Multiple land use activities drive riverine salinization in a large, semi-arid river basin in western Canada

Jason G. Kerr*
Alberta Environment and Parks, Environmental Monitoring and Science Division, Calgary, Alberta, Canada

Abstract

Salinization is increasingly recognized as a global issue. However, the relative importance of different drivers across a broad range of ions and ecosystems is not well understood. This study examined spatial and temporal dynamics in riverine salinity (conductivity, Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$, SO$_4^{2-}$, Cl$^-$ and HCO$_3^-$) in the South Saskatchewan River Basin (SSRB), a semi-arid, mixed land use watershed in Alberta, Canada. A significant temporal increase ($p<0.05$) in the concentration of one or more ions was observed at all 12 study sites. While all ions exhibited a significant increase in concentration over time, the rate of change was generally highest for Cl$^-$ ($\approx 1.4$–$3.0\%$ yr$^{-1}$). The observed increase in riverine Cl$^-$ loading downstream of a large urban center ($\approx 1700$ tonnes yr$^{-1}$) was attributed to increasing inputs from road salt ($\approx 1800$ tonnes yr$^{-1}$) and to a lesser extent municipal wastewater ($\approx 400$ tonnes yr$^{-1}$). For most other salts, spatial variation was driven not by urbanization but by the proportion of salt affected soils and/or cropland. A distinct Na$_2$SO$_4$ signal was observed at stations draining salt affected soils which strengthened over time at 7/12 sites indicating temporal trends in Na$^+$ and SO$_4^{2-}$ have been driven largely by soil processes. A strong relationship between crop-land and salt chemistry across the basin suggests agricultural activities have also contributed to observed trends. Therefore, in regions with similar climatic and anthropogenic characteristics to the SSRB, multiple stressors are likely to be operating and as such, these systems may be at particular risk from salinization.

Freshwaters naturally contain dissolved salts. However, if the concentration of dissolved salts increases due to one or more anthropogenic activities this process is commonly termed secondary salinization (Williams 2001). Secondary salinization is widely recognized as a significant threat to the sustainability of freshwater ecosystems (Williams 2001; Kaushal et al. 2005; Novotny et al. 2009; Cañedo-Argüelles et al. 2013; Herbert et al. 2015). The salinity of freshwater ecosystems is generally determined by the concentration of calcium (Ca$^{2+}$), magnesium (Mg$^{2+}$), potassium (K$^+$), sodium (Na$^+$), chloride (Cl$^-$), sulphate (SO$_4^{2-}$), carbonate (CO$_3^{2-}$) and bicarbonate (HCO$_3^-$) ions (Cañedo-Argüelles et al. 2013). Temporal increases in one or more of these ions (or some measure of overall salinity) have been reported in lakes (Rosfjord et al. 2007; Likens and Buso 2009; Müller and Gächter 2011; Winter et al. 2011), and in streams and rivers (Jolly et al. 2001; Parr and Mason 2003; Thunqvist 2004; Kaushal et al. 2005; Corsi et al. 2015). Increased salinity in freshwater ecosystems has been linked to a number of deleterious impacts including shifts in aquatic community composition (Collins and Russell 2009; Bâthe and Coring 2011; Porter-Goff et al. 2013), alteration of biogeochemical processes (Löfgren 2001; Swan and DePalma 2011), and the degradation of potable water supplies (Kaushal 2016). Importantly, once freshwater systems become salinized, mitigation can be costly and time consuming (Novotny and Stefan 2010; Jin et al. 2011). Therefore, early detection of shifts in riverine salt chemistry and the identification of key drivers of these changes are critical.

Secondary salinization of freshwater ecosystems is driven by a range of anthropogenic activities, some of which are largely climate specific. Historically, salinization was viewed as a problem confined mostly to arid and semi-arid environments (Williams 2001). In particular, changes to natural flow regimes in support of urban and/or agricultural developments have increased salinity in semi-arid waters (Nielsen et al. 2003; Vengosh 2014). In addition, the clearing of deep rooted native vegetation and replacement with annual crops or pastures has resulted in rising groundwater tables and subsequent accumulation of salts at the soil surface (Jolly et al. 2001; Parr and Mason 2003; Thunqvist 2004; Kaushal et al. 2005; Corsi et al. 2015). Increased salinity in freshwater ecosystems has been linked to a number of deleterious impacts including shifts in aquatic community composition (Collins and Russell 2009; Bâthe and Coring 2011; Porter-Goff et al. 2013), alteration of biogeochemical processes (Löfgren 2001; Swan and DePalma 2011), and the degradation of potable water supplies (Kaushal 2016). Importantly, once freshwater systems become salinized, mitigation can be costly and time consuming (Novotny and Stefan 2010; Jin et al. 2011). Therefore, early detection of shifts in riverine salt chemistry and the identification of key drivers of these changes are critical.

