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Relationship between soil and groundwater salinity in the Western Canada Sedimentary Basin

Parker J. Banks · John C. Banks

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Abstract Saturated soil paste extracts indicate soluble ions in soil pore water that are available to vegetation. As such, they are thought to accurately describe the relationship between soil and groundwater salinity. To test this assumption, soil and groundwater samples were collected from 575 monitoring wells in saline regions of the Western Canadian Sedimentary Basin (WCSB). Samples were analyzed for electrical conductivity (EC) and Cl^{-}, Na^{+}, Ca^{2+}, Mg^{2+}, K^{+}, SO^{42-}, and HCO^{-3} content. We compared groundwater ionic concentrations to paste extracts derived from matching soils, finding that differences from in situ soil porosity cause saturated pastes to underestimate groundwater salinity. Therefore, we provide pedotransfer functions for accurately calculating groundwater quality from soil data. In addition, we discuss the effects of porosity and soil composition on the saturated paste method, as measured through hydraulic conductivity, saturation percent, and sample lithology. Groundwater salinity may also influence further leaching of salts from soil. As produced water (NaCl brine) spills are common across the sulfate-rich soils of the WCSB, we considered the effects of NaCl on leaching of other ions, finding that influx of Na^{+} into groundwater is associated with increased sulfate leaching from soil. Therefore, considering the secondary effects of produced water on groundwater quality is essential to spill management.

Keywords Saturated paste method · Groundwater quality · Soil salinity · Salt leaching · Lithology

Introduction

Saturated paste extracts are thought to accurately represent the salts involved in groundwater transport (Richards 1954; Longenecker and Lyerly 1964). Consequently, they have been used since the 1950s to assess the soluble ions within the soil (Richards 1954). Typically, the saturated paste method is preferred when evaluating soil salt content under regulatory oversight. It also provides an accurate measure of soil water content at saturation, from which soil wilting point can be derived (Karkanis 1983). As soil wilting point affects nutrient availability for plant uptake (Viets 1972), paste extracts are also preferred for assessing the effects of salinity on plant growth. While the saturated paste method is relatively time-consuming, it is more ecologically valid than the use of faster, fixed 1:1, 1:2, or 1:5 water to soil extracts that fail to account for soil composition (Zhang et al. 2005; Hogg and Henry 1984). Furthermore, fixed weight extracts are more subject to errors resulting from peptization, hydrolosis, cation exchange, and

P. J. Banks (✉) · J. C. Banks
Matrix Solutions Inc., Suite 600, 214-11 Ave SW, Calgary, AB T2R 0K1, Canada
e-mail: banksp@mcmaster.ca

mineral dissolution (Rhoades and Chanduvi 1999). Consequently, the use of saturated pastes has remained the preferred method for understanding salt transport through groundwater from soil data.

Direct analysis of groundwater (soil pore water) is theoretically better than paste extracts for understanding transport of salts and their potential uptake by plants. However, such measurements require installation of a groundwater well, which is often impractical due to land use permissions, disruption of agriculture, groundwater depth relative to the root zone, budget limitations, or the inaccessibility of remote monitoring sites. Therefore, soil data is much more abundant than groundwater for most site assessments. As such, there is a distinct advantage in being able to confidently understand groundwater concentrations through soil data. For example, transport modelling relies on salt mass calculations that are sensitive to errors in soil-water conversions. Such errors could lead to flawed predictions of future site conditions.

Understanding groundwater salinity is particularly important in the oil and gas industry, which globally produces 250 million barrels of wastewater each day (Igunnu and Chen 2012). This produced water often consists of elevated concentrations of sodium chloride brine (Neff et al. 2011) and can dominate production fluids as well output declines. In late-stage production of oil and gas wells, 9 barrels of water must be treated and disposed of for every barrel of oil produced (Veil et al. 2004). Therefore, produced water presents substantial environmental risks through the introduction of sodium chloride into the ecosystems.

In order to mitigate environmental hazards, current regulatory guidelines provide numerical criteria for defining receptor risks. As chloride concentrations in produced water can exceed 20000–40000 mg/L, these criteria are often defined in terms of chloride concentrations (Alberta Environment 2001, 2019). For example, groundwater must meet criteria of 250 mg/l Cl^- for use as potable water, 120 mg/L for aquatic life, or 100 mg/L for soil irrigation. Problems that may arise in mitigating a site impacted by produced water are:

(i) Chloride criteria are very low, so that remediation of produced water to these low levels is often economically infeasible.
(ii) Since chloride is a conservative element, groundwater is the primary transport mechanism for salt. However, remediation via this process often extends for decades.
(iii) Soil salinity criteria have been developed to mitigate risks to vegetation (within the root zone). Since typical impacts extend below the root zone, often well past groundwater levels, existing criteria do not address the effects of salts on deeper soils (for which no criteria are provided).
(iv) Environmental risk assessment focuses on groundwater salinity and salt transport. However, most available data is in the form of measured soil salinity.

Therefore, significant advantages can be gained in understanding the relationship between soil and groundwater salinity. In the present study, we investigated the relationship between soil and groundwater salt concentrations across 144 sites within the Western Canadian Sedimentary Basin, many of which are impacted by produced water. We provide formulas for converting between soil and groundwater concentrations. Furthermore, we investigate how lithology affects the relationship between soil and groundwater salt concentrations, and how the solubility of a single ion is affected by other ions.

Site description

The study area was located within the arid and semi-arid regions of Western Canada Sedimentary Basin (WCSB), extending from the Fort St. John group in north-east B.C. to the Riding Mountain formation in south-western Manitoba (Fig. 1). This region is home to considerable diversity in petroleum reserves and geology, having formed through multiple episodes of tectonic uplift, subsidence, and glacial deposition (Porter et al. 1982). As such, uppermost strata can be divided into eight distinct formations marked by sandy clastic or limited coarse-grained deposition (Wheeler et al. 1996; Mossop and Shetsen 1994).

