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The distribution and migration of sodium from a reclaimed upland to a constructed fen peatland in a post-mined oil sands landscape

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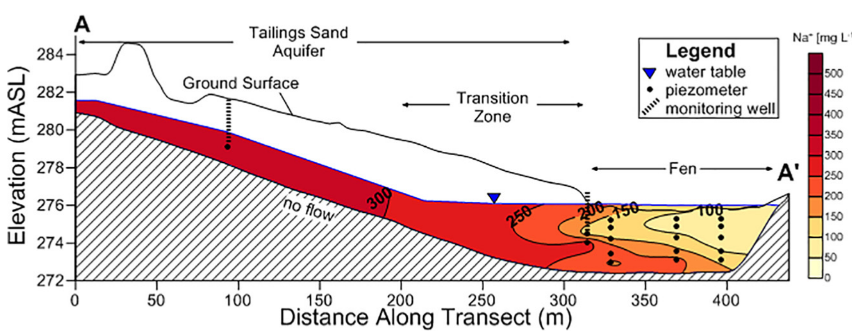
HIGHLIGHTS

- Upland recharge was an important control of sodium migration.
- The driest year had the highest solute flux from upland to fen.
- Arrival times of sodium at the fen surface were estimated to be 4–11 years.
- Sodium concentrations rose from 87 to 200 mg L^{-1} in the fen rooting zone.

GRAPHICAL ABSTRACT



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ABSTRACT

Post-mine landscape reclamation of the Athabasca Oil Sands Region requires the use of tailings sand, an abundant mine-waste material that often contains large amounts of sodium (Na^+). Due to the mobility of Na^+ in groundwater and its effects on vegetation, water quality is a concern when incorporating mine waste materials, especially when attempting to construct groundwater-fed peatlands. This research is the first published account of Na^+ redistribution in groundwater from a constructed tailings sand upland to an adjacent constructed fen peat deposit (Nikanotee Fen). A permeable petroleum coke layer underlying the fen, extending partway into the upland, was important in directing flow and Na^+ beneath the peat, as designed. Initially, Na^+ concentration was highest in the tailings sand (average of 232 mg L^{-1}) and lowest in fen peat (96 mg L^{-1}). Precipitation-driven recharge to the upland controlled the mass flux of Na from upland to fen, which ranged from 2 to 13 tons Na^+ per year. The mass flux was highest in the driest summer, in part from dry-period flowpaths that direct groundwater with higher concentrations of Na^+ into the coke layer, and in part because of the high evapotranspiration loss from the fen in dry periods, which induces upward water flow. With the estimated flux rates of 336 mm yr^{-1}, the Na^+ arrival time to the fen surface was estimated to be between 4 and 11 years. Over the four-year study, average Na^+ concentrations within the fen rooting zone increased from 87 to 200 mg L^{-1}, and in the tailings sand decreased to 196 mg L^{-1}. The planting of more salt-tolerant vegetation in the fen is recommended, given the potential for Na^+ accumulation. This study shows reclamation designs can use layered flow system to control the rate, pattern, and timing of solute interactions with surface soil systems.

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1. Introduction

By 2013, open pit mining to extract oil sands had disturbed over 895 km^2 of the boreal forest within the Athabasca Oil Sands Region (AOSR; Government of Alberta, 2017). Given that wetlands comprise ~50% of this region, most of which are reported to be fen peatlands (Vitt et al., 1996), fen peatland construction has been incorporated into the mine closure planning, at least on a pilot scale, to reclaim the disturbed land to an equivalent land class (Daly et al., 2012). The construction of a fen peatland (in this case "Nikanotee Fen") is a pioneering attempt at post-mined AOSR landscape reclamation (Daly et al., 2012; Ketcheson et al., 2016). Nikanotee Fen relies on groundwater derived from an adjacent upland aquifer constructed from tailings sand to sustain water levels essential for fen peatland development (Price et al., 2010). Tailings sands are an abundant waste product that require reclamation, containing residual concentrations of sodium (Na^+; Rezanezhad et al., 2012a, b; Holden et al., 2011), amongst other constituents (MacKinnon et al., 2001; Scott et al., 2005). Given that Na^+ rich leachate from tailings sands may be toxic to fen vegetation (Pouliot et al., 2012; Rezanezhad et al., 2012a), the influence of process-affected materials on water quantity and quality must both be considered (Daly et al., 2012; Ketcheson et al., 2016; Simhayov et al., 2017). Na^+ flushed from the tailings sands upland aquifer is expected to create pulses of elevated (above background) Na^+ concentrations that will enter the fen peat (Daly et al., 2012; Simhayov et al., 2017). Once in the fen, Na^+ transport is anticipated to slow due to adsorption to pore surfaces within the peat (Rezanezhad et al., 2012b; Simhayov et al., 2017). Given high upland to fen ratios likely for extensive reclamation in the AOSR, and thus tailings sand to peat volumes, there will be more Na^+ within the upland areas than can be adsorbed by the peat (Simhayov et al., 2017), eventually leading to elevated concentrations within the fen rooting zone. In response, vegetation species will likely shift to salt-tolerant species (Trites and Bayley, 2008, 2009; Rezanezhad et al., 2012a). Therefore, understanding Na^+ transport dynamics will offer valuable insight into the ecological development of constructed wetlands.

In the Nikanotee Fen Watershed, the adjacent reclaimed hillslopes and constructed upland currently provide sufficient water to maintain adequate saturation within the fen under moderate seasonal water deficits (Ketcheson and Price, 2016a; Ketcheson et al., 2017). The relatively permeable (2 orders-of-magnitude higher than peat) petroleum coke underdrain plays an important role in distributing flows beneath the fen (Ketcheson et al., 2017). Given the hydraulic gradients from upland to fen (Ketcheson et al., 2017), and the presence of Na^+ solutes in the upland (Simhayov et al., 2017), Na^+-enriched groundwater is expected to migrate from the tailings sand upland, through the petroleum coke underdrain, to beneath the fen. Biagi (2015) documented the migration of Na^+ in the nearby constructed Sandhill Fen Watershed, in which Na^+ moved from underlying composite tailings into overlying peat materials. Sandhill Fen was constructed with a system of tile drains beneath it, to mitigate near surface salt accumulation, if needed (Nicholls et al., 2016). Both constructed systems highlight the importance of underdrain engineering to manage salinity within the groundwater, to delay the arrival time of Na^+ to the fen surfaces.

The hydrological connections between Nikanotee Fen and the constructed upland has been well-documented by Ketcheson et al. (2017); however, the migration and fate of solutes within the upland-fen system are thus far undocumented, and are poorly understood. Rezanezhad et al. (2012b) reported a high attenuation capacity (sorption) of Na^+ for fen peat; however, other solutes generated from tailings materials (e.g., Ca^{2+}, Mg^{2+}) will compete for sorption sites, reducing the Na^+ adsorption capacity of the fen. Laboratory experiments confirmed that the delayed transport (retardation) of Na^+ in fen peat from the AOSR was primarily due to sorption (Rezanezhad et al., 2012a; Simhayov et al., 2018) rather than diffusion of solute into dead-end pores (Hoag and Price, 1997). Simhayov et al. (2018) showed in laboratory columns, that Nikanotee Fen peat was sufficiently degraded that it behaves as a single porosity medium, but that kinetically controlled adsorption retards the migration of Na^+.

The success of future constructed wetlands will depend on designs accounting for the management of Na^+ transport from tailings sand materials as early arrival times of salts to the wetland surface can jeopardize vegetation establishment. Therefore, the primary research objectives of this study are to: 1) characterize the distribution of Na^+ as it migrates from a tailings sand upland to the fen; 2) evaluate how the system design and hydraulic properties of construction materials control the transport and fate of Na^+ in the system; and 3) suggest strategies to manage Na^+ migration in future designs.