*Correspondence: jason.kerr@gov.ab.ca

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This process, sometimes referred to as dryland salinity, has led to the transport of soluble salts in runoff and/or groundwater flow and the subsequent salinization of dryland rivers (Williams 2001). Irrigation of semi-arid croplands has also increased salts at or near the soil surface through the evaporation of irrigation water, and/or the recharge of saline groundwater (Houk et al. 2006; Vengosh 2014). Importantly, this has resulted in the salinization of rivers via irrigation return flows (Causapé et al. 2006).

There is increasing evidence that the application of road salt for winter road maintenance is a major cause of salinization in cold, humid surface waters (Godwin et al. 2003; Thunqvist 2004; Rosfjord et al. 2007; Kelly et al. 2008; Novotny et al. 2009; Müller and Gächter 2011). The most common de-icing agent used for winter road maintenance in North America is NaCl and its use has increased significantly in the last 50 yr to 60 yr (Jackson and Jobbágy 2005; Ramakrishna and Viraraghavan 2005). Within the USA, the majority of road salt usage occurs in the northeastern states (Jackson and Jobbágy 2005), while densely populated regions of Ontario and Quebec have the highest usage within Canada (Environment Canada 2001). As such, most of the research within North America relating road salt application to salinization of freshwaters has occurred within NE USA, and to a lesser extent SE Canada. These studies demonstrate a link between increased Na$^+$ and/or Cl$^-$ concentrations and the application of road salt (Kaushal et al. 2005; Kelly et al. 2008; Novotny et al. 2008; Winter et al. 2011; Perera et al. 2013; Corsi et al. 2015). Importantly, for a number of waterbodies the concentration of Cl$^-$ exceeded critical thresholds for the protection of aquatic health (Kaushal et al. 2005; Trowbridge et al. 2010).

In addition to the climate specific drivers of salinization described above, municipal and/or industrial wastewater effluents (Cortecce et al. 2002; Parr and Mason 2003; Aitkenhead-Peterson et al. 2010; Müller and Gächter 2011), oil and gas activities (Bern et al. 2015), and mining (Bätte and Coring 2011; Zipper et al. 2016) are additional sources of salinity to rivers. Finally, changes in precipitation and/or evapotranspiration as a result of climate change may further accelerate riverine salinization (Cañedo-Argüelles et al. 2013). Therefore, rather than a problem confined mostly to semi-arid and arid regions, salinization is increasingly seen as a global issue (Cañedo-Argüelles et al. 2013; Kaushal 2016) with the dominant drivers of salinization dependent in large part on climatic regime. However, while semi-arid regions appear to be at greatest risk from water withdrawals and agricultural pressures, and cold, humid environments at greatest risk from road salt application, we know very little about systems in which both of these conditions exist (i.e., semi-arid environments which receive a substantial proportion of winter precipitation as snow). Understanding the extent of salinization in these systems and the relative importance of different drivers is critical if we are to understand the global reach of salinization, and the relative importance of multiple stressors under different hydroclimatic and disturbance regimes. Therefore, this study will examine spatial and temporal dynamics in riverine salt concentration and composition in a cold, semi-arid river basin subject to multiple land-use pressures. It is hypothesized that salinization in these systems will be driven by multiple causes and characterized by a number of associated chemical signatures. The overall aim of this work is to provide new insights into how multiple land use pressures operate at the basin scale to alter the concentration and composition of riverine salts.

**Methods**

The South Saskatchewan River Basin

The South Saskatchewan River Basin (SSRB) is located in western Canada and encompasses an area of 172,900 km$^2$. The basin extends from the Continental Divide in Alberta’s Rocky Mountains, through to south-central Saskatchewan where it merges with the North Saskatchewan River to form the Saskatchewan River. For the purpose of this study, only the area located within the province of Alberta is included. Within Alberta, the SSRB encompasses an area of approximately 121,100 km$^2$ and includes three major sub-watersheds; the Bow River Watershed, the Oldman River Watershed, and the Red Deer River Watershed. Within Alberta, the Bow and Oldman Rivers flow into the South Saskatchewan River while the Red Deer River joins the South Saskatchewan River to the east of the provincial border with Saskatchewan (Fig. 1). Discharge to major river systems in the SSRB originates from melting snowpack in the Rocky Mountains which typically contribute about 70% of annual river flow (Tanzeeba and Gan 2012). Peak river discharge generally occurs in late May to early June. While the SSRB contains a number of different eco-regions, the majority of the basin is grasslands (Downing and Pettapiece 2006). The climate is predominately semi-arid (annual precipitation $\approx 435$ mm) with higher levels of precipitation falling mostly as snow in the Rocky Mountains and lower levels (median annual precipitation $\approx 270$ mm) in the dry mixed grasslands. In much of the basin evapotranspiration exceeds precipitation (AMEC 2009). The region is characterized by cold winters and short summers with a mean annual temperature of approximately $+3^\circ$C (Downing and Pettapiece 2006).

