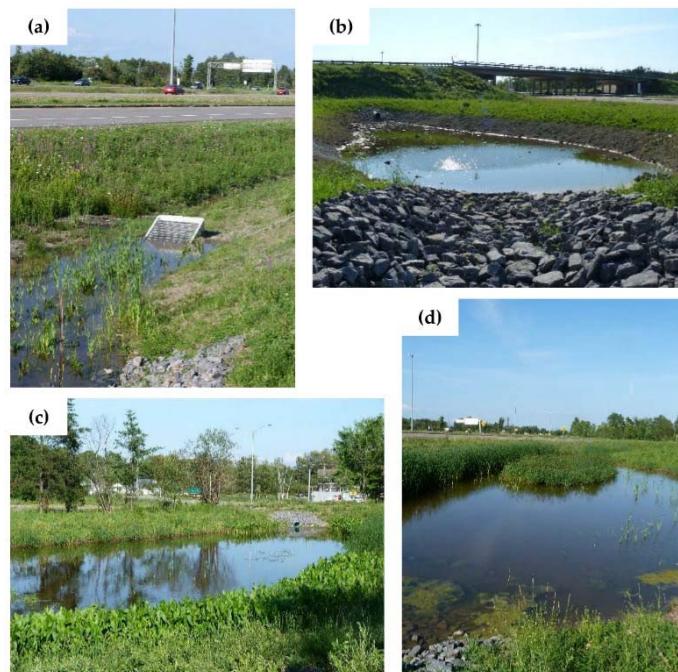


Figure 3. Pictures of the pilot site located in Saint-Augustin Lake watershed (Quebec): (a) active filtering bed entrance sump; (b) detention basin; (c) adapted constructed wetland—deep zone; and (d) adapted constructed wetland—shallow zone.

The active filtering bed uses an underground reactive media composed of a limestone (80%) and dolomite (20%) mixture. The functioning is based on the runoff circulating through a granular bed (the reactive media) whose chemical properties enhance contaminant retention. Clean stone from the reactive media has an average diameter between 5 mm and 10 mm, which could ensure a permeability of 10 cm/s. The active filtering bed is 27 m long and 4.8 m wide (average width of the trapezoid), which gives a flow section of 2.88 m^2 . The active filtering bed is totally isolated from both soil and surface by an impermeable geotextile, and also from the base by the superposition of granular material and a geomembrane.

The adapted constructed wetland is a free surface-flow system where substrate, water and plants imitate the processes occurring in natural swamps. The adapted constructed wetland consists of a shallow pond divided into 3 plots of land where aquatic plants were cultivated. There is an inlet control equipped with a drainage pipe and a valve at the outlet that controls the water level as needed

by the plants. The adapted constructed wetland dimensions are: 850 m³ volume and 5/1 length/width. The bottom is impermeable, made from a bentonite geomembrane. Planted were typical wetland plants, such as *Typha angustifolia* and *Eleocharis palustris*. Moreover, halophyte plants were planted, such as *Atriplex patula*, *Spergularia canadensis*, and *Salicornia europaea*. Halophyte plants are an interesting alternative to plants traditionally used to treat the road effluents: (i) they are resistant to strong salinity; (ii) they can accumulate moderate to high quantities of salt in its biomass; and (iii) they can reduce the load of trace metals by bioaccumulation and biostabilisation [9,24].

2.3. Sampling Strategy

2.3.1. Groundwater

A strategic sampling has been set up to characterize and monitor groundwater based on the location of the A40 highway, the rainwater drainage, and the piezometric and well network installed in the study area. Indeed, several sampling wells and piezometers are installed in the Saint-Augustin Lake watershed as part of previous research studies [20,25]. Based on these studies, several wells to collect groundwater samples were selected (Figure 1): piezometer G1 (road control); monitoring well G2 (residential control); and monitoring wells G3, G4, and G5 (study wells). Special attention was placed on the geological composition and land use of sites where selected wells are installed were as similar and comparable as possible. The piezometer G1 is located upstream of the Highway A40. In this area groundwater is not affected by runoff pollution from Highway A40. The G2 groundwater well is located in a residential area but away from the road area. In this area, groundwater is not affected by runoff pollution but by residences. Therefore, G1 and G2 are considered as reference monitoring points not affected by road de-icing salts due to its location. In contrast, the G3 to G5 groundwater wells are located upstream of Saint-Augustin Lake and placed in a North-West/South-East axis, being affected by both road and residential areas. Groundwater samples in duplicate were taken, with a single valve bailer, once a week from July to November 2013 (135 days) in order to monitor Cl and Na concentrations. Other major cations and anions (K⁺, Ca²⁺, Mg²⁺, HCO₃⁻, CO₃²⁻, SO₄²⁻, and PO₄³⁻), as well as trace metals (Cd and Pb), were also monitored in this period. Some water quality indicators (temperature, pH, and electrical conductivity (EC)) were measured in situ using a multiprobe meter (Model YSI 6600 V2, YSI Inc., Yellow Springs, OH, USA). Water level was measured with an electrical probe called “Deep water level”.

2.3.2. Surface Water

Surface water was sampled and monitored in all cases at two points at the pilot site (Figure 2). The first sampling point (S1) is located in a Highway A40 ditch at the entrance of the pilot site corresponding to influent water. The second sampling point (S2) is located at the exit of the pilot site corresponding to treated water or effluent water. Pilot site construction and plantation were finished during summer 2011. Between July and October 2011 some water quality indicators were measured in situ (temperature, pH, and EC) or in the laboratory (total suspended solids (TSS)). Then, the system was allowed to stabilize until spring 2012 (snow-melting period), when the monitoring was conducted. Surface water samples were taken once a week from March to November 2012 (238 days) in order to monitor Cl and Na concentrations, water quality indicators (temperature, pH, EC, and TSS), as well as trace metal concentrations (Cd and Pb). Removal efficiency (%) of Na, Cl, Cd, and Pb was calculated in terms of concentration by the following Equation (1):

$$\text{Removal efficiency} = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \quad (1)$$

where C_{in} and C_{out} are the average influent and effluent concentrations, respectively, of Na, Cl, Cd, or Pb (mg/L).