2. Study site

2.1. Constructed upland – fen system

The study site, a constructed upland – fen peatland system, forms the core of the Nikanotee Fen Watershed (56°55.944′N, 111°25.035′ W) ~30 km north of Fort McMurray, Alberta. The upland – fen system was designed such that lateral groundwater flow from upland to fen sustains a water table at or near the surface of the fen under periods of drought (Price et al., 2010; Daly et al., 2012). A combination of salvaged (peat and forest soil) and mine waste materials (tailings sand and petroleum coke) layers were incorporated into construction designs to convey water and manage the transport of solutes from the constructed upland to the fen (Daly et al., 2012). The upland (7.7 ha) is a 2–3 m thick tailings sand aquifer on a 3% basal slope, underlain by an impermeable engineered geosynthetic clay liner and overlain by a relatively thin (0.3–0.5 m) LFH-mineral mix reclamation forest soil material (herein referred to as 'LFH'; Fig. 1). The lower lying fen (2.9 ha), situated at the toe of the upland, is ~2 m of moderately decomposed fen peat (average bulk density 0.2 g cm^{-3}; anisotropy of ~1.0; Nwaishi et al., 2015) overlying a ~0.5–0.7 m thick petroleum coke layer (herein referred to as the 'petroleum coke underdrain') that extends ~100 m into the upland, beneath the tailings sand (this portion of the upland is referred to as the 'transition zone'; Fig. 1). The soil hydraulic properties of construction materials are provided in Appendix A, and a detailed description of the system design and hydrological performance is presented by Ketcheson et al. (2017). Briefly, the strong contrast in saturated hydraulic conductivities between the tailings sand and petroleum coke directs lateral flow into the petroleum coke underdrain, which transmits most of the flow towards and beneath the fen, after which it migrates upwards through the peat profile (Fig. 1; Ketcheson et al., 2017). An outlet (spill box) was installed at the north-east corner of the fen to allow for outflow through a flume to an outflow pond.

The 7.7 ha constructed upland and 2.9 ha fen were encompassed by reclaimed slopes, forming a watershed area of 32.1 ha. Hillslopes to the southeast and west (8.2 and 2.4 ha, respectively; Fig. 1) were reclaimed in 2011, and occasionally contribute surface runoff that recharges the upland aquifer, ultimately reaching the fen peatland. The 8.1 ha east hillslope slope was constructed earlier, five years before the fen (2007); it provides runoff during the spring freshet (Ketcheson and Price, 2016b), but little surface runoff to the upland-fen system during the snow free season (Ketcheson and Price, 2016a). Four recharge basins (Fig. 1) were excavated behind "hummocks" (earthen mounds constructed originally to increase ecological and topographical diversity), with the intent of increasing the infiltration and detention volume of overland flow (Kessel, 2016). These were created in the autumn of 2013 by removing the LFH behind them to expose the more permeable tailings sands; concurrently, furrows were tilled across the upland LFH to improve surface water detention (Ketcheson, 2015), and thus recharge. A small depression in the south of the upland lined with 0.5 m thick peat-mineral mix material (referred to as the 'peat-lined basin'; Fig. 1) acts as a recharge window (mostly from snow trapped within it); it has a negligible contributing area (Ketcheson, 2015).

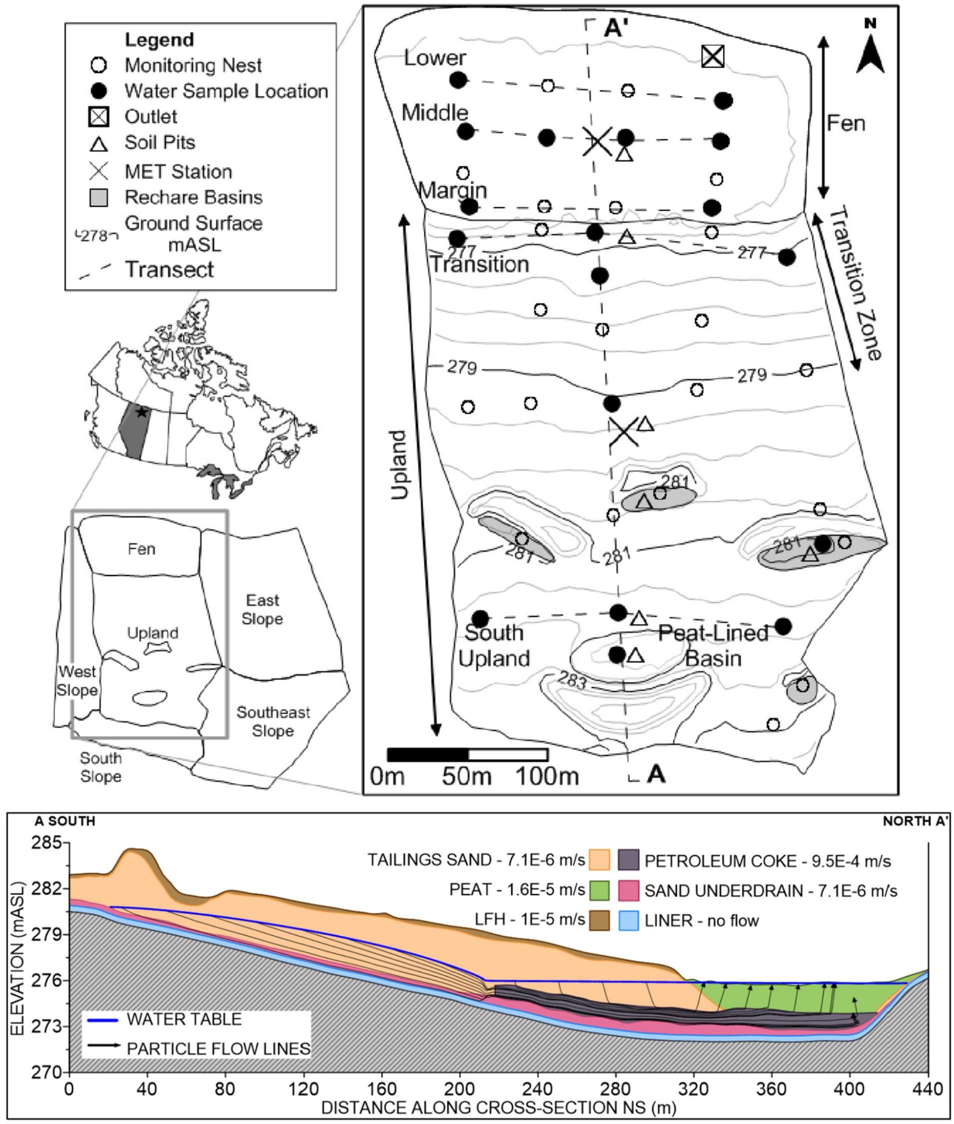

Fig. 1. Map of Nikanotee Fen with site instrumentation (top; plan view) and layer properties (bottom; A-A′ cross-section). Cross-section includes particle flow lines for particles dropped every 25 m within the upland tailings sand aquifer. Flow lines generated using MODFLOW (from Ketcheson et al., 2017) display convergence of flow into the petroleum coke underdrain and then 'bottom up' flow within the fen peatland.

The fen was vegetated using a factorial design consisting of control (bare peat), moss seedlings, seedling-moss and seeds with combinations of mulch-covered and non-mulch covered plots. More detail on the planting design can be found in Murray et al. (2017). Evapotranspiration rates (*ET*) within the fen were highest over open water (4.4 mm day^{-1}) and lowest over moss-mulch plots (2.4 mm day^{-1}; Scarlett et al., 2017), averaging 3.3 mm day^{-1} in 2014. *ET* from the fen was the dominant water flux within the entire upland – fen system, which typically created a seasonal water deficit for the fen (Ketcheson et al., 2017; Scarlett et al., 2017). High *ET*, in combination with the hydraulic design generates an upward flow from the petroleum coke underdrain into the peat profile. Early season ground frost constrained upwards groundwater fluxes; once thawed, vertical hydraulic gradients generated upward flow for ~85% of the 2014 snow-free (Ketcheson et al., 2017).