Due to water withdrawals from the basin’s major river systems, the SSRB is considered to be one of Canada’s most threatened watersheds (Swainson 2009; Sauchyn et al. 2016). Water allocation to meet increasing municipal, industrial, and agricultural needs has led to large reductions in natural river flows (Schindler and Donahue 2006). More than 70% of water withdrawals in the SSRB are used to supply water to 13 irrigation districts, all of which are located within the SSRB (Nicol and Klein 2006). The main types of salt affected soils in the SSRB are saline and sodic soils (Harker and
Fig. 1. Location of major rivers and long-term river network (LTRN) stations within the SSRB.
Most sodic soils in Alberta are classified as solonetzic and a large region of solonetzic soil extends across the eastern grasslands of the SSRB (Miller and Brierley 2011). While the basin contains naturally saline and sodic soils, land clearing and subsequent increases in groundwater recharge (i.e., dryland salinity) has led to the formation of salt seeps throughout the region (Greenlee et al. 1968; Wiebe et al. 2007). The SSRB includes a number of major urban centers, the largest of which are the city of Calgary with a population of almost 1.2 million and the smaller cities of Red Deer, Lethbridge, and Medicine Hat with populations ranging from 60,000–100,000 (Alberta Municipal Affairs 2014). Average annual snowfall in these four major cities ranges from 60,000–100,000 (Alberta Municipal Affairs 2014). Average annual snowfall in these four major cities ranges from 60,000–100,000 (Alberta Municipal Affairs 2014). Average annual snowfall in these four major cities ranges from 60,000–100,000 (Alberta Municipal Affairs 2014). Average annual snowfall in these four major cities ranges from 60,000–100,000 (Alberta Municipal Affairs 2014). Average annual snowfall in these four major cities ranges from 60,000–100,000 (Alberta Municipal Affairs 2014).

Data analyses

To investigate salinization in the SSRB, long-term records of salt chemistry (conductivity, Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$, SO$_4^{2-}$, HCO$_3^-$ and Cl$^-$) in the SSRB’s major river systems were analyzed for spatial patterns and temporal trends. To enable comparison of the relative contribution of individual ions to overall salinity, all concentration data were converted to equivalents. In addition to analyses of ion concentrations, several key ion ratios (e.g., Na:Cl and sodium adsorption ratio [SAR]) were calculated to determine temporal and spatial changes in salt composition. The SAR was derived from the concentration of Na$^+$, Ca$^{2+}$, and Mg$^{2+}$ (meq L$^{-1}$) using the following equation:

$$\text{SAR} = \frac{[\text{Na}^+]}{\sqrt{([\text{Ca}^{2+}] + [\text{Mg}^{2+}])] \times 0.5}$$

The SAR is commonly used to assess the suitability of water for irrigation and in particular the risks associated with sodic soils (Jackson and Reddy 2007). All raw chemistry data were obtained from the Alberta Environment and Parks long-term river network (LTRN). Monthly water quality data were analyzed from 13 stations across five rivers sampled year round (Fig. 1). Although the duration of record varied among stations, most stations had approximately 30 yr of continuous data and with the exception of RDR4, all stations had a minimum of 15 yr of data (Supporting Information Table 1). Analytical methodologies used as part of the LTRN program over the past 40 yr are summarized in Supporting Information Table 2. When changes in analytical methodologies were made, data were assessed for evidence of a change in concentration related to the introduction of the new protocol. First, data were compared between the pre- ($n=12$) and post-change ($n=12$) periods graphically and statistically. Additionally, data were assessed for any synchronicity of change across multiple stations which would be expected to occur if changes in concentration were due to a change in analytical methodology. Because changes in analytical methodology occurred at different times for different analytes, pre- and post-change data were also assessed for any deviations in the cation to anion balance. In addition, synchronicity among related ions was examined to determine if shifts in the concentration of a particular anion/cation following a change in methodology was accompanied by a concurrent shift in a related cation/anion which was not subject to a change in analytical methodology. Based on the above criteria, there was no evidence of shifts in concentration due to method changes and therefore all data were merged prior to analysis.