The Belly River and Wapiti were the first of these to form. During the initial transgression of the Pakowki sea (83 Ma), the Rocky Mountains shed clastics eastwards, forming beds of medium- to fine-grained sandstone and siltstone. Floodplain and fluvial processes continued to deposit the Foremost, followed by the Oldman during renewed mountain formation. About 76 Ma, the transgression of the Bearpaw sea

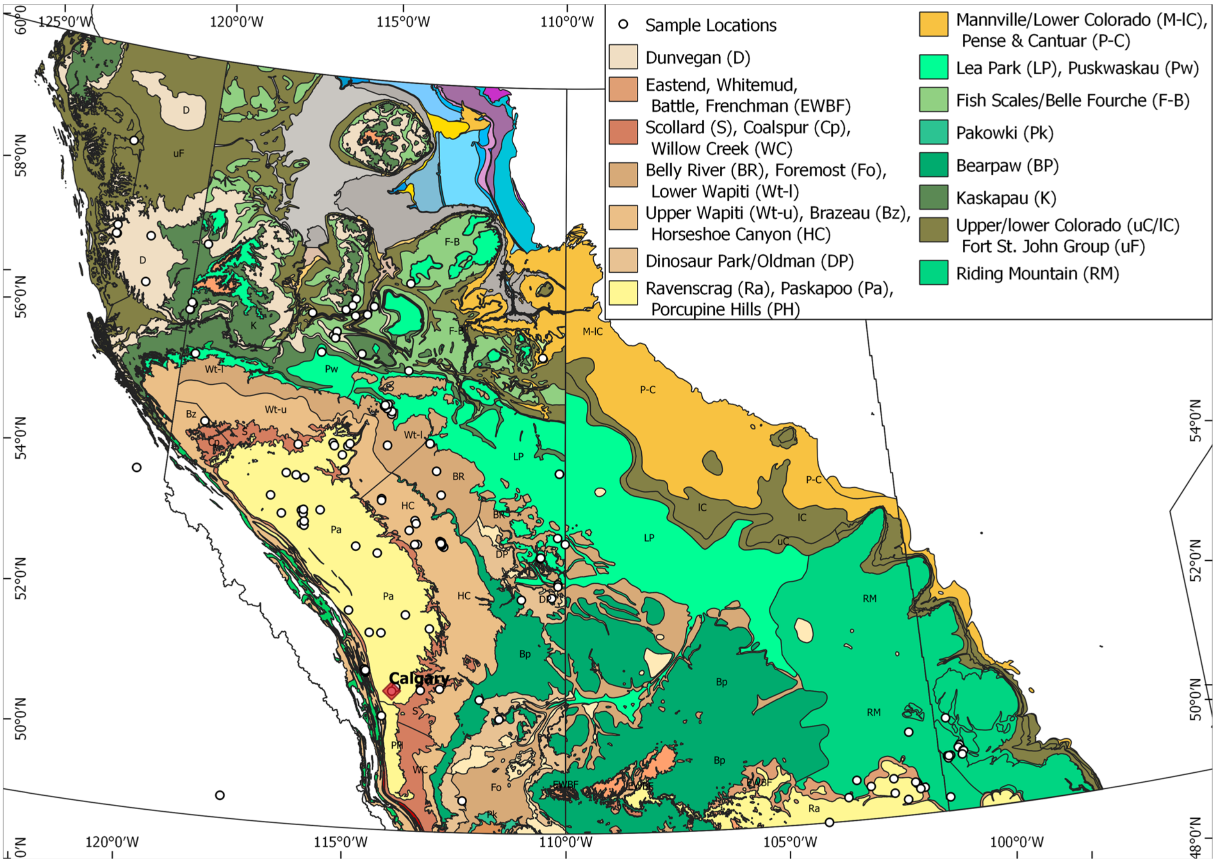

Fig. 1 Assessed remediation sites and bedrock geology. Composited from Cui et al. (2015), Prior et al. (2013), Macdonald and Slimmon (1999), and Manitoba Land Initiative (2014)

deposited laminated shale and siltstone. Further east, the Bearpaw merged with the Lea Park and Pakowki formations, which are marked by fine-grained, dark gray to brown mudstone and siltstone, to become Riding Mountain. As the Bearpaw sea retreated, the Horseshoe Canyon and equivalent Eastend prograded southwards, forming interbedded sandstone, siltstone, and mudstone. Several coal seams were also deposited in fluvial and lacustrine environments. These were overlain by the Whitemud and Battle formations, which consist of dark gray shales atop white kaolinitic siltstone.

About 65 Ma renewed tectonic activity deposited the lower Scollard, Coalspur, and Willow Creek formations. These consist of thick buff to gray sandstone and siltstone beds that also correlate to the Frenchman. Afterwards, sedimentation declined during a period of tectonic quiescence, giving rise to a stable alluvial plain and coal-beds that comprise the upper Scollard and lower Ravenscrag. Renewed tectonic activity and consequent deposition of clastics resulted in the Paskapoo and Porcupine Hills formations. These are marked by thick- and coarse-grained sandstone interbedded with siltstones and shales that are overlain by sand and gravel sheets. The upper Ravenscrag in southern Saskatchewan exhibits similar lithology but is also rich in coal seams.

Field sampling and analysis

In this study, we reviewed data from 575 groundwater wells installed at 144 environmental monitoring sites from 1991–2012, primarily by Matrix Solutions Inc. Many of the sites studied were affected by anthropogenic salts or exhibited naturally elevated soil salinity. Wells generally consisted of a PVC slotted screen and riser pipe installed in 15 cm boreholes typically drilled with an auger rig. 949 soil samples were collected from between the auger flights, at depths ranging from 0–18 m, with a mean depth of 4.02 m. Of these samples, 135 were removed from chemical analysis for being dominantly peat or not being collected within the soil saturated zone or screened interval of the monitoring well. The screened interval in all wells was short (typically 3 m) to ensure that fully purged samples were obtained and to avoid sampling biases caused by long screened wells (Einarson 2006). Multiple samples from a single well were composited by averaging soil parameters weighted by sample thickness within the screened interval. The remaining 538 composite soil samples were paired with 538 groundwater samples collected after purging each well within several months of installation (CCME 1994). The mean groundwater depth across all wells was 2.26 m below surface.