3. Methods

This study covered the first four-year period following construction of Nikanotee Fen. All data were collected in the snow-free periods between late May to early October for 2013, 2014 and 2015. The 2016 study period was confined June to early October due to a wildfire in the area.

3.1. Instrumentation and monitoring

A grid network of monitoring locations (herein referred to as 'nests') of wells and piezometers was employed to target water levels and pressure heads within distinct constructed material layers (Fig. 1). Instrumentation began in late April 2013 and was generally completed by July 2013; hydrological and geochemical monitoring detail (i.e. sample sizes) increased as additional nests were installed. All wells instrumented within the upland and transition zone were 2.54 cm inner diameter (I.D.) polyvinyl chloride (PVC) pipes installed in a pilot hole made using a portable earth auger (Stihl BT 121; 5.08 cm auger diameter). All piezometers within the upland (max depth of 275 cm below ground surface (bgs)) and transition zone (225 cm and 275 cm bgs) were stainless steel drive points (Solinst Canada Ltd., Model 615; 10 or 20 cm slotted intake) installed using a portable rock percussion hammer (Pionjar 120) and lined with 1.2 cm I.D. low density polyethylene tubing to prevent contamination of water samples by the galvanized steel casings. A total of 14 nests were installed throughout the fen, each consisting of a well (150 cm total depth) and five piezometers installed to targeted depths of 50, 90, and 150 cm bgs within peat and 225 and 275 cm bgs within the petroleum coke underdrain and tailings sand that underlies it, respectively. All wells and piezometers within the fen were constructed from PVC pipe (2.54 cm I.D.) and fen piezometers had a 20 cm slotted intake; both were wrapped with filter sock and installed into pilot holes drilled using a hand auger. All absolute positions and elevations (mASL; ±0.5 cm vertical accuracy) of wells and piezometers were determined annually by a Topcon HiPER GL RTK GPS system (2013) and a Leica Geosystems Viva GS14 GNSS RTK GPS system (2014 to 2016).

3.2. Metrological variables

Precipitation (P) was measured using a tipping bucket rain gauge (Texas Instruments Canada Ltd. TR-525M) located at the upland meteorological station (Fig. 1) which recorded cumulative rainfall over 30-minute periods. Data gaps in recorded P were filled using a secondary tipping bucket (Onset Hobo RG3-M) located on the east slope. Evapotranspiration (ET) from the upland and fen was measured with eddy covariance systems at their respective meteorological stations. The specific details of meteorological instrumentation and protocols are reported by Ketcheson et al. (2017). Discharge (Q) was measured using a Palmer-Bowlus© type flume equipped with a logging pressure transducer in 2013, and through a v-notch weir in 2014–2016 (Fig. 1). A base-line Q of 0.14 L s^{-1} was added to the 2015 and 2016 outflow rates as Q through the V-notch was often nil, yet water was seeping into the spill box laterally though the peat deposit, resulting in a loss of water from the system. Q is expressed as a depth of runoff (R_{out}; mm) normalized to the upland-fen area (2.9 ha) for seasonal totals.

3.3. Transport calculations

Specific discharge in the horizontal (q_{horz}) and vertical (q_{vert}) directions, were calculated using Darcy's Law (Eq. (1); Freeze and Cherry, 1979).

$$q_{horz,vert} = -K_{sat} \cdot i_{horz,vert}, \qquad (1)$$

where saturated hydraulic conductivity (K_{sat}) for each construction material reported by Ketcheson et al. (2017), were used (Appendix A).

The mean advective mass flux (J; Eq. (2)) was determined by using the respective average q and average seasonal Na^+ concentrations (C_{avg}).

$$J_{horz,vert} = q_{horz,vert} \cdot C_{avg}, \qquad (2)$$

Horizontal flow (q_{horz}) from upland to fen was assumed to only be through the petroleum coke underdrain given its high permeability relative to the tailings sand (Ketcheson et al., 2017), determined by using the seasonal average i_{horz} (8 to 15 measurements per year) from 3 pairs of nests starting at the boundary between the upland and transition zone, to the fen margin (Fig. 1). Here, only the K_{sat} and C_{avg} of petroleum coke underdrain was used. J_{horz} (kg m^2 yr^{-1}) for the petroleum coke was applied across the layer thickness and width (0.7 m × 200 m = 140 m^2; Worley-Parsons Canada, 2014), and reported in kg yr^{-1}.

Vertical flow (q_{vert}) within the fen was determined by using the average i_{vert} between petroleum coke underdrain and water table from all 14 nests (8 to 15 measurements per year). The harmonic mean (Eq. (3); Freeze and Cherry, 1979), using respective layer thicknesses, each with the geometric means of all peat K_{sat} at 50, 90, and 150 cm bgs, from 2013 to 2016 to estimate an effective K_{sat} for the fen.

$$K_{sat} = \frac{b}{\sum_{i=1-3}^{n} {b_i}/{K_{sat,i}}}, \qquad (3)$$

C_{avg} for the petroleum coke underdrain were used in Eq. (2) to determine J_{vert} entering the fen, which was applied over the fen area (2.9 ha).

The aforementioned calculations for J_{horz} and J_{vert} were compared against the estimated vertical mass flux using the seasonal ET flux total (as previously mentioned) averaged over the total ice-free days for each season (~102 days per study period) to estimate a q_{vert}. C_{avg} of the petroleum coke underdrain was used to calculate the mass flux, J_{vert}, as before (Eq. (2)).

3.4. Pore water sampling and analysis

Pore water samples were extracted from a sub-set of wells and piezometers (at previously noted depths) from the hydrological monitoring network (Fig. 1). All wells and piezometers were purged (minimum 3 well volumes) ~24 h prior to water sample extraction. Standard sampling protocols included evacuating a volume of water (~50 mL) into a clean reservoir to measure electrical conductivity (EC), temperature (T), and pH using a multiparameter probe (Thermo Scientific™ Orion™ Star A329 pH/Conductivity Portable Multiparameter meter). EC probes were calibrated to 1413 μS cm^{-1} monthly and pH probes were three-point calibrated before each day used. Water samples were taken in 60 mL high density polyethylene vials, stored, and transported in a cooler, filtered through 0.45 μm nitrocellulose filters and frozen within 24 h. Frozen samples were shipped in coolers for analysis. Major cation (Na^+, Ca^{2+} and Mg^{2+}) and anion (Cl^- and SO_4^{2-}) concentrations were determined in all water samples by a Dionex ICS-1600 Method EPA 300.0 with AS-DV auto-sampler, with analytical precision to ±1.0 mg L^{-1}, or less. Field blanks, bottles filled with de-ionized water (<1 μS), and sample duplicates were taken periodically through the sample campaigns for quality assurance and quality control.

4. Results

4.1. Hydrological context

Total rainfall (P) between 17 May and 27 August 2013–2016, was 254, 193, 126 and 223 mm, respectively (Table 1). Nikanotee Fen Watershed received infrequent but intense P events, with only a few (2 to 6) major (>10 mm day^{-1}) events each season (Fig. 2). Large proportions of the seasonal P were received in the early half (May and June) of the 2013 and 2014 seasons and the latter half of these seasons were relatively dry, with events of similar frequency but lower intensity (<10 mm day^{-1}). The 2015 season received very little rainfall in May and June, with most received in July and August, albeit less than normal. In 2016, rainfall was more evenly distributed over the field season. The

Table 1
Meteorological variables (P, fen ET, and R_{out}) reported for the periods from 17 May to 27 August (day of year (DOY) 137 to 238) in 2013–2016. All values are expressed as depth (mm), and R_{out} is with respect to the fen area (2.9 ha).