2.4. Analytical Methods

The collected ground and surface water samples were filtered under vacuum through a 0.45- μm -pore-size cellulose ester membrane filter (Advantec Inc., Montreal, QC, Canada). Then the filtrate was frozen (for major anion quantification) or acidified to pH 2 by adding the necessary volume of concentrated HNO₃ and kept at 4 °C until analysis (for major cation and trace metal quantification). Major anions (Cl[−], HCO₃[−], CO₃^{2−}, SO₄^{2−}, and PO₄^{3−}) were quantified by high performance liquid chromatography (1525 Binary HPLC Pump System, Waters Inc., Brossard, QC, Canada). Major cations were quantified by flame atomic emission (Na) or absorption (K, Ca, Mg) spectroscopy (AA240FS, Varian Inc., Mississauga, ON, Canada). Trace metals (Cd, Pb) were quantified by graphite furnace atomic absorption spectroscopy with Zeeman background correction (AA240Z, Varian Inc.). Detection limits in $\mu\text{g/L}$ were: Cd = 0.01, Pb = 0.2, Na = 2, K = 30, Ca = 10, Mg = 3 (spectroscopy method), Cl[−] = 25, HCO₃[−] = 50, CO₃^{2−} = 50, SO₄^{2−} = 75, PO₄^{3−} = 125 (HPLC method). Total suspended solid quantification was conducted following standard analytic procedures [26]. All chemicals and reagents were analytic grade from Fisher Scientific Inc. (Ottawa, ON, Canada) and EMD Chemicals Inc. (Etobicoke, ON, Canada). All glassware used was rinsed with nanopure water (Ultrapure Water System, Barnstead Nanopure, Mississauga, ON, Canada).

2.5. Statistical Analyses

Significance of differences of Cl, Na, Cd, Pb, TSS concentrations and temperature, pH, EC values between influent and effluent surface water (S1 and S2) were investigated by means of *t*-student test. Pearson correlation coefficients were calculated to relate water parameters to each other in order to support data interpretation. Analyses were conducted using Statistical Package for the Social Sciences v. 17 (SPSS Inc., Amonk, NY, USA) software. Graphics were created using GraphPad Prism v. 6.01 software (GraphPad Inc., La Jolla, CA, USA).

3. Results and Discussion

3.1. Groundwater Quality

3.1.1. Hydrochemical Groundwater Characterization

The Stiff diagram (Figure 4) represents the hydrochemical groundwater characterization in the wells G1 (road control well positioned upstream the Highway A40), G2 (residential control well), and G3, G4 and G5 (study wells), based on the monitoring conducted between July and November 2013.

Stiff diagram representation allows the mapping, for each well, of a polygon that assumes a geometry depending on the content of hydrochemical elements and gives an idea of the dominant species and its chemical relationship. The three axes of the diagram are, respectively, up to down: (1) Na⁺–K⁺–Cl[−]; (2) Ca²⁺–HCO₃[−]–CO₃^{2−}; and (3) Mg²⁺–SO₄^{2−}. As shown in Figure 4, higher HCO₃[−] and Ca²⁺ contents were measured in groundwater from G1 and G5 wells. Stiff diagrams allow the chemical classification of the waters according to the anion and cation facies. In this regard, the water type that corresponds to the groundwater sampled in G1 and G5 wells was calcium–sodium bicarbonate–chloride sulphate. Interestingly, EC mean values in groundwater were higher in these wells (0.75 and 0.84 dS/m, G1 and G5, respectively) than in the others (0.25–0.56 dS/m). The other wells (G2, G3, and G4) presented a different hydrochemical water type based on their chemical composition. Indeed, groundwater samples collected in the G2 well were sodium–calcium bicarbonate–chloride sulphate type. In the study wells, the water-type profiles were calcium–sodium chloride–sulphate bicarbonate (the G3 well) and sodium–calcium chloride–sulphate bicarbonate (the G4 well). The relative heterogeneity of results corresponds to the alluvial nature of soils in the study area. The agricultural lands that are in proximity to the G1 well could be partially responsible of its high HCO₃[−] and Ca²⁺ contents, although this would not explain the data of G5. Indeed, results are consistent with the most common facies found in the territory of the Metropolitan Community of Quebec

City (Ca^{2+} - HCO_3^- and Na^+ - Cl^- , associated with recharge areas), and show the typical intrinsic heterogeneity in the spatial distribution of geological deposits of glaciofluvial origin [27]. These facies are found almost exclusively on the north shore of the St. Lawrence River, in the St. Lawrence Platform formation.

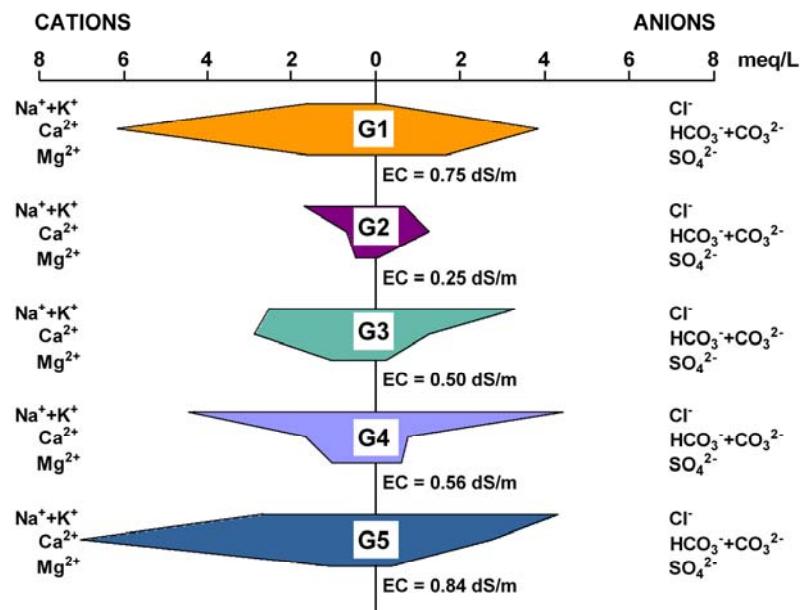


Figure 4. Stiff diagram of the groundwater of sampling wells (G1, road control; G2, residential control; and G3 to G5, study wells). For each ion, data are expressed as mean values (meq/L) of the monitoring performed from summer to autumn 2013. EC: electrical conductivity.