Groundwater samples were collected in .5-L bottles and sent to a laboratory for routine potable water analysis. Soil samples were collected within the auger flights and sealed in glass jars with Teflon-lined lids or soil bags. Soluble salts were extracted from soil samples for analysis using the saturated paste method (McKeague 1978). Deionized water was added to air-dried soil samples to the point of saturation (Carter and Gregorich 2008). After standing a minimum of 4 h, the resulting mud was pressure or vacuum filtered, yielding the saturated paste extract. Ion chromatography and colorimetric methods were used to analyze Cl^- and SO_4^{2-} concentrations, while Na^+, Ca^{2+}, Mg^{2+}, and K^+ concentrations were measured via inductively coupled plasma spectrometry (APHA 2005; McKeague 1978). Groundwater samples were also tested for HCO_3^- via ion chromatography or titration (APHA 2005). Any readings at detection limit (DL) were replaced with DL/2, which is shown to produce the best results following statistical analysis (Farnham et al. 2002).

Hydraulic conductivity (K_{sat}) values were measured at 165 wells via the (Hvorslev 1951) slug test; 155 of which remained after sample quality control. In addition, the lithology of 935 soil samples was classified according to the Canadian and Unified Soil Classification Systems (Soil Classification Working Group 1998; ASTM Committee D-18 2011) and reflected that of the underlying parent material. Sample lithology was recorded as dominantly consisting of clay ($N = 329$), silt ($N = 34$), sand ($N = 190$), loam ($N = 363$), or peat ($N = 19$). Of the 191 non-peat samples collected within the Paskapoo, Scollard, Coalspur, Willow Creek, and Ravenscrag formations (dating from the Paleocene), 83.8% were predominantly sand or loam and had a mean hydraulic conductivity of 2.56×10^{-7} m/s. In contrast, 54.2% of the 699 samples dating from the upper Cretaceous were identified as sand or loam and had a mean hydraulic conductivity of 1.33×10^{-7} m/s. The remaining 26 non-peat samples were collected in formations dating from the Jurrasic, Neoproterozoic, and lower Cretaceous.

Data was received from multiple consulting firms conducting site assessment and monitoring activities over several decades. Therefore, differences in procedural guidelines may have caused variability in data quality. However, most procedural guidelines were derived from or similar to standard industry practices as presented in EPA SW-846 (2015). Soil samples were collected over intervals of 10–150 cm, with the majority spanning 50 cm. When lithology or visual characteristics differed over the sampling interval, a representative sample was collected in proportion to the different horizons within the interval. The majority of analytical testing was completed by the four main laboratories in the Canadian prairies: ALS, AGAT, Element, and Maxxam. As they have been engaged in inter-laboratory testing programs since the 1980s, differences in testing methodology were judged to be inconsequential. During soil groundwater sampling, quality control (QC) was conducted using duplicates, field blanks, and trip blanks. These data were evaluated on a per-site basis by the consulting firm conducting the assessment and removed as needed. Laboratories also maintained internal QC through the use of spiked samples, blanks, duplicates, and internal reference samples. While the use of multiple data sources may introduce variability, data quality was judged to be more than adequate for our study.

Results and discussion

Saturated pastes underestimate groundwater salinity

Measured relationships between groundwater and soil salinity are provided in Table 1. As seen in Fig. 2, saturated pastes reliably capture groundwater mass concentrations (in mg/l) of K^+ and SO_4^{2-} ions, having soil/water ratios close to 1. However, electrical conductivity (in uS/cm) and groundwater concentrations of Cl^-, Na^+, Ca^{2+}, and Mg^{2+} were 1.87–2.94

Table 1 Groundwater/soil concentration ratios and regression coefficients for EC and six ions

Ion	Ratio	B_0	B_{ion}	R^2
EC	2.0649	$1.7112 \pm .2022^{\ddagger}$	$.8668 \pm .0269^{\ddagger}$	.6599
Cl^-	2.1805	$.4997 \pm .1301^{\ddagger}$	$1.0681 \pm .0272^{\ddagger}$	.7440
Na^+	1.8717	$.2043 \pm .1280$	$1.0909 \pm .0257$‡	.7726
Ca^{2+}	2.6607	$2.5953 \pm .1196^{\ddagger}$	$.6547 \pm .0246^{\ddagger}$	.5721
Mg^{2+}	2.9411	$1.6870 \pm .0901^{\ddagger}$	$.8349 \pm .0223^{\ddagger}$	.7248
K^+	.7637	$.0846 \pm .1064$	$.8516 \pm .0410^{\ddagger}$	.4478
SO_4^{2-}	1.0563	$-.0101 \pm .1898$	$1.0119 \pm .0331^{\ddagger}$	.6388

Groundwater concentrations can be estimated from soil values (X, in uS/cm or mg/L) using the formula $y = exp(B_{\text{ion}} \times ln(X) + B_0)$. $^*p < .05$, $^{\dagger}p < .01$, $^{\ddagger}p < .001$

times higher than those reported from associated saturated paste extracts. Therefore, saturated pastes often underestimate groundwater salinity measured in situ. This inconsistency may partly stem from differences in soluble ion composition and cation exchange in soil pastes. For example, sodium binds to clay particles in soil (Hanson et al. 1999), which may cause differences in ionic balances between the two media. In our data, median Na^+/Cl^- ratios were 1.318 and 1.884 in groundwater and soil respectively, while Ca^{2+}/SO_4^{2-} ratios were 2.242 and .959 (mEq basis). Therefore, shifts in soluble ion composition due to cation exchange may partially contribute to the discrepancy between soil and groundwater samples.