Year	P	AET	R_{out}
2013	254	309	117
2014	193	381	132
2015	126	408	18
2016	223	365[a]	29[b]

[a] Measured values only available from 27 June 2016 to 19 August 2016 (DOY 179 to 232; 206 mm); seasonal total was based on average daily rate during this period.
[b] Value only from 17 May 2016 to 18 August 2016 (DOY 122 to 231).

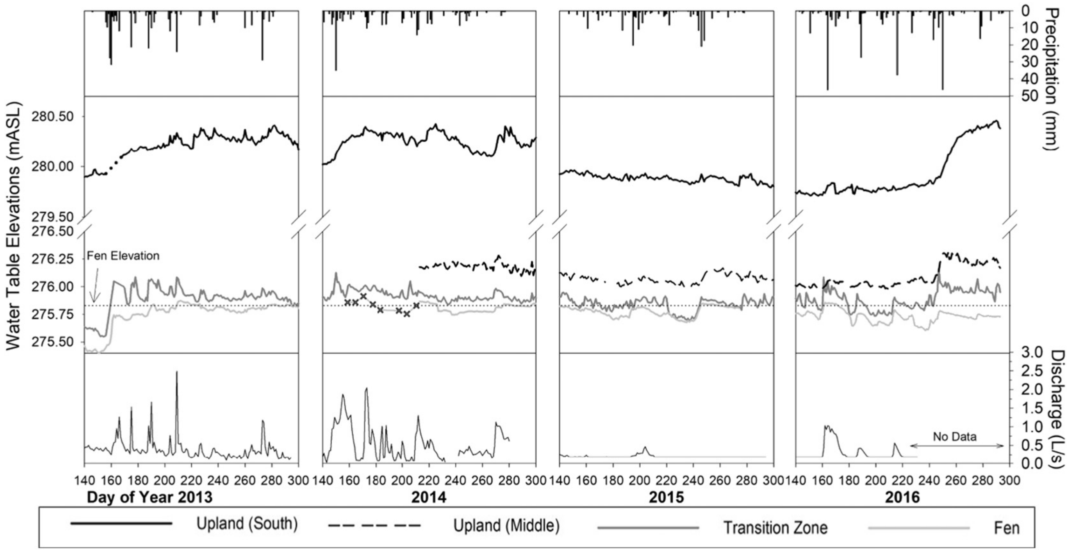

Fig. 2. Precipitation, runoff, and average water table elevations in the southern and central portions of the upland, transition zone, and fen for 2013–2016. Average fen elevation is also shown (note: depth to water table was less in the west, where ponded water was present, and greater in the east, due to variability in the local peat surface elevation; see Ketcheson et al., 2017 for a more detailed description of water table variability within the fen). Precipitation is expressed as daily total values.

fen ET rates ranged from 1 to 6 mm day^{-1}, with seasonal totals exceeding rainfall (Table 1). Q had a rapid response time to P, in 2013 and 2014 (<1 day) and maintained a base-line outflow (~0.1 mm day^{-1}) via groundwater seepage through the outlet during most of the seasons (Fig. 2). In 2015, Q was typically low, with only two periods of flow following the spring freshet. Q was also low in 2016, except for runoff caused by three separate precipitation events.

Given the water table elevations in the upland, transition zone and fen (Fig. 2) the general groundwater flow direction was south to north; from upland to fen (Fig. 3). The water table was parallel to the slope of the basal liner within the southern part of the upland, forming a hydraulic gradient (i_{horz}) of ~0.03. Within the upland, the water table was generally ~1.5 to 2 m bgs, except near the center of the upland where thicker tailings sand placement resulted in a water table that exceeded ~3 m bgs. Following large P events, the upland water table increased (Fig. 2), and mounding occurred in the south-east and east side of the upland proximal to the recharge basins (Fig. 3). This water table mounding increased the local i_{horz} (~0.05) along the east side of the upland for periods of up to 3 weeks following the recharge event. The upland water table flattened and i_{horz} diminished substantially across the transition zone, projecting near horizontally (~0.003) towards the fen outlet. The average water table within the transition zone was ~1 m

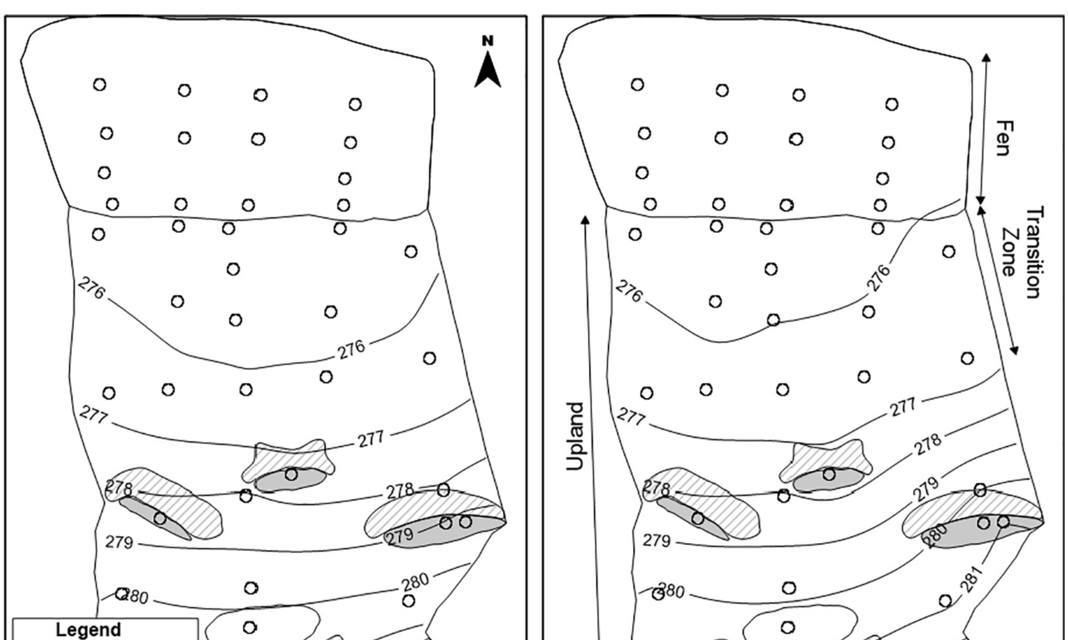

Fig. 3. Average water table elevation contours (left), and during typical recharge scenarios (i.e. mounding along the east side of the upland; right) in 2014, similar recharge scenarios can be seen in all years studied.

bgs near the fen and remained somewhat stable (±20 cm) throughout the duration of study. In the fen, the average water table was shallow and remained near the surface for the entire 2013 and 2014 seasons (Fig. 2). During relatively dry conditions in 2015, the average water table within the fen dropped to ~20 cm bgs for a portion of the season (Fig. 2). During this time, corresponding drops in the average water table elevation were observed within the upland (~30 to 40 cm) and the transition zone (~15 cm). Average upland water levels in 2016 were initially low, following the dry conditions experienced in 2015, and increased in the later portion of the season following heavy rainfall (Fig. 2).

4.2. Groundwater geochemistry

Groundwater electrical conductivity (*EC*) typically ranged between ~1000 and 4000 μS cm^{-1} in all construction materials (mean of 2550 μS cm^{-1}) with no notable differences between material types. *EC* was typically dominated by Ca^{2+} and SO_4^{2-} with lesser amounts of Na^+, Mg^{2+} and Cl^- (Table 2). Thus, *EC* could not reliably be used to determine Na^+ concentrations, but it did respond to seasonal inputs of fresh water. The petroleum coke underdrain was slightly basic (mean pH of 7.5) and the tailings sand upland was slightly acidic (mean pH of 6.5) in all periods studied. The pH of fen peat was generally neutral (mean pH of ~7.2) throughout the study.