To investigate spatial variability in salinity across the SSRB, a number of approaches were used. For the purpose of comparing salt chemistry among stations, a uniform period of time (2000–2015) was used in the calculation and comparison of all summary statistics. The exception to this was RDR4 which only had data for the period 2007–2015. The degree of similarity in salt chemistry among stations was first examined graphically by employing an ALSCAL multidimensional scaling (MDS) analysis. For the MDS analysis, the median concentrations of major ions (i.e., Ca$^{2+}$, Mg$^{2+}$, Na$^+$, K$^+$, SO$_4^{2-}$, Cl$^-$, and HCO$_3^-$) were first transformed to z scores to account for differences in concentrations among ions. Transformed data at each station were used to create Euclidian distance measures of dissimilarity among all 13 LTRN stations and plotted to examine if, and to what extent, stations clustered based on similarities in salt chemistry. To examine the influence of land use on salt chemistry, the median concentration of each ion at each station was analyzed as a function of land use and soil variables. The percent watershed area of urban land, cropland, saline soil, and solonetzic soil were calculated for each station and the spatial variation in salt chemistry was then analyzed as a function of these variables by linear regression.

To determine the degree of seasonality in the concentration of cations and anions, a seasonality index was calculated for each ion at each station. The seasonality index is defined here as the ratio of ice season (Nov–Mar) to open water season (Apr–Oct) concentration. Temporal trends in salt concentrations were assessed using a seasonal Mann-Kendall test (Helsel et al. 2006). This method has the advantage of being robust against seasonality, non-normality, missing values and serial correlation (Hirsch and Slack 1984). To account for the potential effect of discharge on temporal trends, all trend tests were performed on flow adjusted data. All discharge data were obtained from Water Survey of Canada hydrometric stations (Water Survey of Canada, www.wateroffice.ec.gc.ca) located at or near each of the LTRN sites. For all trend analysis, only sites with a minimum of 15 yr of data were included (i.e., all stations except RDR4).

To investigate mass fluxes of Cl$^-$ across the SSRB, Cl$^-$ loads were calculated at the mouth of each of the major watersheds.
Fig. 2. Median concentration ± 1 SD for conductivity and major anions and cations in river water at 13 LTRN stations within the SSRB.
(ER1, BR4, OMR3, RDR4, and SSR1). Cl\textsuperscript{–} loads were estimated by the composite method (Aulenbach and Hooper 2006) using monthly concentration data and daily discharge data at each station. Briefly, the composite method combines two common approaches: the regression model method and the period-weighted method. Concentration vs. discharge relationships were first used to derive daily estimates of load. The residuals of these relationships (i.e., actual load from monthly chemistry and discharge measurements vs. modelled load) were then used to adjust all modelled data using a method akin to the mid-point method described elsewhere (Kerr et al. 2016). To examine the effect of urban inputs from the City of Calgary on Cl\textsuperscript{–} loads in the Bow River, Cl\textsuperscript{–} loads from wastewater treatment plant (WWTP) effluent and road salt application were compared with increases in Cl\textsuperscript{–} loads downstream of Calgary (i.e., BR1 vs. BR2) and temporal trends at the mouth of the Bow River (BR4). WWTP and road salt data were provided by the City of Calgary.

Results

Spatial variability in riverine salinity in the SSRB

The ion composition of rivers in the SSRB was dominated by Ca\textsuperscript{2+} and HCO\textsubscript{3}\textsuperscript{–} which made up 48–66% and 63–83% of cation and anion charge, respectively. Together with Mg\textsuperscript{2+}, which made up between 30–32% of cation charge, the concentrations of Ca\textsuperscript{2+} and HCO\textsubscript{3}\textsuperscript{–} did not vary substantially among stations relative to other ions (Fig. 2). Conversely, Na\textsuperscript{+} and Cl\textsuperscript{–} generally made up a small proportion of overall ionic charge (i.e., 3–28% and 1–8% of overall cation and anion charge, respectively) but varied considerably among stations (Fig. 2). In terms of Cl\textsuperscript{–}, stations within (ER1) or downstream of Calgary (BR2–4 and SSR1) exhibited relatively high concentrations. This is reflected in the MDS plot which shows a distinct clustering of these stations along the vertical axis (Dimension 2) (Fig. 3). Increasingly positive values along this axis were correlated with increases in Cl\textsuperscript{–} concentration at each station ($r^2 = 0.58$) indicating that these stations are distinguished from other sites primarily by elevated Cl\textsuperscript{–}. High Cl\textsuperscript{–} concentrations relative to other ions at these stations also resulted in lower Na:Cl ratios and a higher proportion of anion charge as Cl\textsuperscript{–} (Fig. 4a,b). Furthermore, relative to other ions and stations, Cl\textsuperscript{–} seasonality ($C_{ice}/C_{open}$) was high within and downstream of Calgary (Table 1). In terms of land use drivers of spatial variability in Cl\textsuperscript{–} concentration, neither saline soils, solonetzic soils nor percent cropland were important. Conversely, as the proportion of urban land at each station increased there was an increase in median Cl\textsuperscript{–} concentration ($p < 0.05; r^2 = 0.73$) and a decrease in the Na:Cl ratio ($p < 0.05; r^2 = 0.45$) (Fig. 5). In addition, as urban land use increased, the proportion of anion charge as Cl\textsuperscript{–} (Cl\textsuperscript{–}/SO\textsubscript{4}\textsuperscript{2–} + HCO\textsubscript{3}\textsuperscript{–}) also increased ($p < 0.05; r^2 = 0.84$). Urban land use was not correlated with spatial variability in any other ions or ratios.