Soil porosity and bulk density likely contribute the most to the difference between groundwater and saturated paste salinity. Groundwater samples collected in situ correspond to the water present in saturated soil pores. This pore space is constrained by the compressive force of overlying soil, limiting the soil's water capacity at saturation. However, in collecting a paste extract, the soil is dried, ground, and re-saturated before extracting the soil pore water through pressure filtration. Processing lowers the bulk density of

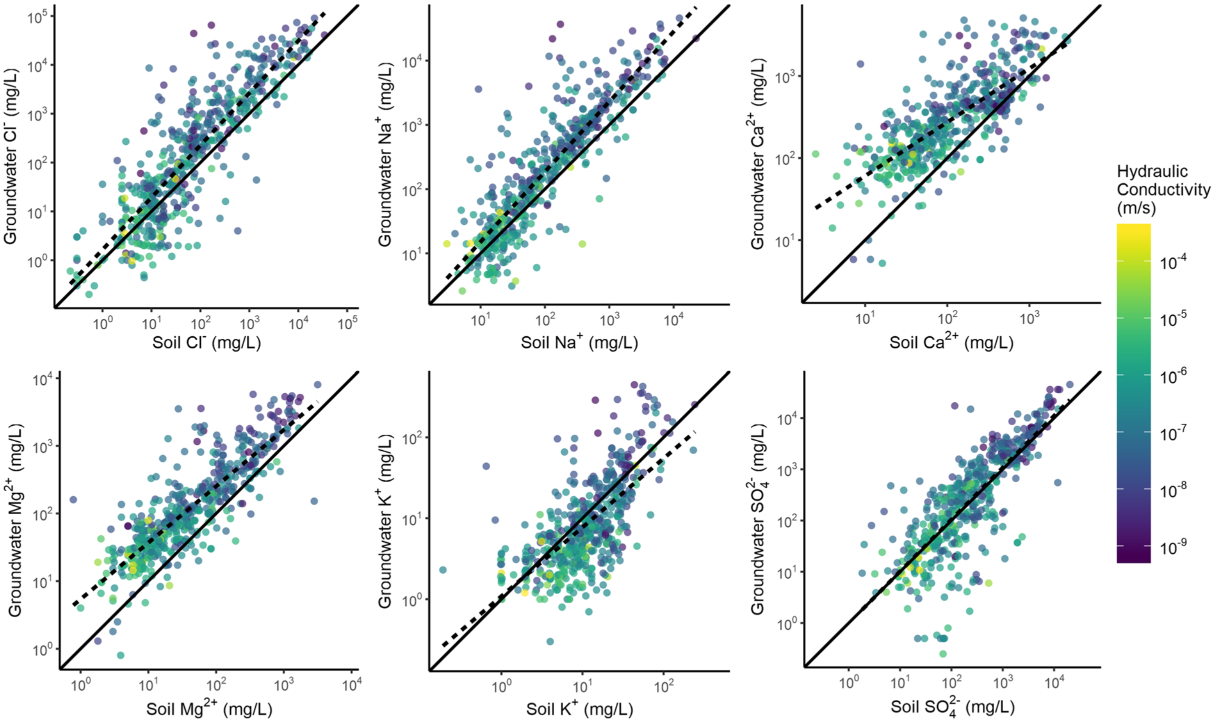

Fig. 2 Relationship between water and soil ion concentrations for soils of varying hydraulic conductivities. For all ions, increasing hydraulic conductivity decreases groundwater salinity, relative to soil. The solid line indicates a 1:1 correspondence between groundwater and soil values, while the dashed line indicates the observed trend

soil, homogenizes particle sizes, and increases porosity. Consequently, more water is required to saturate soil, resulting in dilution of solutes relative to groundwater samples. Fine-grained and clay soils are the most susceptible to these issues, being heavily subject to compaction and swelling relative to sandy, coarse-grained soils (Mohan et al. 1993; Revil et al. 2002). In such cases, measurement of soil water content at the point of collection would allow calibration of paste extracts against in situ saturation conditions. However, in the absence of such data, Table 1 provides formulas for converting between extract and groundwater concentrations. These conversions are accurate at estimating EC, Cl^-, Na^+, Mg^{2+}, and SO_4^{2-}, having R^2 values exceeding .6 (Table 1). However, conversions for Ca^{2+} and K^+ are less accurate and should be used with discretion.

An important final consideration is whether the vertical distribution of salts influence the relationship between soil and groundwater salinity. In our study, the depth and length of the screened interval had no effect on EC, Cl^-, Na^+, Ca^{2+}, Mg^{2+}, or K^+ ($p > .202$ in all cases). This lack of an effect is likely because salt impacts at most of the sites were several decades old, and more uniformly distributed throughout all soil layers sampled. SO_4^{2-} water/soil ratios were significantly affected by sample depth ($F(1, 529) = 5.669, p = .018$). Given the prevalence of naturally occurring sulfate throughout Alberta (Alberta Environment 2001), it is unlikely that produced water impacts caused this effect. However, Bourke et al. (2015) compared soil deuterium, Cl^- and $NO_3^- - N$ profiles against groundwater samples in recent impacts. They found large effects of sample depth and location of the screened interval on relationships between measured soil and groundwater concentrations. Therefore, in more recent impacts, where downward transport of salts dominates, sample depth is an important factor when comparing soil and groundwater ionic concentrations.

Soil composition mediates groundwater salinity

Given the importance of soil porosity, sorting, and grain size to both the water-holding capacity of soil and the transport of salts through groundwater (Nimmo 2004), we evaluated their effects on the correspondence between groundwater and soil salinity. In particular, the hydraulic conductivity (K_{sat}) of a saturated soil indicates the rate at which groundwater moves through soil pore spaces and provides an effective proxy for porosity and particle size (Chapuis 2004; Sperry and Peirce 1995). Therefore, formulas for estimating groundwater salt concentrations from both soil salinity and hydraulic conductivity are given in Table 2.