In 2013, the tailings sand upland aquifer had the highest concentrations of Na^+, Cl^-, and SO_4^{2-} (average of 232, 53, and 882 mg L^{-1}, respectively), compared to other materials. In all subsequent years (2014–2016), the petroleum coke underdrain had the highest concentrations of these ions (maximum seasonal average of 303, 60, and 1190 mg L^{-1}; Table 2). Peat had the highest concentrations of Ca^{2+} and Mg^{2+} in all four years (303 and 110 mg L^{-1}, respectively). Peat also had notably high concentrations in SO_4^{2-} (maximum seasonal average of 934 mg L^{-1}; Table 2).

4.3. Na^+ distribution

Generally, Na^+ concentrations were highest within the tailing sand upland and petroleum coke underdrain, and lowest within the fen peat (Table 2). Decreases in Na^+ concentration occurred in all materials immediately following the substantial rainfall received in early 2013 and 2014. In the dry 2015 season, Na^+ concentrations within the tailings sand upland and petroleum coke underdrain were relatively high (>200 mg L^{-1}; Fig. 4). Na^+ concentrations were generally low within the fen peat throughout 2013 and 2014, but also increased during 2015 (Table 2 and Fig. 4). Na^+ concentrations were slightly lower in all construction materials in 2016, which was much wetter than the previous year, despite increasing in the near surface peat layers (50 cm bgs).

The spatial patterns of average Na^+ concentrations show relatively high concentrations throughout the tailings sand upland and petroleum coke underdrain beneath the transition zone in 2013 (mean of 216 mg L^{-1}; Figs. 5 and 6), with highest concentrations within the southwest portion of the upland (mean of 428 mg L^{-1}) and lowest immediately downslope (~5 m) of the peat-lined basin (mean of 202 mg L^{-1}). Within the fen, the average 2013 Na^+ concentrations were highest in the petroleum coke underdrain adjacent to the upland and decreased with increasing distance from the upland (Figs. 5 and 6). In 2014, following surface tilling and the creation of recharge basins (Fig. 3), Na^+ concentrations decreased by an average of 68% in the southeast region of the tailings sand upland and along the east side of the system (Fig. 5). Lower Na^+ concentrations were observed in the southeast region of the upland in 2015, although concentrations remained high (>175 mg L^{-1}), similar to 2013, throughout much of the upland and the transition zone (Fig. 5). Na^+ concentrations increased throughout the petroleum coke underdrain beneath the fen (303 mg L^{-1}) and in the basal peat layers (188 mg L^{-1}) by 2015 (Figs. 5 and 6). Na^+ concentrations generally decreased site-wide in 2016, including within the petroleum coke underdrain layer beneath the fen (226 mg L^{-1}; Figs. 5 and 6).

The i_{horz} that drive Na^+ transport towards the fen (Fig. 5) were much higher in the upland (~0.036) than in the transition zone and fen (~0.003), reflecting the slope of the water table (Figs. 3 and 6) as driven by the system geometry (Fig. 1). The vertical hydraulic gradients (i_{vert}) within the fen were usually upwards (average ~0.013) and generally stronger towards the toe of the upland (Fig. 5).

Table 2
Groundwater analysis for *EC*, major ions and compounds[a] within all construction materials for 2013–2016. Bolded values are the highest average that year (excluding wells). Locations of samples were kept identical between years to ensure unbiased interpretation. Sample numbers range from 8 to 48 for each material in each year.

Parameter	Year	Peat				Petroleum coke	Tailings sand
		Well	50 cm	90 cm	150 cm		
EC	2013	2250 ± 515	2040 ± 658	2010 ± 533	2020 ± 605	1490 ± 508	**2450 ± 1020**
(μS cm^{-1})	2014	2280 ± 545	2710 ± 1030	2399 ± 779	2620 ± 664	**3000 ± 380**	2370 ± 515
	2015	2770 ± 725	2700 ± 889	2450 ± 628	2660 ± 557	**2910 ± 384**	2330 ± 590
	2016	–	**3570 ± 1580**	2620 ± 1220	2680 ± 1080	3100 ± 782	2410 ± 911
Na^+	2013	119 ± 38.9	87.3 ± 35.5	78.5 ± 35.4	121 ± 66.3	200 ± 119	**232 ± 140**
(mg L^{-1})	2014	90.3 ± 64.9	79.7 ± 64.1	66.3 ± 42.9	97.4 ± 71.2	**182 ± 103**	138 ± 111
	2015	207 ± 110	153 ± 105	129 ± 73.8	188 ± 79.4	**303 ± 78.7**	238 ± 146
	2016	–	200 ± 126	123 ± 62.7	149 ± 83.5	**226 ± 83.1**	196 ± 108
Ca^{2+}	2013	411 ± 117	268 ± 144	278 ± 123	**284 ± 151**	161 ± 67.8	184 ± 134
(mg L^{-1})	2014	135 ± 76.1	**117 ± 69.8**	112 ± 49.5	99.6 ± 49.3	84.4 ± 32.4	69.7 ± 32.0
	2015	345 ± 133	285 ± 152	250 ± 87.5	**303 ± 84.6**	251 ± 48.5	225 ± 88.5
	2016	–	**219 ± 150**	124 ± 105	157 ± 80.7	169 ± 66.5	194 ± 82.0
Mg^{2+}	2013	59.6 ± 44.3	**52.3 ± 38.2**	48.8 ± 20.1	50.1 ± 32	33.2 ± 16.9	39.2 ± 30.6
(mg L^{-1})	2014	54.9 ± 51.1	**45.8 ± 35.2**	39.2 ± 29.7	36.2 ± 23.1	35.5 ± 29.3	26.3 ± 19.3
	2015	98.8 ± 32.7	**91.3 ± 42.7**	68.8 ± 31.9	78.2 ± 29.5	74.8 ± 20.6	65.1 ± 28
	2016	–	**110 ± 47.4**	79.1 ± 34.8	63.7 ± 31.5	63.2 ± 21.5	62.2 ± 22.8
Cl^-	2013	16.3 ± 7.30	14.2 ± 7.20	15.1 ± 7.5	28.1 ± 19.2	43.6 ± 34.9	**52.6 ± 48.1**
(mg L^{-1})	2014	23.2 ± 18.8	21.0 ± 19.4	22.2 ± 17.2	28.4 ± 21	**41.3 ± 29.1**	30.2 ± 35
	2015	48.5 ± 31.4	31.2 ± 17.9	28.8 ± 18.2	46.6 ± 20.2	**59.7 ± 21.1**	43.9 ± 40.1
	2016	–	39.1 ± 20.9	24.5 ± 14.2	36.7 ± 18.6	**47.8 ± 20.5**	38.5 ± 25.5
SO_4^{2-}	2013	1230 ± 544	658 ± 369	671 ± 397	798 ± 471	759 ± 349	**882 ± 716**
(mg L^{-1})	2014	560 ± 314	570 ± 367	490 ± 252	595 ± 297	**723 ± 310**	511 ± 253
	2015	961 ± 505	779 ± 566	596 ± 301	934 ± 271	**1190 ± 257**	964 ± 437
	2016	–	900 ± 625	442 ± 312	634 ± 230	**937 ± 242**	926 ± 290

[a] Annual mean (arithmetic) ± standard deviation (SD) of ion concentrations within all materials over the duration of the study period (2013–2016).