For several ions (i.e., Na\textsuperscript{+}, SO\textsubscript{4}\textsuperscript{2–}, Mg\textsuperscript{2+}, K\textsuperscript{+}), concentration increased with distance downstream (Fig. 2). This is reflected in the distribution of stations along the horizontal axis (Dimension 1) of the MDS plot (Fig. 3). A high degree of similarity between the two most upstream stations (OMR1 and BR1) was observed and in rivers with multiple stations, increasingly negative values along the horizontal axis were observed with distance downstream (e.g., OMR1 < OMR2 < OMR3). Increasingly negative values along this axis were correlated with increases in Mg\textsuperscript{2+} ($r^2 = 0.94$), K\textsuperscript{+} ($r^2 = 0.80$), Na\textsuperscript{+} ($r^2 = 0.74$) and SO\textsubscript{4}\textsuperscript{2–} ($r^2 = 0.76$) and reflect an overall shift from lower to higher conductivity among stations. Longitudinal...
increases in Na\(^+\) and SO\(_4^{2-}\) coupled with the relative stability of Ca\(^{2+}\) and HCO\(_3^-\) produced shifts toward higher SAR and to a lesser extent a higher proportion of anions as SO\(_4^{2-}\) (SO\(_4^{2-}\)/Cl\(^-\) + HCO\(_3^-\)) in downstream vs. upstream stations (Fig. 4c,d). Spatial variability in Na\(^+\), K\(^+\), SO\(_4^{2-}\), Mg\(^{2+}\), and SAR were related to the proportion of saline and/or solonetzic soils at each station (Fig. 5). Because HCO\(_3^-\) concentration did not change as a function of soil salinity (p > 0.05; \(r^2 = 0.01\)), the proportion of anion charge as SO\(_4^{2-}\) (SO\(_4^{2-}\)/Cl\(^-\) + HCO\(_3^-\)) also increased as a function of percent saline soil at each station (p < 0.05; \(r^2 = 0.55\)). The proportion of land associated with crop farming was also an important driver of spatial variability. As the percentage of cropland increased at each station, the concentrations of Na\(^+\), Mg\(^{2+}\), K\(^+\), HCO\(_3^-\), and SAR also increased (Fig. 5). With the exception of Mg\(^{2+}\), for parameters where saline/solonetzic soils and cropland were both related to spatial variability (i.e., Mg\(^{2+}\), Na\(^+\), K\(^+\), and SAR), multiple step-wise regressions indicated that more variation was explained by the combined effect of cropland and saline or solonetzic soils (Supporting Information Table 3). For Mg\(^{2+}\), cropland alone was the best predictor of concentration.

**Temporal trends in riverine salinity in the SSRB**

Significant temporal increases in flow adjusted concentrations were observed across all stations and ions. In terms of relationship to flow, ions generally exhibited a negative relationship with discharge (Supporting Information Table 4) and discharge did not decrease during the study period (data not shown). The number of individual salts exhibiting a significant increase in concentration at each station ranged from two salts at OMR1 (Ca\(^{2+}\) and Cl\(^-\)) through to all seven salts at BR2–4 and SSR1 (Table 2 and Supporting Information Table 5). Of the seven major anions and cations, only Cl\(^-\) increased significantly at all 12 stations. The rate of change in concentration (\(\mu\text{eq L}^{-1}\ \text{yr}^{-1}\)) was generally highest for Ca\(^{2+}\) but as a percentage of median concentration (% yr\(^{-1}\)), the rate of change in Cl\(^-\) concentration was frequently greater than other salts (Table 2). Although generally lower

![Fig. 4. Median (± 1SD) Cl\(^-\)/SO\(_4^{2-}\) + HCO\(_3^-\) (a), Na : Cl (b), sodium adsorption ratio (SAR) (c), and SO\(_4^{2-}\)/Cl\(^-\) + HCO\(_3^-\) (d) in river water at LTRN stations.](image-url)
Table 1. Seasonality index (C_{Na}−C_{open}) for major anions and cations at LTRN stations within the SSRB.