Since hydraulic conductivity readings were only available for 155 of 538 wells, excluding the remaining 383 samples through listwise deletion might bias estimated water/soil relationships, especially if there was a systematic cause for their omission (Tsikriktsis 2005). For example, the average water and soil Cl^- concentrations were 183 and 74 mg/L for data in which hydraulic conductivity was not measured, but 223 and 101 mg/L for samples where it was. Instead, missing conductivity values were estimated from sample depth, saturation percent (θ_{SP}), lithology, ionic concentrations, and available hydraulic conductivities via multiple imputation with the "Amelia II" R package (Honaker et al. 2011).

Multiple imputation uses the expectation maximization (EM) algorithm to predict the posterior probability distribution of missing data values (i.e., K_{sat}) from observed patterns in available data (Dempster et al. 1977). Because these predictions contain error, the estimated posteriors account for the uncertainty present in each prediction. Multiple imputed datasets are formed by repeatedly sampling from each posterior and assessing the average effect of hydraulic conductivity across all these datasets. This method of repeated sampling while accounting for error results in unbiased estimates of the effects of K_{sat} without adding information that is not already present in the data (Donders et al. 2006).

One hundred imputations were conducted per solute analyzed. To further increase estimate reliability, a bayesian prior was used for imputation, derived from 6339 hydraulic conductivity values sampled by Matrix Solutions between 1991–2017 across the WCSB,[1] primarily using the Hvorslev method, giving a μ of 3.2452×10^{-7} m/s and a log-σ of 3.5566. Groundwater estimates derived from imputed hydraulic conductivities did not significantly differ from measured values ($p > .133$ in all cases), validating the inclusion of imputed data in our analysis.

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[1] Unpublished data

Table 2 Regression coefficients for estimating groundwater ionic concentrations from soil values (X_1, mg/L) and hydraulic conductivity (X_2, m/s) using the formula $y = \exp(B_{\text{ion}} \times ln(X_1) + B_{K_{\text{sat}}} \times ln(X_2) + B_0)$

Ion	B_0	B_{ion}	$B_{K_{\text{sat}}}$	R^2
EC	$1.4121 \pm .2779^{\ddagger}$	$.8432 \pm .0306^{\ddagger}$	$-.0307 \pm .0194$	.6636
Cl^-	$-1.2256 \pm .5436^{*}$	$1.0356 \pm .0288^{\ddagger}$	$-.1202 \pm .0366^{\dagger}$	.7560
Na^+	$-1.1580 \pm .3419^{\ddagger}$	$1.0432 \pm .0279^{\ddagger}$	$-.1027 \pm .0240^{\ddagger}$	.7878
Ca^{2+}	$2.0323 \pm .2589^{\ddagger}$	$.6237 \pm .0272^{\ddagger}$	$-.0459 \pm .0184^{*}$	.5839
Mg^{2+}	$.6277 \pm .2963^{*}$	$.7814 \pm .0256^{\ddagger}$	$-.0814 \pm .0213^{\ddagger}$	.7419
K^+	$-1.4301 \pm .3027^{\ddagger}$	$.7613 \pm .0431^{\ddagger}$	$-.1122 \pm .0205^{\ddagger}$	.5019
SO_4^{2-}	$-1.3579 \pm .4603^{\dagger}$	$.9457 \pm .0397^{\ddagger}$	$-.1107 \pm .0347^{\dagger}$	.6545

$^{*}p < .05$, $^{\dagger}p < .01$, $^{\ddagger}p < .001$

As seen in Table 2, the effect of hydraulic conductivity on EC was non-significant, while those for Ca^{2+} and Mg^{2+} were statistically significant, but weak. As Ca^{2+} and Mg^{2+} bind strongly to clay, and are naturally occurring, it follows that hydraulic conductivity should have less effect on their transport through groundwater. However, hydraulic conductivity (i.e., soil porosity) does influence the association between soil and groundwater Cl^-, Na^+, K^+, and SO_4^{2-} concentrations. As seen in Figs. 2 and 3, sandier soils have smaller groundwater/soil ratios. In sand (e.g., $K = 10^{-4}$ m/s), groundwater flows freely due to higher porosity. These conditions are maintained in laboratory paste extracts, so that there is a closer correspondence between saturated paste extracts and groundwater salinity. However, in clay, changes in soil porosity in laboratory conditions cause underestimation of groundwater salinity. Our conversions correct these biases caused by soils exhibiting low hydraulic conductivity and porosity.

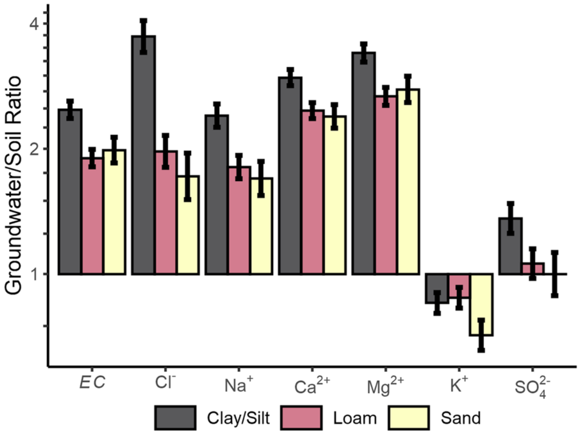


Fig. 3 Effect of soil type on ratios of groundwater to soil parameters, as calculated from Table 3. Ratios were calculated at the geometric average for each soil parameter, where EC = 1970.6 uS/cm, Cl^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, and SO_4^{2-} = 81.8, 133.9, 121.1, 47.2, 11.7, and 260.0 mg/l