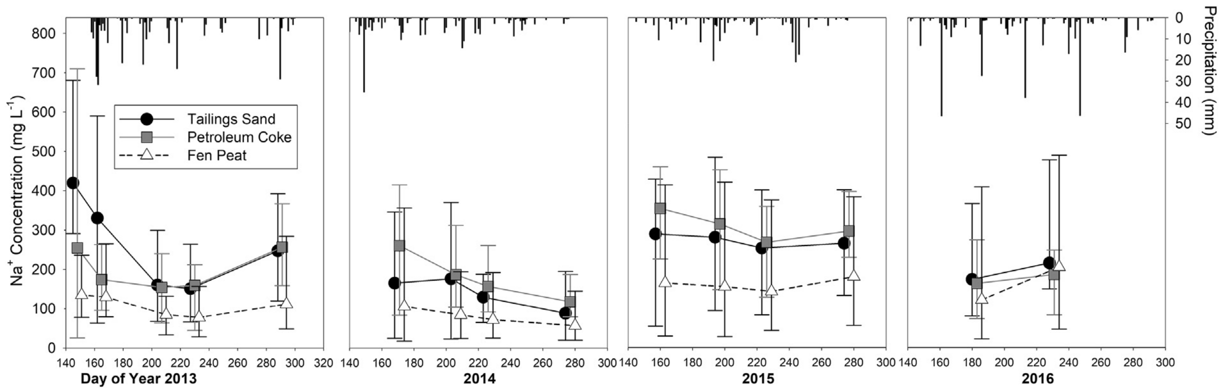

Fig. 4. Average Na^+ concentrations in construction materials over the duration of the four-year study period. Whiskers are the minimum and maximum observed concentrations. The fen peat values are averages and ranges of all peat layers. Daily precipitation is shown at the top.

5. Discussion

5.1. Transport of Na^+

Precipitation-driven recharge to the upland (Ketcheson et al., 2017), including intermittent runoff from the reclaimed slopes captured in recharge basins, was the driving force for water flow and thus the re-distribution of Na^+ within the system. Generally, groundwater flow was from tailings sand upland to petroleum coke underdrain (Fig. 2), and upwards into the fen peat (Fig. 1). The general patterns of Na^+ reflect its origin, thus higher concentrations in tailings sands, in 2013, and low concentrations in the fen (Figs. 5 and 6). Over time, the expectation is for freshwater recharge to flush the tailings sands, reducing Na^+ concentrations there, and progressively increasing these concentrations under and within the fen. Indeed, in 2014 there was a decrease in Na^+ concentration in the upland, and evidence the Na^+ plume was moving in the coke layer beneath the fen (Fig. 6). However, in 2015, a dry year, Na^+ concentrations increased, in both upland and in the coke layer beneath the fen, contrary to expectations (Table 2). Simhayov et al. (2017) found leachable concentrations of Na^+ in tailings sand was ~75 mg kg^{-1}, which equates to an equilibrium porewater concentration (assuming bulk density = 1.45 g cm^{-3} and porosity = 0.45; Appendix A) of 242 mg L^{-1}. While percolating water could leach some of this from the vadose zone, we do not believe this is an important source of Na^+, because recharge was low in 2015, and the upland water table declined ~22 cm from the previous year (Fig. 2). Moreover, Na^+ in the saturated zone of the upland was stratified, with higher concentrations at the basal layer (Appendix A). We believe the increase in Na^+ concentration in 2015 was an artefact of this stratification, and sampling with wells. Water samples drawn in wells during dry conditions (lower water table), preferentially sample deeper, more concentrated solutions. However, since the coke layer sits at the bottom of the profile, is of limited vertical extent (~0.7 m), is fully saturated and well mixed (high flow rate), we believe its changes in Na^+ concentration reflect inputs from the base of the tailings sand aquifer, and different dominant flowpaths in wet

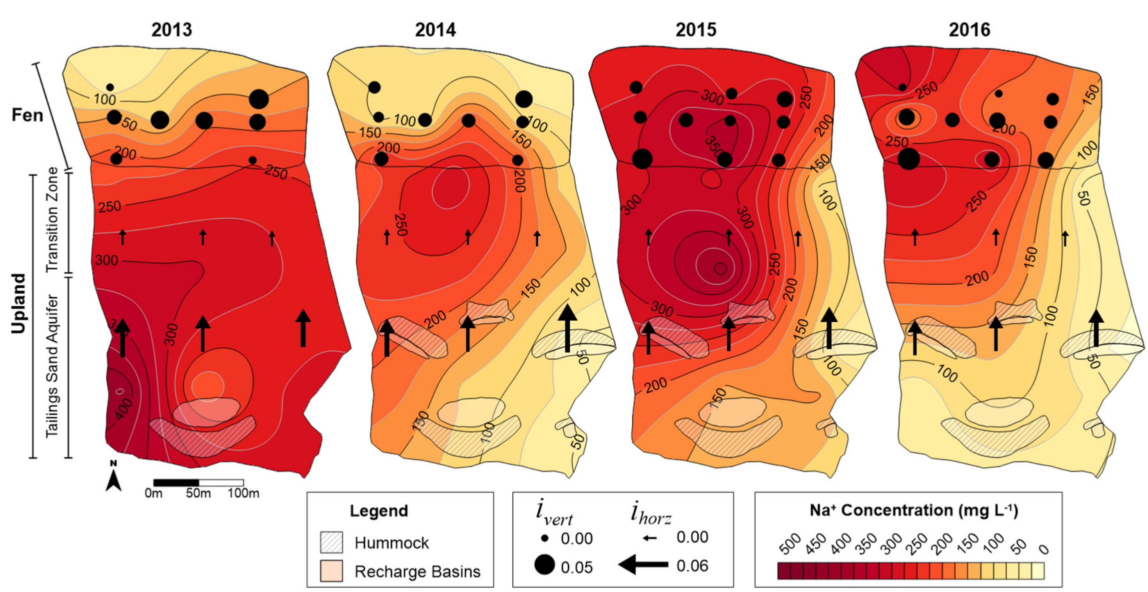

Fig. 5. Spatial distribution of the seasonal average Na^+ concentrations (colour scale; mg L^{-1}) within the tailings sand upland and petroleum coke underdrain (beneath peat) for 2013–2016. Included are seasonal average i_{horz} (upland and transition zone) and i_{vert} (fen) represented by arrows and circles, respectively.