| Site | Cond | Na⁺ | Mg²⁺ | K⁺ | Ca²⁺ | SO₄²⁻ | Cl⁻ | HCO₃⁻ |
|------|------|-----|------|-----|------|--------|-----|--------|
| BR1  | 1.14 | 1.02| 1.13 | 1.00| 1.17 | 1.22   | 1.05| 1.07   |
| BR2  | 1.22 | 1.42| 1.13 | 1.36| 1.15 | 1.28   | 1.70| 1.12   |
| BR3  | 1.21 | 1.38| 1.08 | 1.23| 1.17 | 1.23   | 1.91| 1.13   |
| BR4  | 1.18 | 1.04| 1.06 | 1.11| 1.24 | 1.04   | 1.63| 1.17   |
| ER1  | 1.25 | 1.54| 1.19 | 1.20| 1.26 | 1.33   | 1.63| 1.21   |
| OMR1 | 1.17 | 1.23| 1.18 | 1.09| 1.19 | 1.37   | 1.36| 1.14   |
| OMR2 | 1.18 | 1.21| 1.14 | 0.92| 1.16 | 1.27   | 1.15| 1.15   |
| OMR3 | 1.23 | 1.37| 1.15 | 1.11| 1.20 | 1.32   | 1.54| 1.19   |
| SSR1 | 1.22 | 1.12| 1.08 | 1.11| 1.28 | 1.14   | 1.95| 1.18   |
| RDR1 | 1.21 | 1.01| 1.21 | 0.75| 1.24 | 1.32   | 0.71| 1.18   |
| RDR2 | 1.31 | 1.25| 1.23 | 0.89| 1.30 | 1.37   | 1.27| 1.32   |
| RDR3 | 1.31 | 1.23| 1.30 | 0.90| 1.41 | 1.41   | 1.36| 1.32   |
| RDR4 | 1.21 | 0.85| 1.28 | 0.79| 1.39 | 1.10   | 1.09| 1.33   |

Discussion

This study documents the widespread salinization of a cold, semi-arid river basin in western Canada. This is important given the paucity of data on large river systems in general, and more specifically for rivers within western Canada and across semi-arid regions subject to substantial snowfall. In this regard, results from the SSRB support the growing consensus that salinization is a global threat to freshwater ecosystems (Kausral et al. 2005; Cañeddo-Argüelles et al. 2013; Herbert et al. 2015). In terms of what drives salinization in the SSRB, the study also provides important insights. A clear shift away from a Ca(HCO₃)₂ dominated system was observed. The prevalence of Ca²⁺ and HCO₃⁻ in the system is driven by the dissolution of carbonate minerals in the headwaters of the SSRB (Grasby and Hutcheon 2000) and therefore represent natural or background conditions. Importantly, the observed longitudinal and temporal shifts away from background conditions appear to be driven by a number of different anthropogenic stressors. This differs from many other systems where salinization has been attributed primarily to a single driver (Jolly et al. 2001; Godwin et al. 2003; Kelly et al. 2008; Bern et al. 2015; Corsi et al. 2015; Zipper et al. 2016). As such, the SSRB, and systems with comparable climatic and anthropogenic characteristics, face a unique set of pressures and management challenges in terms of riverine salinization. Furthermore, results from the SSRB highlight the importance of considering a range of potential sources when examining the impact of anthropogenic activities on riverine salinity in other systems.

Road salt as a driver of riverine salinization in the SSRB

Multiple lines of evidence indicate an important role for urbanization, and more specifically road salt, as a driver of salinization in the SSRB. The relationship between urban land area and Cl⁻ concentrations across the basin, together with the observation that Cl⁻ fluxes were substantially higher in the more urbanized watersheds (BR and ER), indicate an urban effect. Furthermore, estimates of urban Cl⁻ inputs clearly demonstrate that road salt, and to a lesser extent WWTPs, are the primary drivers of increased Cl⁻ at stations within or downstream of Calgary. In addition, the relatively high degree of seasonality for Cl⁻ at these stations is indicative of seasonal inputs from road salt runoff (Kaushal et al. 2005; Novotny et al. 2008; Gardner and Royer 2010). A final line of evidence for road salt as a driver of Cl⁻ trends in these stations was the Na:Cl ratios. If the primary source of Na⁺ and Cl⁻ in water is halite (NaCl), the molar ratio of Na⁺ to Cl⁻ should be close to 1:1 (Godwin et al. 2003; Novotny et al. 2008; Jin et al. 2011) and this was clearly observed at ER1 and stations downstream of Calgary. Furthermore, a clear link between urban area and decreasing Na:Cl was observed across the basin. Finally, at all stations Na:Cl decreased toward a 1:1 ratio over time indicating that the proportion of Na⁺ associated with Cl⁻ has increased. This is again consistent with increasing inputs from road salt.
Relative to stations within or downstream of Calgary (i.e., ER1, BR2–4, and SSR1), the weight of evidence for a road salt effect in the RDR and OMR was weaker and the overall impact appeared to be relatively small. Increased Na\(^+\) and Cl\(^-\) concentrations downstream of the cities of Red Deer (RD1 vs. RDR2) and Lethbridge (OMR1 vs. OMR3) are consistent with inputs from WWTPs and/or road salt. However, temporal increases in Na\(^+\) and Cl\(^-\) upstream of these urban centers are not the result of wastewater inputs. Furthermore, OMR2 is located within the city of Lethbridge but upstream of the WWTP. Again, increases in Cl\(^-\) concentration at this station cannot be attributed to wastewater discharges. Conversely, NaCl distributed across the road network could cause increases downstream of urban centers and also explain the temporal trends observed across each watershed. The observation that Na:Cl ratios in the OMR (OMR1 and OMR2) and RDR (RDR1–RDR3) stations decreased over time further supports a road salt effect. However, the relatively high Na:Cl ratios in the OMR and RDR stations (i.e., 4.0–9.8) indicate that Na\(^+\) chemistry in these systems is not dominated by halite. Furthermore, the absence of a strong Cl\(^-\) seasonality in the RDR and OMR indicate that while seasonal inputs from road salt may be occurring, their impact on seasonality is relatively small. Overall, the weaker road salt signal in the RDR and OMR likely reflects less urban development in these watersheds and this is consistent with results reported elsewhere (Daley et al. 2009; Novotny et al. 2009; Trowbridge et al. 2010).