Overall, the groundwater sampled in the study wells (G3 to G5) is much more loaded in Cl than in G1 and G2 control wells: 54% (G3), 67% (G4), and 42% (G5) of the sum of ions. As discussed below, results suggest that the road runoff loaded with Cl from de-icing salts leached into the soil and reached the groundwater downstream of the road network. In contrast, Na was not always the predominant cation in the study wells. This is probably because of Na ions are more easily retained in soils than Cl ions and, therefore, lower Na concentrations in groundwater are expected. If we consider the predominant ions: HCO_3^- was the predominant anion (>50% of the sum of anions) in groundwater in G1, G2, and G5 wells, and Cl in G3 and G4 wells. Considering the cations, Ca was predominant in G1, G3, and G5 wells, but Na in G2 and G4 wells.

Within the groundwater characterization, trace metal (Cd and Pb) concentration was also monitored. However, Cd and Pb concentrations were in all cases below the detection limit.

3.1.2. Water Quality Indicators

Groundwater quality indicators (temperature, pH and EC) in G1, G2, G3, G4 and G5 wells were monitored during summer and autumn seasons and are presented in Figure 5.

The groundwater temperatures ranged from 10 °C to 13 °C (Figure 5a). An increase in groundwater temperature was observed during the summer period, with a decrease in autumn. The highest temperatures were measured in the G1 well, road control (during the summer period), and the smallest values in the G2 well, residential control. The temperatures in groundwater in the G3 to G5 wells were intermediaries.

Regarding pH (Figure 5b), no clear temporal variation pattern was observed. Values fluctuated from 5.6 to 8.3 in the G3 to G5 wells. In the case of control wells, the pH values in G1 were neutral (around 7) while pHs tended to basicity in the G2 with values ≥ 8 . The lowest pH values were measured in the G4 and G5 study wells. Relationships between the pH values and the other measured

water parameters were not observed. Differences in the content and composition of some reactive fractions (carbonate, organic matter, etc.) of soils located in the area of the wells, mainly due to different lithology and land use, may account for the observed pH range.

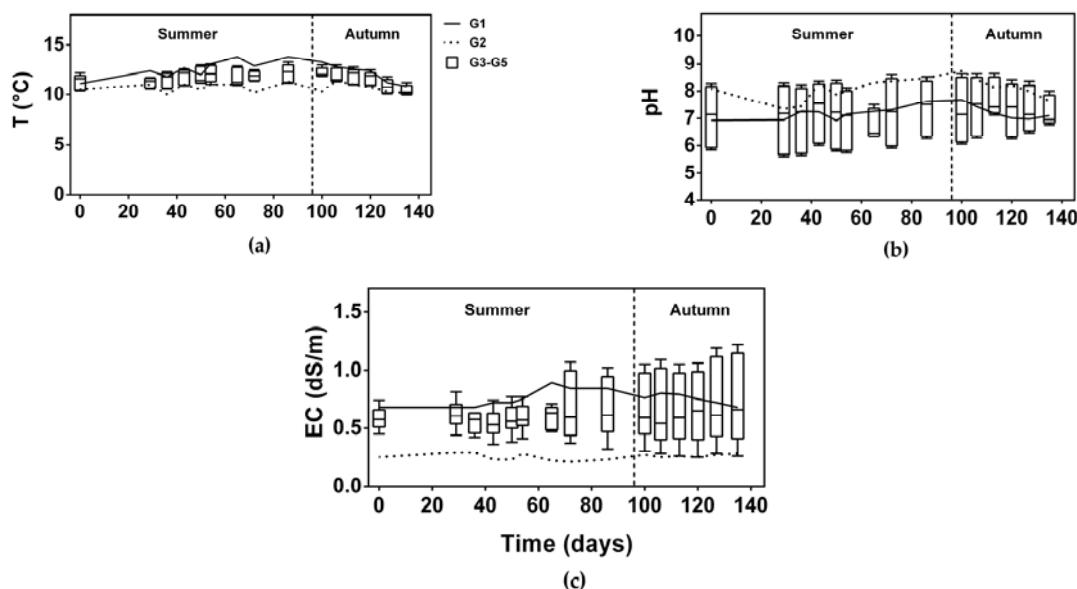


Figure 5. Water quality indicators in groundwater sampling wells (G1, road control; G2, residential control; and G3 to G5, study wells) for the monitoring performed from summer to autumn 2013: (a) temperature (T); (b) pH and (c) electrical conductivity (EC). Solid and dotted lines represent mean values for G1 and G2, respectively. Boxplots show the lower, median, and upper quartiles for G3–G5, with whiskers extending to the most extreme data point.

Figure 5c shows the EC values in groundwater sampled in control and study wells over time. As explained above, significant differences among EC values in the road control well (G1) and the study wells (G3–G5) could not be established. In contrast, the G2 well showed the lowest EC values (0.25 dS/m). In order to support the discussion, Pearson's correlation analysis was performed (Table 1). Interestingly, EC was highly significant and positively correlated with Cl in the study wells ($p < 0.001$) but not in the control wells. This showed a differentiating pattern between control and study wells and suggested that the EC measured in the study wells (G3, G4 and G5) would be mostly due to the NaCl content, in contrast to control wells. Pearson's correlations were also calculated among EC values and the other major anions and cations. The EC values over time were significantly and positively correlated with: (i) Ca^{2+} in the G1, G3 and G5 wells ($p < 0.001$); (ii) HCO_3^- in the G1, G2, and G5 wells ($p < 0.001$); (iii) Mg^{2+} in the G3 and G5 wells ($p < 0.001$); and (iv) SO_4^{2-} in the G1 ($p < 0.01$), G3 and G5 wells ($p < 0.001$). As stated above, groundwater in the G1, G3 and G5 wells belongs to the calcium–sodium facies, which is affecting its EC values. As described above, the intrinsic heterogeneity of the study area could be partially responsible of the EC spatial pattern. Results therefore suggest that the EC spatial pattern in the groundwater of the selected wells is determined by both anthropic (de-icing salts; G3, G4 and G5 wells) and natural influences (weathering of rocks and leaching of ions from soil; G1, G3 and G5 wells).

Table 1. Pearson's correlation coefficients among groundwater parameters at each well.