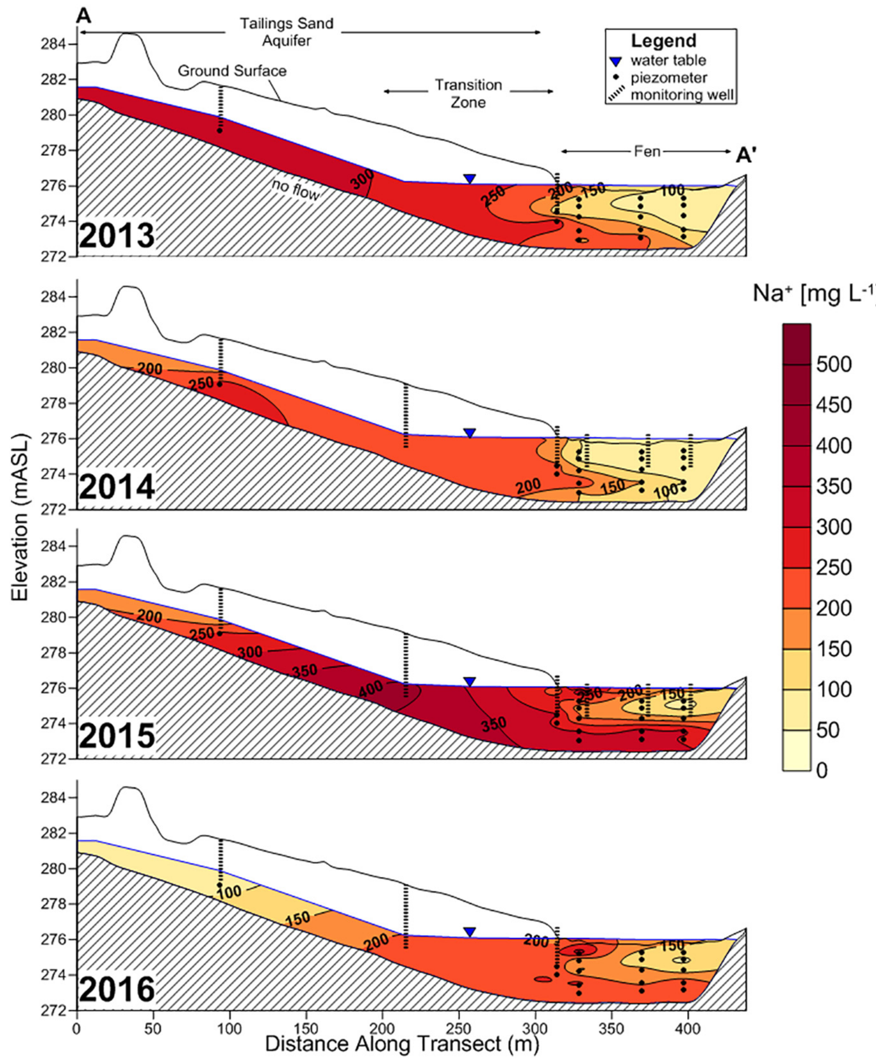

Fig. 6. Cross-sectional visualization of average (arithmetic) Na^+ concentration distributions in groundwater (colour scale; mg L^{-1}) along the primary A–A′ transect (Fig. 1) for 2013–2016. Piezometers are denoted by dots and wells are illustrated with hashed lines, both of which are to true sampling depths. The vertical exaggeration is 12:1.

and dry years, rather than sampling bias. During dry conditions when the water table in the sand aquifer is low, water and solutes flow mostly from the base of the water column directly into the up-slope face of the petroleum coke, which is at the approximate elevation of the water table (see flowlines in Fig. 1). This is supplemented by fresher water from the top of the water column that recharges across the top (horizontal) face of the coke layer (see Fig. 1). However, this is less in dry years, so Na^+ concentrations in the coke more strongly reflect basal concentration in the tailings sand.

Given the above, the most reliable approach to calculating the mass flux ($J_{horz,vert}$) of Na^+ from the upland to the fen, relies on using estimates of water and solute flux calculated in three ways: 1) using Eq. (2) with i_{horz}, K_{sat} and C_{avg} of the petroleum coke layer; 2) based on i_{vert} and the harmonic mean of K_{sat} within the fen (Eq. (3); Ketcheson, 2015) and C_{avg} in the petroleum coke underdrain; and 3) using ET, which is the primary water sink for the fen (Table 1), and C_{avg} in the petroleum coke underdrain. Based on these approaches, the mass flux across the respective flow areas in 2013, 2014, 2015 and 2016 ranged from 5280 to 6400, 1990–7200, 3440–12,800, and 2850–8560 kg yr^{-1}, respectively (Table 3). Note that the greatest mass flux was in 2015, the driest year. Estimating the mass flux is sensitive to measurements of i, K_{sat} and C, as well as choices of averaging techniques and time periods. Using multiple approaches to estimate the mass flux, as was done here, provides a range of values that more likely encompasses the true value.

5.2. Attenuation of Na^+ in peat

Peat can remove Na^+ from mobile water in active pores by diffusing into closed or dead-end pores (Hoag and Price, 1995, 1997) and by adsorbing to the peat surface (Rezanezhad et al., 2012b, 2016). However, Simhayov et al. (2017) showed in scanning electroscope images that the cellular structure of this particular peat was degraded, such that Na^+ transport could be modelled using a single porosity model with kinematic adsorption. While Na^+ was delivered quickly in the coke layer to the base of the peat profile, changes in concentration in the peat profile were slow (Fig. 6), because of its adsorption by peat (Table 2, Figs. 4 and 6), given the high initial availability of sorption sites (cf. Goldberg et al., 2007; Rezanezhad et al., 2012b). Solute retardation (R), which is the ratio of fluid velocity ($v = q/n_e$) to solute velocity (v_s) such that $R = v/v_s$, ranges from 1.22–3.07 for Na^+ (McCarter

Table 3
Average hydraulic and transport parameters for 2013 to 2016 for the transition zone (petroleum coke) and fen (peat). The flow direction in each region is listed. All values are in respective units indicated.

Year	Region	Flow Direction	i	K_{sat}[a]	q	C_{avg}	J	q wrt. fen area	q wrt. fen area	$J \cdot A$	J wrt. fen area
			–	m yr^{-1}	m yr^{-1}	mg L^{-1}	kg m^{-2} yr^{-1}	mm yr^{-1}	mm d^{-1}	kg yr^{-1}	kg m^{-2} yr^{-1}
2013	Transition	Horizontal	–	–	–	–	–	–	–	–	–
	Fen	Vertical	0.014	65	0.91	164	0.149	184	2.49	4330	0.149
	Fen *ET*	Vertical	–	–	1.11	164	0.181	309	3.03	5260	0.181
2014	Transition	Horizontal	0.0026	30,000	78	182	14.2	377	3.69	1990	0.051
	Fen	Vertical	0.015	65	0.98	182	0.177	259	2.67	5150	0.177
	Fen *ET*	Vertical	–	–	1.36	182	0.248	381	3.74	7200	0.248
2015	Transition	Horizontal	0.0027	30,000	81	303	24.5	391	3.83	3440	0.085
	Fen	Vertical	0.012	65	0.78	303	0.236	227	2.14	6850	0.236
	Fen *ET*	Vertical	–	–	1.46	303	0.442	408	4.00	12,800	0.442
2016	Transition	Horizontal	0.0030	30,000	90	226	20.3	434	4.26	2850	0.072
	Fen	Vertical	0.011[b]	65	0.72	226	0.162	200	1.96	4690	0.162
	Fen *ET*	Vertical	–	–	1.31	226	0.295	365	3.58	8560	0.295

[a] Values taken from Ketcheson et al. (2017).
[b] Low i_{vert} due to shorter seasonal measurement period (27 June to 27 August 2016).

and Price, 2017; Rezanezhad et al., 2012b; Simhayov et al., 2018). The average arrival time of solute at the fen surface (t_a), is

$$t_a = \frac{hR}{v}, \tag{4}$$

and given a typical vertical water flux of $q = $ ~338 mm per ice-free season (averaged 2014 to 2016; Table 3), effective porosity $n_e = 0.6$ cm^3 cm^{-3} (Simhayov et al., 2018), through 2 m thick peat (h), the average arrival time, t_a, ranges from ~4 to 11 years. This could be assisted by Na^+ being out-competed for sorption sites by other abundant ions including calcium (Stassart et al., 1981), which is abundant in peat porewater at this site (Table 2). Hydrodynamic dispersion will accelerate the front of the plume, and solute with reduced Na^+ concentrations than the input concentration at the coke layer (Table 2), will arrive much sooner. This is already evident at the 50 cm deep peat, in which Na^+ concentrations have risen from ~87 to 200 mg L^{-1} between 2013 and 2016. Na^+ concentrations at the surface are also likely to become elevated due to evapo-concentration (Simhayov et al., 2017; Simhayov et al., 2018).