While hydraulic conductivity is the most direct measurement of the potential movement of water, it is difficult to estimate, requiring the installation of a monitoring well. In contrast, soil saturation percent (θ_{SP}) is easily determined from saturated pastes. Saturation also correlates with hydraulic conductivity (Gamie and De Smedt 2018), being directly related to soil texture (Mbagwu and Okafor 1995; Stiven and Khan 1966). However, its relationship with saturated hydraulic conductivity relies on numerous other factors such as soil structure, bulk density, organic matter, and water content (Tietje and Hennings 1996; Jabro 1992; Hendry and Paterson 1982). Whereas K_{sat} provides an indicator of in situ soil porosity, θ_{SP} relates to porosity in disturbed soil. For example, in our data, θ_{SP} and hydraulic conductivity exhibited a weak correlation ($R(145) = -.319, p < .001$). While dependence on these factors varies between undisturbed and ground, re-saturated soils, we tested whether saturation percentage is sufficient to capture the differences between fine and coarse-grained soils. Therefore, multiple linear regression was conducted, testing for the effect of saturation percentage on groundwater ionic concentrations, after controlling for soil concentrations in non-peat soils. It had significant effects on EC ($F(1, 521) = 7.312, p = .007$) and K^+ ($F(1, 517) = 7.518, p = .006$) but not on any other ions ($p > .170$ in all cases). Given these small (relative to hydraulic conductivity) and inconsistent effects, saturation percentage was not an effective measure of water/soil relationships within the current study.

Sample lithology provides an additional easy measure of soil composition. Therefore, we tested for the effects of lithology on groundwater/soil relationships. Given the impracticality of compositing soil classifications, for this analysis, the dataset comprising 814 non-composited soil samples was used. While pairing multiple soil samples with a single groundwater sample is expedient, it may introduce additional variability into our analysis, as the thickness of soil samples in relation to the screened interval is no longer accounted for. Regardless, there was a significant effect of soil type on the relationship between groundwater and soil EC ($F(2, 796) = 8.333, p < .001$), Cl^- ($F(2, 782) = 18.086, p < .001$), Na^+ ($F(2, 785) = 6.762, p = .001$), Ca^{2+} ($F(2, 785) = 5.577, p = .004$), Mg^{2+} ($F(2, 785) = 6.240, p = .002$), and SO_4^{2-} ($F(2, 796) = 6.290, p = .002$). As seen in Fig. 3, saturated paste extracts share the greatest correspondence with groundwater samples in sand-dominated soils for all ions except K^+. Cl^- ions are most influenced by soil type due to their mobility through soil (Letey and Klute 1960). Therefore, formulas for predicting groundwater parameters from soil values and lithology are given in Table 3.

Ion-ion interactions

While the pedotransfer functions in Tables 1–3 provide useful tools for estimating groundwater quality, they can be further improved through an understanding of how interactions between soil and groundwater ions drive groundwater salinization. For example, the introduction of NaCl through produced water has an immediate impact on the ecosystems, but can have secondary effects by enhancing leaching of naturally occurring ions into groundwater, such as sulfate. Given the importance of SO_4^{2-} as a measure of groundwater quality (Alberta Environment 2019) leaching of otherwise insoluble sulfate salts could present additional environmental risks. In laboratory settings, sodium chloride is found to increase the solubility of barium, sodium, and calcium sulfate (i.e., gypsum) (Templeton 1960; Mockobey 1932; Meijer and Van Rosmalen 1984; Salman et al. 2015). The presence of $NaNO_3$, $Mg(NO_3)_2$, and $MgCl_2$ also increase gypsum solubility, while $CaCl_2$ and $Ca(NO_3)_2$ cause it to decrease (Shternina 1960; Seidell and Smith 1904). In addition, Curtin and Syers (1990) demonstrated that NaCl increases SO_4^{2-} leachability from soil. These additional salinization pathways are particularly relevant to the WCSB, which is rich in naturally occurring sulfates found within salt beds, sour gas, and gypsum deposits (Govett 1958; 1961; Bailey et al. 1974). Therefore, elevated NaCl within our observational data may also be associated with increased leaching of other ions into groundwater (i.e., increased groundwater/soil ratios), such as sulfate.

However, one shortage to the regression analyses of Tables 1–3 is that they are correlative, rather than demonstrating causal links between soil and groundwater salinity. The problem of correlation is especially evident in deep formation waters of the WCSB, where Cl^-, Na^+, Ca^{2+}, and Mg^{2+} are strongly related (Hitchon and Holter 1971; Walter et al. 2017). In our data, soil calcium and magnesium are heavily correlated with a R^2 of .809. We attempted to disentangle the causal relationships between soil and groundwater properties from these confounds using structural equation modelling. Structural equation models exhibit several advantages over traditional regression. In particular, they allow causal relationships to be inferred from non-gaussian structures in observational data (Shimizu 2014). Furthermore, all interactions between

Table 3 Regression coefficients for estimating groundwater ionic concentrations from soil values (X_1, mg/L), and soil type (X_2) using the formula $y = \exp(B_{ion} \times ln(X_1) + B_{soil} + B_0)$ where $B_{soil} = 0$, B_{loam}, and B_{sand} for clay, loam, and sand respectively

Ion	B_0 (Clay)	B_{ion}	B_{loam}	B_{sand}	R^2
EC	1.9760‡	.8594‡	−.2673‡	−.2231*	.6740
Cl^-	1.2379‡	1.0174‡	−.6358‡	−.7741‡	.7444
Na^+	.6773‡	1.0407‡	−.2848†	−.3478†	.7486
Ca^{2+}	2.9056‡	.6210‡	−.1830†	−.2151†	.5549
Mg^{2+}	1.9825‡	.8030‡	−.2401‡	−.2011*	.7164
K^+	.1067	.8916‡	.0289	−.1787	.4764
SO_4^{2-}	.7110‡	.9275‡	-.2502*	−.3081*	.5959