Well	Parameters	T	pH	EC	WL	Cl ⁻	Na ⁺	Ca ²⁺	Mg ²⁺	K ⁺	HCO ₃ ⁻	SO ₄ ²⁻
G1	T	1	0.674 **	0.788 ***	0.611 *	-0.458	-0.252	0.696 **	0.506	0.588 *	0.813 ***	0.472
	pH		1	0.549 *	0.234	-0.395	-0.192	0.553 *	0.458	0.251	0.368	0.416
	EC			1	0.368	-0.514 *	-0.224	0.968 ***	0.366	0.708 **	0.879 ***	0.660 **
	WL				1	0.075	0.057	0.176	0.680 **	0.568 *	0.535 *	0.067
	Cl ⁻					1	0.547 *	-0.544 *	-0.398	-0.252	-0.437	-0.438
	Na ⁺						1	-0.321	-0.344	0.056	-0.336	0.092
G2	T	1	0.309	-0.220	0.167	0.021	-0.367	-0.105	-0.247	-0.260	-0.122	0.330
	pH		1	-0.593 *	0.474	0.413	-0.146	-0.503	-0.672 **	-0.207	-0.593 *	-0.145
	EC			1	-0.109	0.232	0.335	0.532 *	0.520 *	0.356	0.930 ***	-0.157
	WL				1	0.506	-0.162	-0.201	-0.168	0.350	-0.137	-0.009
	Cl ⁻					1	0.307	-0.269	-0.150	0.258	0.063	-0.085
	Na ⁺						1	-0.048	0.230	0.311	0.014	0.024
G3	T	1	-0.270	0.472	0.554 *	0.563 *	0.148	0.392	0.400	-0.440	-0.198	0.283
	pH		1	-0.876 ***	-0.727 **	-0.903 ***	-0.442	-0.871 ***	-0.725 **	0.100	0.350	-0.905 ***
	EC			1	0.808 ***	0.951 ***	0.405	0.968 ***	0.889 ***	-0.201	-0.076	0.916 ***
	WL				1	0.822 ***	0.337	0.769 **	0.668 **	-0.087	-0.226	0.778 **
	Cl ⁻					1	0.508	0.889 ***	0.754 **	-0.288	-0.293	0.872 ***
	Na ⁺						1	0.304	0.143	-0.089	-0.518 *	0.466
G4	T	1	-0.259	-0.349	0.334	-0.323	-0.133	-0.499	-0.185	0.378	-0.328	0.412
	pH		1	0.615 *	0.618 *	0.605 *	0.542 *	0.399	0.283	-0.238	0.258	-0.446
	EC			1	0.280	0.787 ***	0.881 ***	0.495	0.447	-0.219	0.591 *	-0.510
	WL				1	0.251	0.387	0.035	0.366	0.046	0.035	-0.438
	Cl ⁻					1	0.551 *	0.367	0.458	-0.466	0.120	-0.342
	Na ⁺						1	0.295	0.534 *	-0.109	0.469	-0.530 *
G5	T	1	0.059	0.287	0.454	0.287	0.156	0.302	0.255	0.138	0.233	0.238
	pH		1	-0.661 **	-0.600 *	-0.694 **	-0.510	-0.660 **	-0.681 **	0.059	-0.528 *	-0.689 **
	EC			1	0.946 ***	0.973 ***	0.818 ***	0.989 ***	0.950 ***	0.295	0.937 ***	0.978 ***
	WL				1	0.941 ***	0.704 **	0.930 ***	0.905 ***	0.145	0.897 ***	0.886 ***
	Cl ⁻					1	0.751 **	0.944 ***	0.952 ***	0.276	0.888 ***	0.952 ***
	Na ⁺						1	0.788 ***	0.777 **	0.318	0.700 **	0.833 ***

Notes: T: Temperature; EC: Electrical conductivity; WT: Water level. * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$, statistical significance at these probability levels ($n = 15$).

Regarding the temporal pattern, although mean EC values in control wells remained almost constant throughout the study period (0.68–0.89 dS/m in G1 and 0.21–0.29 dS/m in G2), a different pattern was observed in the study wells (G3, G4 and G5). Thus, the range of variation is much larger in autumn (linked with more precipitation) than in summer. When the EC values in groundwater are evaluated separately taking into account each of the study wells, it was observed that EC: decreases (from 0.64 to 0.36 dS/m in G3), is almost constant (from 0.53 to 0.63 dS/m in G4), or increases (from 0.58 to 1.17 dS/m in G5) over time. Results were compared to the water levels measured at each well over time (data not shown). Pearson correlation analysis showed significant and positive correlations between EC values and the water level in the G3 and G5 wells ($p < 0.001$) (Table 1). Results therefore showed that the higher the water level, the higher the EC in groundwater and that this pattern was not apparently seasonally dependent. Nonetheless, the longer residence time of groundwater than that of surface water could be offsetting the seasonal pattern. Higher infiltration of water charged in salts during the increase of water levels may account for the positive correlation obtained between the EC values and the water level, which deserves further studies.

3.1.3. NaCl Concentration

The concentration of Cl and Na in groundwater (G1 to G5 wells) has been monitored from summer to autumn 2013 (Figure 6).

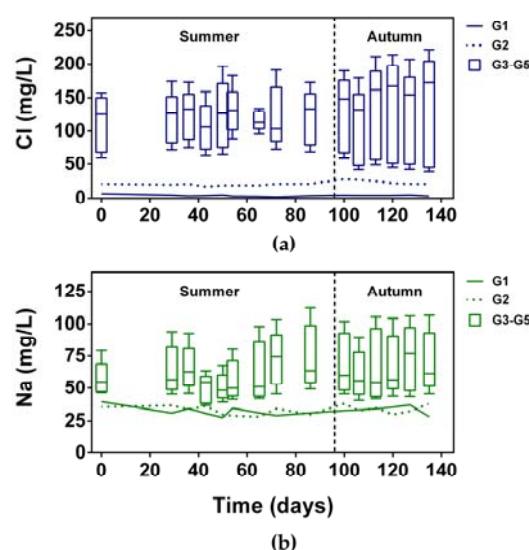


Figure 6. Concentration of Cl and Na in groundwater sampling wells (G1, road control; G2, residential control; and G3 to G5, study wells) for the monitoring performed from summer to autumn 2013: (a) Cl and (b) Na. Solid and dotted lines represent mean values for G1 and G2, respectively. Boxplots show the lower, median, and upper quartiles for G3–G5, with whiskers extending to the most extreme data point.