Non-road salt drivers of salinization in the SSRB

The presence of increasing trends in all seven salts in the SSRB is indicative of one or more non-road salt drivers. Although CaCl\(_2\) and MgCl\(_2\) are used in Alberta for winter road maintenance (note: only CaCl\(_2\) and NaCl are used by

![Fig. 5. Median salt concentrations (meq L\(^{-1}\)), Na : Cl ratios, and SAR in river water as a function of land use and soil properties at LTRN stations. Note: only significant relationships (p < 0.05) are plotted.](image-url)
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Salinization of the SSRB

Temporal trends in flow adjusted ion concentrations shown as rate of change per year. Values in parentheses show the rate of change as a percentage of the long-term (2000–2015) median concentration.

|                | Conductivity (µS cm−1 yr−1) | Na+ (µeq L−1 yr−1) | Mg2+ (µeq L−1 yr−1) | Ca2+ (µeq L−1 yr−1) | SO42− (µeq L−1 yr−1) | Cl− (µeq L−1 yr−1) | HCO3− (µeq L−1 yr−1) |
|----------------|-----------------------------|--------------------|---------------------|---------------------|----------------------|-------------------|---------------------|
| BR1            | 0.7 (0.2)                   | 0.3 (0.3)          | 2.4 (0.2)           | 1.5 (0.2)           | 1.2 (0.2)           | 0.6 (0.0)         | 3.0 (0.3)           |
| BR2            | 2.5 (0.7)                   | 7.8 (1.6)          | 6.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| BR3            | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| BR4            | 3.0 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| OMRI           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| OMR2           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| OMR3           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| SRRI           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| SRRII          | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| SRRIII         | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| RDR1           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| RDR2           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |
| RDR3           | 1.3 (0.9)                   | 7.8 (1.6)          | 5.9 (1.6)           | 5.9 (1.6)           | 7.8 (2.9)           | 9.2 (0.9)         | 4.3 (0.1)           |

*Seasonal Mann-Kendall test shows significant trend at α = 0.05.

The City of Calgary), their contribution relative to NaCl is minimal (e.g., <1% Canada wide, Environment Canada 2012). In addition, there were no distinct increases in the concentrations of these cations upstream vs. downstream of large urban centers. Furthermore, because HCO3−, K+, and SO42− are not part of any winter road maintenance programs in the basin, the observed increases in these salts must have come from other sources. While WWTPs might explain some of the trends in non-halite salts, inputs from WWTP effluent alone cannot explain all of the observed spatial and temporal patterns. In particular, temporal trends in non-halite salts upstream of major urban centers cannot be explained by wastewater effluent. Furthermore, with the exception of Cl−, urbanization was a poor predictor of spatial variation in ion chemistry. In terms of temporal trends in non-Cl− ions, if we assume that the processes operating on spatial variability are also drivers of temporal change, soil salinization and/or crop farming may have been important.

**Soil salinization and agriculture in the SSRB**

Within central and southern Albertan soils, the dominant soluble salt is Na2SO4 (Pawluk and Bayrock 1969). This is consistent with the observation that as the proportion of salt affected (saline and/or solonetzic) soils increased at each station, there was a concurrent increase in the concentrations and relative proportions of riverine Na+ and SO42−. Importantly, the four key parameters related to spatial patterns in salt affected soils (i.e., SO42−, Na+, SAR, and SO42−/Cl−+HCO3−) were also observed to change with time. This suggests an increase in the area, and/or influence, of salt affected soils in the region may be an important driver of temporal change. Cannon and Wentz (2000) used aerial photography to show that total visible salinity has increased in the region during the period 1950–2000. Conversely, Wiebe et al. (2007) reported that due to improved land use practices, the area of land at risk from dryland salinity has declined within Alberta (1981–2001). Although the extent of change in soil salinity in the SSRB is unclear, it is important to note that the observed trends in Na+ and SO42− do not necessarily require an expansion of salt affected soils. Bern et al. (2015) attributed increased riverine salinity in a semi-arid watershed to the disturbance of native soils from oil and gas activities. While, a determination of the relative importance of disturbance vs. an increase in the area of salt affected soils is beyond the scope of this study, it is clear that spoil and temporal patterns in Na+ and SO42− appear to be driven largely by soil processes, and in particular the dissolution and transport of soluble salts.