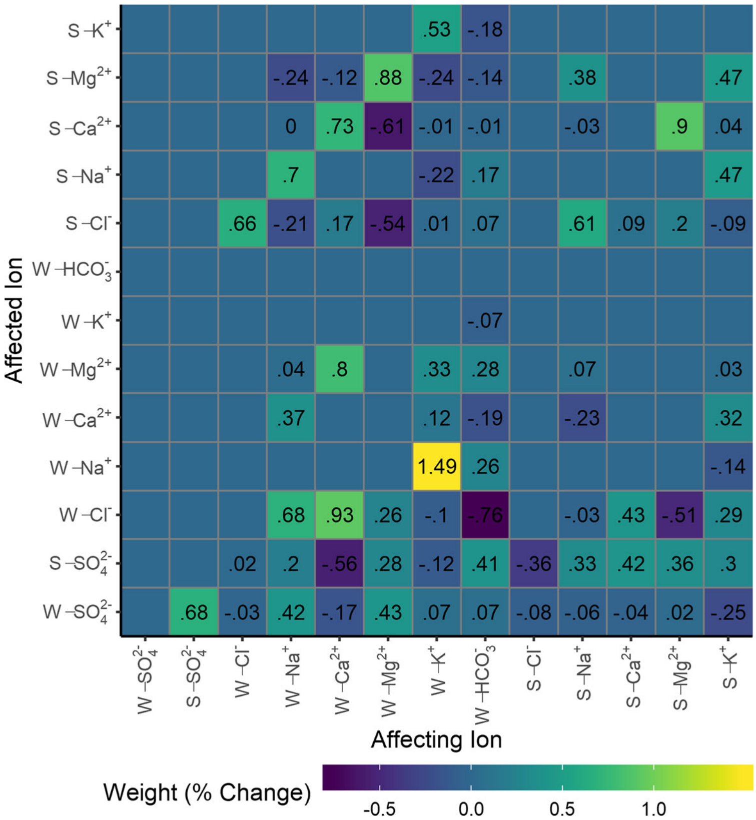
* $p < .05$, $^†p < .01$, $^‡p < .001$

soil and groundwater ions can be explored within a single model, rather than treating groundwater ions as independent from one another. Our model was generated using the Direct-LiNGAM algorithm of Shimizu et al. (2011). This algorithm produces a directed acyclic graph of causal connections from observational data. Each connection represents the percent change in an affected ion, given a 1% change in an affecting ion. To ensure that latent confounds were minimized, groundwater HCO_3^- observations were included with all other salinity data. However, electrical conductivities were omitted due to high collinearity with other variables (VIF $>$ 11 in all cases). We also calculated 95% confidence intervals for each connection from 2000 bootstrap replicates

Results of the Direct-LiNGAM algorithm are given in Fig. 4. To aid in the detection of patterns among variables, connections were sorted according to the angular order of their eigenvectors. Both groundwater Na^+ and Mg^{2+} ions increase groundwater SO_4^{2-} by .42 (0, .56) and .43 (0, .74) percent after controlling for the effect of soil sulfate (.68%, 95% CI [.55,.86]). Consequently, sodium-bearing produced water increases leaching of sulfates and associated groundwater/soil ratios. Groundwater calcium has the opposite effect, reducing sulfates in both soil by −.56% (−.86, 0) and water by −.17% (−.51, .17), due to the precipitation of relatively insoluble gypsum (calcium sulfate). These results are consistent with previously observed effects of Na^+, Ca^{2+}, and Mg^{2+} on sulfate leaching from soil (Curtin and Syers 1990).

Movement of chloride into groundwater is also heavily influenced by groundwater Na^+ (.68% [0, 1.12]) and Ca^{2+} (.93% [0, 1.43]). That the effect of calcium is significantly greater than that of sodium coincides with the difference in charge between the two ions. However, the effects of Mg^{2+} on groundwater Cl^- are considerably lower, likely due to the greater abundance of Na^+ and Ca^{2+} in soils within the WCSB. Conversely, bicarbonates lower Cl concentrations by −.76% (−1.11, −.28). Sodium chloride inhibits precipitation of $CaCO_3$ and $Ca(HCO_3)_2$ (Kitano 1962; He et al. 1999), resulting in antagonism due to shared negative charges (Mitchell et al. 2005). Chloride is thought to exhibit little affinity for soil components, and therefore should be heavily affected

Fig. 4 Directed acyclic graph representing relationships between all soil and groundwater parameters. Weights are given as the expected % change in each affected ion, given a 1% change in each affecting ion



	W-SO_4^{2-}	S-SO_4^{2-}	W-Cl^-	W-Na^+	W-Ca^{2+}	W-Mg^{2+}	W-K^+	W-HCO_3^-	S-Cl^-	S-Na^+	S-Ca^{2+}	S-Mg^{2+}	S-K^+
S-K^+							.53	-.18					
S-Mg^{2+}				-.24	-.12	.88	-.24	-.14		.38			.47
S-Ca^{2+}				0	.73	-.61	-.01	-.01		-.03		.9	.04
S-Na^+				.7			-.22	.17					.47
S-Cl^-			.66	-.21	.17	-.54	.01	.07		.61	.09	.2	-.09
W-HCO_3^-													
W-K^+								-.07					
W-Mg^{2+}				.04	.8		.33	.28		.07			.03
W-Ca^{2+}				.37			.12	-.19		-.23			.32
W-Na^+							1.49	.26					-.14
W-Cl^-				.68	.93	.26	-.1	-.76		-.03	.43	-.51	.29
S-SO_4^{2-}			.02	.2	-.56	.28	-.12	.41	-.36	.33	.42	.36	.3
W-SO_4^{2-}		.68	-.03	.42	-.17	.43	.07	.07	-.08	-.06	-.04	.02	-.25

Table 4 Groundwater/soil concentration regression coefficients for EC and six ions