The Cl concentration in groundwater in the two control wells (4 and 22 mg/L in G1 and G2 respectively, ~13 mg/L on average) was in all cases lower than that quantified in the study wells (from 39 to 222 mg/L, 125 mg/L on average, G3–G5). Interestingly, results showed that even though major ion concentrations (Figure 4) and EC values (Figure 5) are elevated in G1 well, Na and Cl concentrations are very low (Figure 6). The higher Cl concentrations obtained in G2 with respect to G1 can be explained by the residential character of the area. Indeed, septic tanks are possible sources of groundwater salinization [8]. The higher Cl concentration in groundwater in the study wells (G3 to G5) than in the control wells (G1 and G2) indicates that the road network and de-icing salts impact the groundwater chemical composition. This observation was also stated by Daley et al. [8], which reported that sewage and water softeners accounted for only 7% of the Na and Cl input to a single

rural watershed in New Hampshire, whereas NaCl used for de-icing accounted for 91%. Regarding Na, the same pattern was observed as in the case of Cl. Thus, Na concentration in groundwater in the two control wells (G1 and G2) showed the lowest values (33 mg/L on average). In the case of the study wells (64 mg/L Na on average, G3–G5), the maximum Na values even reached up to 112 mg/L. This showed the importance of the Highway and road network on the contamination of groundwater. As it was observed for EC, the range of variability in Cl concentration in the study wells was higher in autumn than in summer, attributed to the cooler temperatures and higher precipitation rates during autumn, which facilitates flushing of salts through groundwater. A relationship with the water level was also observed (Table 1). Thus, significant and positive correlations among the water level were obtained with Cl in the G3 and G5 wells ($p < 0.001$) and with Na in the G5 well ($p < 0.01$). As stated above, EC and Cl and Na ions were significantly positively correlated to each other in the study wells ($p < 0.001$) but not in the control wells. These results are in accordance with the study conducted by Galvez-Cloutier et al. [20] for the same lake and season.

Groundwater is an important source of drinking water. According to McConnell and Lewis [28], 25% to 50% of road de-icing salt may reach groundwater, increasing the salinity of drinking water sources in public distribution networks. Data on drinking water from several Canadian provinces indicate that the Cl concentrations are generally less than 10 mg/L [29]. In the present study, results showed that Cl concentrations in study wells (G3–G5) are very high, up to 22 times higher than 10 mg/L. In the case of Na, the concentration ranges in both the control wells (28–30 mg/L Na) and the study wells (37–112 mg/L Na) were within the normal ranges for groundwater in Canada (from 6 to 130 mg/L) [30]. The concentrations of Cl and Na remained in all cases below the drinking water quality standards of Canada: 250 mg/L and 200 mg/L, respectively [31]. However, results highlight that the highest NaCl concentration is found in the groundwater from the areas of the watershed affected by the road network (study wells), which makes it essential to perform long term monitoring.

3.2. Surface Water Quality

3.2.1. Water Quality Indicators

Table 2 shows the water quality indicators measured in influent (S1) and effluent (S2) surface water at the pilot site during summer and autumn, 2011, after its construction. During the first period of operation (from mid-2011), no significant differences were observed on water quality indicators between S1 and S2 sampling points. Nonetheless, a trend to decrease was observed on EC (from 1.9 dS/m at S1 to 1.1 dS/m at S2) with higher values during the summer period. A decreasing trend on the concentration of TSS was also observed, from 194.5 µg/L (S1) to 24.1 µg/L (S2), suggesting the retention of particles by the system.

Table 2. Water quality indicators (mean and range) measured in influent (S1) and effluent (S2) surface water in the pilot site after its construction, from late summer to autumn 2011.

Parameters	Influent (S1)					Effluent (S2)			
	Units	Min	Max	Mean	SEM	Min	Max	Mean	SEM
T	°C	9.1	17.5	14.1	1.8	8.9	19.2	16.3	2.5
pH	-	7.4	8.2	7.8	0.1	7.1	8.2	7.7	0.2
EC	dS/m	0.7	2.8	1.9	0.4	0.6	2.2	1.1	0.3
TSS	µg/L	2.2	386.8	194.5	192.3	12.4	35.8	24.1	11.7

Notes: SEM: Standard error of mean; T: Temperature; EC: Electrical conductivity; TSS: Total suspended solids.

A comprehensive monitoring was completed between March and November in 2012. Data on climatic conditions (ambient temperature, and total precipitation) during this period are shown in Figure 7. Surface water quality indicators (water temperature, pH, EC, and TSS) in S1 and S2 sampling points along the performed monitoring are shown in Figure 8.

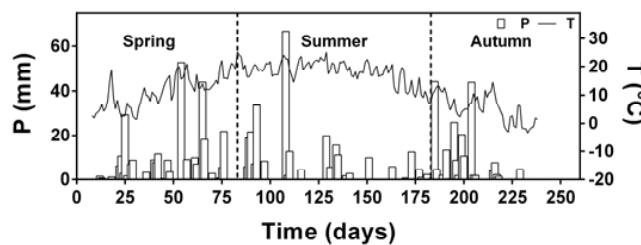


Figure 7. Precipitation and temperature from spring to autumn 2012 (Station Jean Lesage, Quebec).