A number of factors might explain the relationship between the proportion of cropland at each station and riverine salinity. Approximately 20% of water diverted for irrigation from the Bow and Oldman rivers is returned to these rivers further downstream. Importantly, these return flows have been shown to be enriched in salts (Villeneuve and
The mechanisms by which irrigation return flows are salinized are numerous and complex (Vengosh 2014). Historically, the replacement of native vegetation with cropland has accelerated soil salinization within Alberta (Wiebe et al. 2007). It is therefore likely that this has also had an impact on riverine salinity. Furthermore, research indicates that the long-term application of manure in the Oldman River watershed has increased soil salinity in irrigated, and to a larger extent non-irrigated, soils (Hao and Chang 2003). The evaporation of water and subsequent concentration of salts in irrigation channels, soil disturbance and/or the salinization of soils via irrigation waters are additional mechanisms by which crop production might lead to an increase in the concentration of riverine salts (Vengosh 2014). Although identification of the specific mechanism(s) by which crop production has driven spatial patterns in riverine salinity is beyond the scope of this study, results suggest a link between crop production, soil salinization, and riverine salinity. Furthermore, because agricultural activities such as irrigation have expanded in the SSRB over recent decades (Alberta Agriculture and Rural Development 2010), it is reasonable to suggest that this has been an important driver of the observed temporal trends in riverine salinity.

**Salinization and the sustainability of the SSRB**

$\text{Cl}^-$ was well below aquatic health thresholds for acute (640 mg L$^{-1}$) and chronic (120 mg L$^{-1}$) exposures (Canadian Council of Ministers of the Environment [CCME] 2011). For example, the median $\text{Cl}^-$ concentration at ER1 (12 mg L$^{-1}$) was 10 times lower than chronic guidelines for the protection of aquatic life, and based on the current rate of change (i.e., 3% yr$^{-1}$) it would take more than 250 yr to reach this threshold. Having said this, due to the dilution of salts in...
large rivers, it is likely that concentrations in urbanized tributaries of the SSRB are higher than those reported here. This is an important consideration as these smaller tributaries may be experiencing ecological effects not observed in the rivers. In addition, inputs from road salt are sporadic and not well captured by the type of fixed frequency sampling program upon which this study is based. As such, the acute impacts of short-term inputs of NaCl in the SSRB, and the overall impact within urban tributaries of the Bow River watershed remain unclear. Furthermore, if we assume that Cl\textsuperscript{2} concentrations in the most upstream stations are representative of pre-anthropogenic levels, the SSRB is a region where biota likely evolved in a relatively low Cl\textsuperscript{2} environment. As such, increases in Cl\textsuperscript{2} which occur below established toxicity thresholds may be enough to cause shifts in community composition towards more salt tolerant species. Finally, population growth and increased urbanization (Alberta Treasury Board and Finance 2015), increased demand for water (Bruneau and Toth 2007), and increased evapotranspiration due to climate change (Tanzeeba and Gan 2012) are likely to further accelerate salinization in the SSRB. Therefore, despite the fact that Cl\textsuperscript{2} concentrations are currently well below established thresholds, based on the current trajectory, current land use practices will fail to ensure the protection of vital water resources for future generations, albeit over a relatively long time frame. Furthermore, the potential impact of other ions and ion ratios on aquatic biota (Caicedo-Agüelles et al. 2016), together with the impact of episodic inputs to urban tributaries require additional investigation. Importantly, many of the conditions present in the SSRB are not unique. Results from this study suggest that in mixed land use, semi-arid environments receiving regular snowfall, salinization presents a significant threat to the long-term sustainability of freshwater ecosystems.

Fig. 7. Comparison of temporal trends in Cl\textsuperscript{2} loads (kt yr\textsuperscript{-1}) at the mouth of the Bow River (BR4) with trends in road salt application and municipal wastewater discharges (WWTPs) from the City of Calgary (a). Comparison of mean Cl\textsuperscript{2} loads (2008–2014) upstream (BR1) and downstream (BR2) of Calgary with mean Cl\textsuperscript{2} loads from road salt application and municipal wastewater discharges from the City of Calgary (b).

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Conflict of Interest

None declared.

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