Ion	B_0	B_{Cl^-}	B_{Na^+}	$B_{Ca^{2+}}$	$B_{Mg^{2+}}$	B_{K^+}	$B_{SO_4^{2-}}$	R^2
EC	5.0596‡	.1977‡	.2808‡	.1032	.1264*	ns	ns	.6758
Cl^-	$-.4462$	.9737‡	ns	.2851‡	ns	ns	ns	.7540
Na^+	.6853†	.1960‡	.8877‡	$-.2217$*	.1717*	$-.1281$	.0673	.7892
Ca^{2+}	2.8351‡	.1246‡	ns	.5675‡	ns	ns	$-.0635^{\dagger}$	.6284
Mg^{2+}	2.4889‡	.1186‡	ns	$-.4633^{\ddagger}$	1.0733‡	ns	ns	.7598
K^+	$-.0377$	.0827‡	.2485‡	$-.3182^{\ddagger}$	.3076‡	.4254‡	ns	.6480
SO_4^{2-}	.7853†	ns	.3463‡	$-.4619^{\ddagger}$	.5754‡	-.3008‡	.7100‡	.7117

Groundwater concentrations can be estimated from soil values (in uS/m or mg/L) using the formula $y = \exp(B_{EC} \times ln(X_{EC}) + B_{Cl^-} \times ln(X_{Cl^-}) \ldots + B_0)$. *$p < .05$, $^{\dagger}p < .01$, $^{\ddagger}p < .001$

by leaching processes (White and Broadley 2001). However, Svensson et al. (2017) recently demonstrated that Cl pore water concentrations, nitrate, organic carbon, and soil moisture influence chlorination of soils. We provide further evidence that Cl reacts with soil components.

In the case of sodium leaching, it is most strongly affected by potassium. Na^+ and K^+ ions behave similarly, having similar effects on clay dispersion, ionic radii (1.02 and 1.38 Å respectively), and identical charges. Na^+ and K^+ should therefore repel and displace each other from clay binding sites, causing each other to leach into groundwater. Brønsted (1922) asserts that ions of the same sign have a uniform influence on the solubility of one another. Conversely, potassium is known to be less susceptible to leaching than are calcium and magnesium (Wong et al. 1992; Lehmann and Schroth 2003). Our results support this theory, given that groundwater Na^+ exhibits a 1.49% (0, 1.56) increase following a 1% change in K^+, while groundwater K^+ is unaffected by other ions. Mg^{2+} also exhibits a .80% (0, .99) increase in leachability in response to Ca^{2+}. However, Fig. 4 highlights a shortcoming of the Direct-LiNGAM algorithm. While Ca^{2+} and Mg^{2+} uniformly influence one another, Direct-LiNGAM produces acyclic graphs, which only consider unidirectional effects between ions. Therefore, the effects of Mg^{2+} on Ca^{2+} or K^+ on Na^+ should be considered synonymous with those of Ca^{2+} on Mg^{2+} or Na^+ on K^+.

Given the interactions between soil and groundwater ions, multiple soil parameters can be used to predict groundwater quality. Though less comprehensive than our structural equation model, the pedotransfer functions in Table 4 provide improved accuracy over those of Table 1, while remaining simple and usable by industry. Predictors for estimating groundwater concentrations of each ion were selected via forwards and backwards stepwise multiple regression in the R MASS package (Venables and Ripley 2002). An α of p=.15 was used, so that parameters where $p > .15$ were omitted as non-informative. Again, soil EC data were removed due to high collinearity with other variables (VIF > 14.5 in all cases).

These expanded formulas considerably improve estimates of groundwater Ca^{2+}, K^+, and SO_4^{2-} concentrations from soil data. However, the goodness of fit for EC, Cl^-, Na^+, and Mg^{2+} estimates only slightly improved. Consequently, when accurate estimates of Ca^{2+}, K^+, and SO_4^{2-} are needed, the expanded formulas in Table 4 are preferable, but are unnecessary for the other ions.

Conclusions

In the present study, we provide one of the first large-scale comparisons of soil and groundwater data, showing that historical soil data can lead to advancements in modelling contaminant fate and transport. We validated the saturated paste method against data collected from groundwater wells in the Western Canadian Sedimentary Basin. While the saturated paste method underestimates groundwater salinity, when reported in milligrams per liter, its results are highly consistent, allowing us to provide formulas for estimating groundwater salinity from paste extracts. These formulas account for change in soil porosity, grain size, and composition following processing of saturated pastes relative to in situ conditions. Therefore, the common convention of calibrating paste extracts against laboratory measured saturation percent, for the purpose of reporting salt concentrations in milligram/kilogram units, should be avoided. This practice underestimates salt concentrations of soil pore water due to the over-saturation of soil samples in the lab. Instead, saturated pastes should be calibrated against soil-moisture measured on site. However, the conversions given in Tables 2–3 also correct for biases in paste extracts when hydraulic conductivity or soil lithology are known.

In addition to allowing estimation of groundwater salinity from saturated soil pastes, these formulas allow preliminary soil data to be used to determine the ideal screened interval depth when installing monitoring wells. During initial site assessment soil salinity can now be used as a proxy for groundwater concentrations. These estimates may prove especially cost-effective when assessing the salinity of deep groundwater. However, future studies are needed to elucidate the effects of depth on soil/groundwater relationships. In our current study, the effects of depth were minimal, due to the even distribution of solutes through naturally saline regions and old impacts. However, Bourke et al. (2015) demonstrated that sample depth is a contributing factor in recent impacts, where downward (rather than lateral) flow through multiple soil layers dominates solute transport.

Finally, we considered the interdependencies between different ions in soil and groundwater, allowing for the development of more accurate pedotransfer functions. A causal structural equation model revealed the influences of groundwater salinity on the leaching of ions from soil into groundwater. We found that the presence of sodium tends to increase groundwater sulfate concentrations relative to soil. Consequently, produced water exhibiting high concentrations of sodium chloride can have the secondary effect of leaching sulfates into the water table. As the Western Canadian Sedimentary Basin is rich in naturally occurring sulfates, remediation criteria specific to naturally sulfate-rich soils should be developed, for which our findings should prove a useful guideline.

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