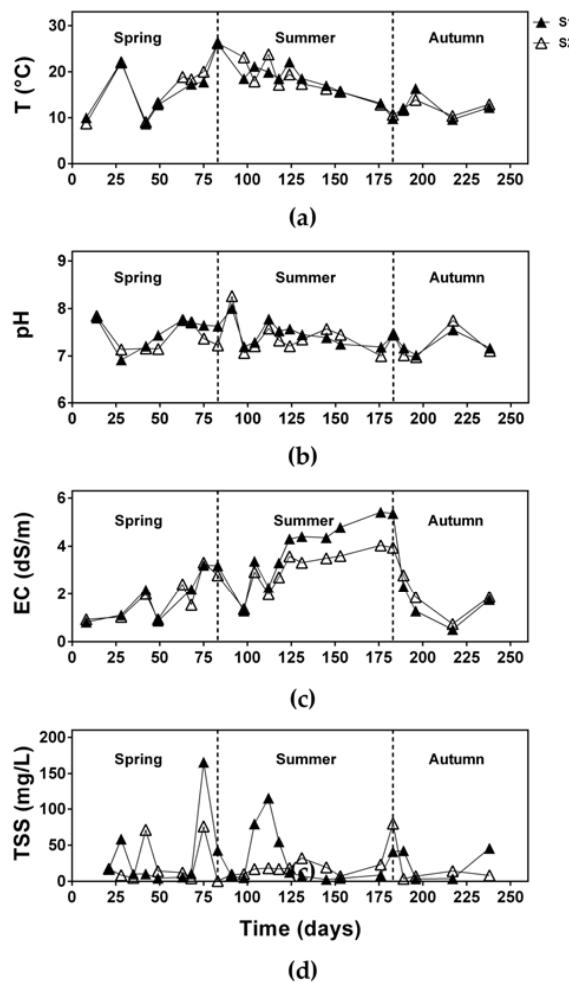


Figure 8. Water quality indicators in influent (S1) and effluent (S2) surface water along the monitoring performed in the pilot site from spring to autumn 2012: (a) temperature (T); (b) pH; (c) electrical conductivity (EC) and (d) total suspended solids (TSS).

Depending on the season, water temperature ranged from 8.6 °C (spring) to 26.2 °C (summer) (Figure 8a), and was generally related to the ambient temperature (Figure 7) with no differences from S1 to S2 sampling points. Water pH values were from neutral to moderately alkaline, between 6.9 and 8.3 (Figure 8b), suitable to permit the existence of most living organisms [32]. No pattern on pH values according to the season was observed, and no differences between influent and effluent water (S1 and S2). The values of EC in surface water (Figure 8c) were highly variable over time, as previously reported for road runoff [5,33]. The EC values ranged from 0.5 to 5.4 dS/m (S1) and from 0.7 to 4.0 dS/m (S2), consistent with the data obtained by Galvez-Cloutier et al. [20] in road side ditches at the same section of the Highway A40. In contrast to that observed for groundwater, in the case of

surface water, the highest EC values were measured in summer and the lowest in both autumn and the early spring. Thus EC values in August and September (3.3–5.4 dS/m) were up to four times higher than in April and May (0.8–2.4 dS/m). Low precipitation, associated with higher ambient temperature, increases salt concentration in water and hence EC. Indeed, this seasonal pattern observed on EC in surface water in both S1 and S2 points (Figure 8c) is consistent with the low total precipitation and high temperatures recorded from mid to late summer (Figure 7). In the fall, EC gradually declined according to the lowest ambient temperature, reaching values similar to those of early spring. Electrical conductivity at S2 was 25% lower than at S1 during August and September, in accordance to that observed in 2011 (Table 2), thus suggesting salt removal by the system during this period.

Regarding particulate matter (Figure 8d), high fluctuations on TSS concentrations were observed in surface water in both S1 and S2 sampling points, with a range from 0.6 mg/L (minimum in S2) to 165.6 mg/L (maximum in S1). Values were low taking into account the traffic intensity of Highway A40 [34] and when compared with the TSS concentration ranges reported for road runoff in other studies: 18–3165 mg/L [33], 13–4800 mg/L [35], and 1–5100 mg/L [2]. Regarding rainfall, a positive relation was observed among the peaks in TSS concentration (Figure 8d) and the peaks in total precipitation (Figure 7), but significant correlations could not be established (Table 3). The concentration of TSS in road runoff is dependent on a number of factors including storms events, rainfall intensity and duration, an antecedent dry weather period, wind, traffic volume and type, traffic during storms, watershed extension, land use in surrounding areas (residential, urban, and agricultural), TSS particle size, and so on [35–38]. The complexity of interactions among these factors may account for the lack of significant correlations between TSS concentration and the climatic data. When comparing the annual average concentration of TSS between S1 and S2 points, a slight overall decrease was observed.

Table 3. Pearson's correlation coefficients among influent (S1) and effluent (S2) surface water parameters.

Sampling Points	Parameters	T	pH	EC	TSS	Cl	Na	Cd	Pb
S1 vs. S1	T	1	0.224	0.169	0.292	0.221	0.308	0.481	-0.069
	pH		1	-0.230	0.149	0.002	-0.063	0.059	-0.293
	EC			1	0.153	0.787 ***	0.880 ***	0.606 **	0.857 ***
	TSS				1	0.092	0.155	-0.120	-0.045
	Cl					1	0.922 ***	0.852 ***	0.611 *
	Na						1	0.766 ***	0.810 ***
	Cd							1	0.481
	Pb								1
S2 vs. S2	T	1	0.094	0.073	-0.321	-0.014	0.122	0.117	0.292
	pH		1	-0.033	-0.025	-0.303	-0.352	-0.282	-0.190
	EC			1	0.358	0.913 ***	0.914 ***	0.670 *	0.810 ***
	TSS				1	0.226	0.227	-0.066	0.140
	Cl					1	0.908 ***	0.757 **	0.701 **
	Na						1	0.775 ***	0.843 ***
	Cd							1	0.514
	Pb								1
S1 vs. S2	T	0.925 ***	-0.034	0.177	-0.383	0.119	0.264	0.426	0.313
	pH	0.335	0.815 **	0.108	0.046	-0.223	-0.201	0.005	-0.083
	EC	0.026	-0.349	0.898 ***	0.384	0.770 ***	0.843 ***	0.726 **	0.807 ***
	TSS	0.372	-0.077	0.118	0.376	0.059	0.233	-0.081	0.189
	Cl	0.109	-0.110	0.852 ***	0.329	0.850 ***	0.871 ***	0.869 ***	0.718 **
	Na	0.157	-0.231	0.902 ***	0.277	0.859 ***	0.918 ***	0.862 ***	0.824 ***
	Cd	0.201	-0.229	0.607 *	-0.188	0.698 **	0.679 **	0.914 ***	0.356
	Pb	-0.132	-0.269	0.820 ***	0.222	0.755 **	0.763 **	0.699 *	0.846 ***

Notes: T: Temperature; EC: Electrical conductivity; TSS: total suspended solids. * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$, statistical significance at these probability levels ($n = 26$).

3.2.2. NaCl Concentration and Removal Efficiency

The concentration of Cl and Na in surface water (S1 and S2 points) along the monitoring performed in the pilot site from spring to autumn 2012 is shown in Figure 9.

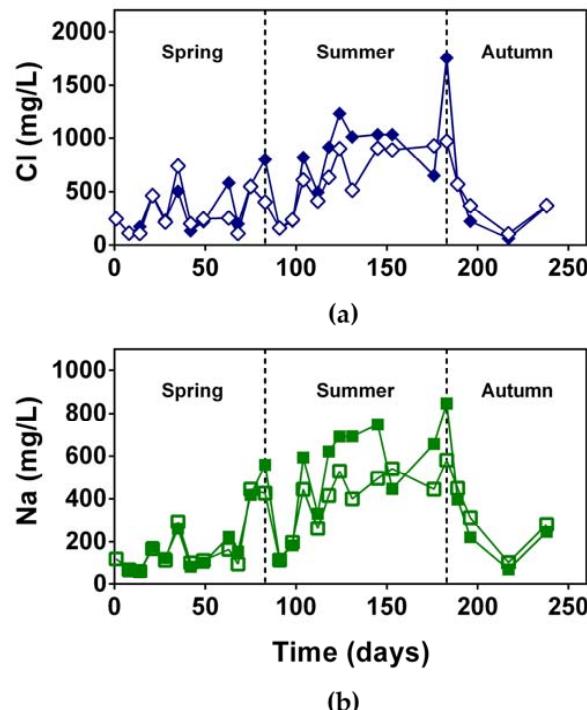


Figure 9. Concentration of Cl and Na in influent (S1) and effluent (S2) surface water along the monitoring performed in the pilot site from spring to autumn 2012: (a) Cl and (b) Na.

Overall, the Cl and Na concentrations measured in influent water (S1) were high and ranged from 63 to 1757 mg/L Cl and from 61 to 846 mg/L Na. Values were in the same magnitude order as the concentrations reported prior to the pilot site construction [20] which suggests that the amount of de-icing salts used on Highway A40 (and in the road network around the highway) during winter road maintenance has not decreased in the last years. Data also revealed a salt pollution problem that may impacts the ecosystem services provided by surface water in the watershed [6]. The extreme salt concentration fluctuations (mostly in summer) can have severe impacts on the aquatic biota [5]. Indeed in 61% of influent water samples both Cl and Na concentrations exceeded the surface water quality criteria established by Quebec Environment Ministry for the prevention of contamination in water and aquatic organisms (250 mg/L Cl, 200 mg/L Na) [39]. Likewise, in 65% of samples, Cl concentrations exceeded the US Environmental Protection Agency chronic effect standard (230 mg/L Cl) and in 23% the acute exposure standard (860 mg/L Cl) for aquatic life in fresh water [40]. Data within the ranges are often reported for road runoff: 17–10,400 mg/L Na [33], 1–298 mg/L Na and 1–573 mg/L Cl [8]. Nonetheless, the very high NaCl concentration fluctuations due to different site-specific and climatic conditions make comparability with other studies limited.

Positive and highly significant correlations ($p < 0.001$) between Cl and Na concentrations and EC values were obtained (Table 3). Indeed, Cl and Na concentrations and EC values in surface water, both in influent (S1) and effluent (S2), showed the same temporal pattern (Figures 8 and 9): increase in the mid and late summer, then decrease in autumn. Seasonal NaCl concentration ranges (both S1 and S2) were as follows: 61–559 mg/L Na and 101–799 mg/L Cl (spring) < 114–846 mg/L Na and 158–1757 mg/L Cl (summer) > 71–449 mg/L Na and 63–569 mg/L Cl (autumn). Higher NaCl concentrations in winter during de-icing salt applications [5,33] or in spring after snow-melting [41] are

often expected. However, data from the present study showed a clear increase of NaCl concentration in summer. The temporal pattern of NaCl concentration is linked to climatic conditions: the highest concentrations of NaCl in water at higher temperature and lower precipitation, i.e., higher temperature/precipitation ratio, as occurs in summer [42]. Indeed, as discussed above for EC, evaporation processes occurring during summer involve a decrease in surface water flow and then an increase in salt concentration in water. Therefore, our study suggests that high Cl and Na concentrations may be expected in the Saint-Augustin Lake watershed in summer. Chloride soluble concentrations in surface water were generally higher than Na concentrations (2–3 fold) (Figure 9). The Cl ions are very soluble and less favourable to be adsorbed onto soil colloids than Na ions which can be adsorbed due to cation exchange reactions by displacing cations such as Ca^{2+} and Mg^{2+} [42,43]. The different affinity of Cl and Na for particle sorption may account for the molar imbalance observed between Cl and Na in surface water.

The seasonal performance of the pilot site (including detention basin, active filtering bed, and adapted constructed wetland units) was evaluated by calculating the Cl and Na removal efficiency considering the concentration difference between influent (S1) and effluent (S2) surface water (Equation (1)). The seasonal removal efficiency was as follows: 16% Cl and 10.6% Na (spring), 18.1% Cl and 24.5% Na (summer), and 19.7% Cl and 3% Na (autumn). Data corresponds to the first year of operation (2012). Higher performance is expected in the subsequent years with the maturation of the system due to growth and development of vegetation and the increase of micro- and macroorganism biomass and activity, as previously reported in adapted constructed wetland under cold climate [44]. Data showed a different removal pattern depending on the element, Cl or Na. Thus Cl removal remained almost constant over the pilot site operational period. However, in the case of Na, the performance of the system was higher during summer, followed by spring, which deserves further studies.

3.2.3. Trace Metal Concentration and Removal Efficiency

The concentration of dissolved Cd and Pb in surface water (S1 and S2 points) along the monitoring performed from spring to summer 2012 is shown in Figure 10.

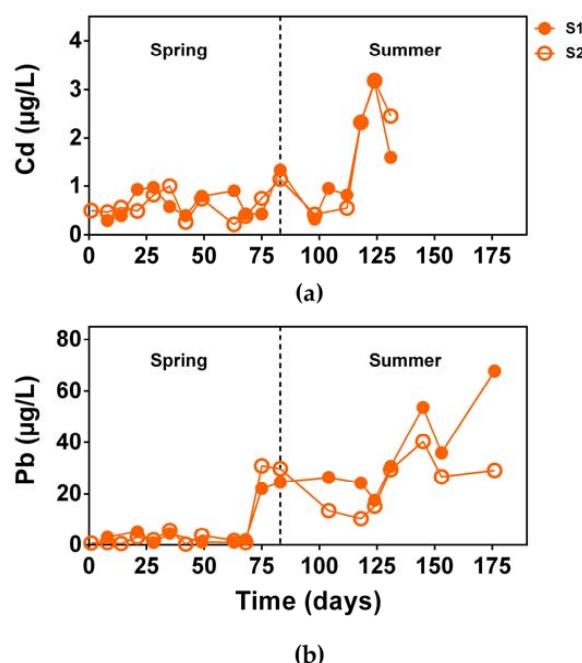


Figure 10. Concentration of trace metals in influent (S1) and effluent (S2) surface water along the monitoring performed in the pilot site from spring to summer 2012: (a) Cd and (b) Pb.

Cadmium and Pb concentration in autumn was not presented because it was below detection limit. Trace metal concentration in both S1 and S2 points ranged as follows: 0.2–3.2 µg/L Cd (0.98 µg/L mean) and 0.3–67.8 µg/L Pb (16.6 µg/L mean). In all cases, Cd concentration was under the surface water quality criteria established by the Quebec Environment Ministry for the prevention of contamination in water and aquatic organisms (5 µg/L Cd) [39]. In contrast, in 56% of water samples, Pb concentrations exceeded the same quality criteria (10 µg/L Pb). However, according to USEPA [40], in 97% (Cd) and 71% (Pb) of water samples the chronic effect standard for aquatic life in fresh water (0.25 µg/L Cd and 2.5 µg/L Pb) was exceeded, as well as in 15% (Cd) and 3% (Pb) of the acute exposure standard (2 µg/L Cd and 65 µg/L Pb). The ranges of Cd and Pb concentration quantified in the present study were expectedly high because of the high daily traffic density in the Highway A40. High metal concentrations can also be related to the high amount of de-icing salts spread [45,46] and the increased tear, wear, and corrosion due to the application of gravel at cold weather conditions [33]. Values are in line with those reported in other studies in road runoff: 0.02–6.1 µg/L dissolved Cd (0.2 µg/L mean) and 0.2–414 µg/L dissolved Pb (5.4 µg/L mean) in urban and non-urban highways [2]; 1.5–72.3 µg/L total Pb in urban highways during storm events [47]; 400 µg/L event mean Pb concentration in highways with >30,000 vehicles/day [48].

Significant positive correlations among Cd, Pb, Na, and Cl concentrations were obtained ($p < 0.001$ Cd and Pb vs. Na, $p < 0.01$ Cd and Pb vs. Cl) (Table 3). Indeed, temporal patterns in Cd, Pb, Na, and Cl concentrations in surface water (both in S1 and S2) were similar: increase from mid to late summer, then a decrease in autumn (Figures 9 and 10). As discussed above, the higher temperature/precipitation ratio occurring in summer can explain this result (Figure 7). The positive correlations may also be related to metal mobilization processes by the formation of metallic chlorocomplexes, favoured at pHs of the present study [41,49]. In this regard, Bäckström et al. [11] studied the seasonal variations of trace metals in soil solution as a function of the distance from the road in Sweden. They observed that the dominant speciation of Cd and Pb at 4 m from the road was: (i) Cd^{2+} , CdCl^+ and CdCl_2 (for Cd); and (ii) Pb^{2+} , PbCl^+ and PbCO_3 (for Pb). In road runoff, particulate matter is considered as one of the major pollutants since many pollutants are attached to it and washed off [37]. In the present study, significant Pearson correlations between Cd and Pb concentrations and TSS were not found (nor between TSS and Cl and Na), even taking into account each season separately (Table 3). Despite Cd and Pb in road runoff are often associated with suspended solids, Cd and Pb adsorption can be far different depending on the fraction sizes, peaking at the fine particles (<40 µm) [48]. In this regard, climatic conditions could play an important role. Westerlund et al. [50] observed that metals during snow-melting were more particulate bound than during the rainy periods with a higher percentage of the dissolved fraction. Nevertheless, at low ambient temperature the accumulation of metals in particles decreases [49]. Further studies considering the seasonal pattern of TSS particle size and trace metal distribution could be of great interest.

The seasonal performance of the pilot site was evaluated by Cd and Pb removal efficiency (Equation (1)). The seasonal removal efficiency in the first year of operation was as follows: 10.3% Cd and 6.8% Pb (spring), and –6.7% Cd and 35.8% Pb (summer). As for NaCl, higher performance is expected in the subsequent years. The negative removal rate of Cd in summer could be the result of pollutants entering the pilot site outlet, considering its proximity to the Highway A40 [34,47].

4. Conclusions

From this research project, conclusions about the groundwater and surface water monitoring conducted in the Saint-Augustin Lake watershed can be established. The concentrations of Cl and Na in groundwater were higher in study wells (affected by road network) than in control wells. Our findings suggest therefore that de-icing salts spread in the watershed roads impact the groundwater chemical composition. Regarding surface water, the pilot site (composed of a detention basin, an active filtering bed and an adapted constructed wetland) has been proposed as a solution to reduce the concentrations of NaCl, as well as trace metals (Cd and Pb), at a watershed scale. Monitoring of

water surface showed that both NaCl concentrations and EC values follow a seasonal dependence. Thus, NaCl concentrations and EC values increased in mid and late summer (linked with the lowest precipitation and highest temperature recorded), and then decrease in autumn. The removal efficiency of Na by the pilot site was also dependent of the season, being the highest in summer, but constant in the case of Cl. Cadmium and Pb concentrations were consistent with the traffic density and followed a similar temporal pattern to that observed for NaCl and EC. The highest Pb removal was found in summer. The pilot site has shown to be a useful strategy to mitigate the impact of anthropogenic salinity at watershed scale, although it would take a few years of operation to observe its optimal performance. It should also be considered that the data are limited as an aggregate comparison of upstream/downstream of the pilot site, which is comprised of three units. This study deserves to be pursued and taken further in order to complete the data and to determine long-term conclusions.

Acknowledgments: The authors especially wish to thank Luke Pettet for English revisions. We thank the editor and the anonymous referees for helpful suggestions.

Author Contributions: Rosa Galvez conceived and designed the experiments; Hamdi Messaoud, Arthur Michaux and Samuel Roy performed the experiments; Gaëlle Guesdon, Ana de Santiago-Martín and Sébastien Raymond analyzed the data and wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest.

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