5.3. Construction design implications on Na^+ transport

The findings of this study highlight the importance of integrating permeable underdrains and landform features that managed the redistribution of solute laden groundwater, in the case of Nikanotee Fen, distributing it beneath the fen area. It is possible to encourage early-season freshwater recharge through snow management strategies and surficial landform features that are designed to maximize water detention during the spring freshet. The recently constructed reclaimed hillslopes to the west and southeast generated surface runoff during precipitation events (Ketcheson and Price, 2016a) that were substantially infiltrated through the recharge basins (Kessel, 2016), and in furrows tilled across the LFH (Ketcheson, 2015). This freshwater recharge was most pronounced in the southeast of the upland (Fig. 5) near the southeast and east recharge basins that received water inputs from the adjacent hillslopes. The diluted groundwater flowed along the east side of the upland aquifer towards the fen, reducing Na^+ concentrations there (Fig. 5). Na^+ concentrations within the west and central regions of the tailings sand upland remained relatively high due to the more limited freshwater recharge; recharge basins on the west side have much smaller contributing areas and rarely contained ponded water (Kessel, 2016).

Up-scaling the design of future constructed fen ecosystems to those integrated into large-scale mine reclamation (10's of km^2) will likely necessitate a higher tailings sand to peat material ratios than at Nikanotee Fen due to limited peat availability; this will magnify the potential for elevated solute concentrations within reclaimed landscapes receiving water discharging from tailings sand sources. The thick (2 m) peat substrate in Nikanotee Fen was designed, in part, to delay and attenuate Na^+ migration to the fen surface (Daly et al., 2012); if thinner peat layers are used in future fen designs, earlier arrival times and higher peak concentrations could occur. Tailings sand aquifers placed at greater thicknesses and lower slope would increase vadose zone storage and reduce precipitation-driven groundwater recharge, which was shown to be an important control on Na^+ concentrations within the saturated zones of Nikanotee Fen watershed. The highly conductive underdrain layer was an important design feature in Nikanotee Fen, because it transmitted and dispersed most of the Na^+ enriched-groundwater beneath the peat deposit, which reduced the potential for shortcutting of Na^+-enriched groundwater directly into the fen along the toe of the upland. Future designs can incorporate layering of materials with variable hydraulic properties to better control the location of groundwater discharge zones where high solute concentrations can be expected.

5.4. Nikanotee Fen trajectory

Areas of the tailings sand aquifer that receive enhanced freshwater recharge from slopes and recharge features will likely 'flush' Na^+ in relatively short time frames and transition to less-saline water sources. Conversely, the remaining upland areas (mostly capped by LFH) will remain as longer-term sources of Na^+-enriched groundwater to the fen. Enhanced flushing of Na^+ (and other solutes) was substantial in recharge areas within the duration of this study; however, it is uncertain for how long these high-Na^+ groundwater sources will persist within Nikanotee Fen. Based on the water flux rates (q) reported here (Table 3), occurring only in the frost-free period (Ketcheson et al., 2017), and the volume of water in the saturated zone of the upland-fen portion of the system (V_{Nik}; ~140,000 m^3), the average residence time (average time a particle of water persists in a hydrologic system) of water (V_{Nik}/q) ranges from 11 to 24 years, averaging 15 years. The residence time of Na^+ will be somewhat longer, because of its retardation and attenuation within the peat material. The residence time of a Na^+ (total mass/mass flux out) within the upland tailing sand aquifer, given a total mass of 24.6 tons Na^+ (Simhayov et al., 2017) and the range of J from the upland to fen provided here (Table 3), is 7 to

44 years, averaging 15 years. Given the importance of freshwater recharge on controlling the migration of Na^+, changes in soil hydraulic properties (e.g. LFH infiltration; Ketcheson, 2015) will likely aid in diluting groundwater fluxes from upland to fen. However, as the adsorbed mass of Na^+ increases in the fen, the peat will lose its ability to decrease peak concentrations and arrival times of subsequent Na^+ pulses. As Na^+ concentrations increase in the rooting zone, the fen vegetation will favour more salt- tolerant species (Rezanezhad et al., 2012a; Trites and Bayley, 2008, 2009). Substantial increases in *EC* within the 50 cm bgs peat layer in 2016 eludes to the accumulation of solutes at the surface. The quality of water discharge from the system was not reported here, but must be considered as Na^+ accumulates at the fen surface, as these constructed wetlands will export solutes to downstream ecosystems.

6. Conclusions

Reclaiming landscapes from mine waste materials, as in the case of the Nikanotee Fen Watershed, will inherently incorporate mobile solutes that may affect newly planted vegetation and down-gradient ecosystems. As such, construction systems should consider designs to best manage both water and solute flows, which we have demonstrated can be done by layering materials of appropriate hydraulic properties. As expected, the tailings sand upland in Nikanotee Fen transmitted large amounts of solutes via groundwater to the adjacent peat deposit within the first several years after construction. The petroleum coke layer directed this underneath the fen. The 2 m thick peat deposit then delayed the arrival time of Na^+ to the fen surface; however, it may not be feasible to use similar peat thicknesses in future peatland designs, given the resource limitation. As such, planting relatively salt-tolerant species is recommended. As Na^+ begins to accumulate within the rooting zone of Nikanotee Fen, increased sampling resolution paired with research focusing on solute cycling in the rooting zone, and plant responses, will provide insight into its potential ecohydrological feedbacks.

In Nikanotee Fen, the petroleum coke underdrain was essential in directing and distributing Na^+ beneath the fen, as designed, which despite increasing the rate of groundwater flow, allowed for relatively even distribution of Na^+ beneath the fen. This greatly reduced the potential for 'hotspots' of high peak concentrations. Thus, it is recommended that future designs incorporate layered materials to create sub-surface hydrological connections that control the flowpath of solutes. Furthermore, underdrain designs could be used to re-direct Na^+-enriched groundwater fluxes to target more salt-tolerant areas, and away from wetland areas with less tolerant species. In addition to underdrain designs, it was evident that recharge features, especially recharge basins, had an influential role on the migration of Na^+; however, their implications on the enhanced flushing of Na^+ (and other solutes) must be investigated further.

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Appendix A

Table A
Bulk density (ρ_b), porosity (n), and saturated hydraulic conductivity (K_{sat}), adapted from Ketcheson (2015), with updated values from Ketcheson et al. (2017) and Simhayov et al. (2017).

Properties		Construction Materials				
		LFH	Tailings Sand	Petroleum Coke	Peat 0–50 cm	Peat 50–200 cm
ρ_b (g cm^{-3})	Average	1.33	1.45	0.64	0.18	0.22
	±SD	0.19	0.14	–	0.04	0.03
	n	21	19	5	36	28
n (cm^3 cm^{-3})	Average	0.5	0.45	0.45[a]	0.92	0.87
	±SD	0.07	0.05	0.07[a]	0.02	0.02
	n	21	19	15	36	28
K_{sat} (m s^{-1})	Average*	1×10^{-5}	4×10^{-6}	9.5×10^{-4}[b]	8×10^{-5}	2×10^{-6}
	Min	5×10^{-7}	1×10^{-7}	–	5×10^{-5}	3×10^{-8}
	Max	2×10^{-4}	3×10^{-5}	–	2×10^{-4}	4×10^{-5}
	n	21	58	–	12	127

Average = arithmetic mean, Average* = geometric mean, SD = 1 standard deviation, n = sample size.
[a] Values taken from Simhayov et al. (2017).
[b] Value taken from Ketcheson et al. (2017).

Appendix B

B.1. Geochemical stratification

Three monitoring locations along the southern transect in the upland (Fig. 1) and the local proximity to the east recharge basin displayed distinct geochemical stratification within the tailings sand aquifer, due to freshwater recharge. Electrical conductivity (*EC*) in the upper layers of the tailings sand aquifer, measured from monitoring wells (maximum sample depth of 279.25 mASL), was consistently lower than that in the basal layer, measured from deep piezometers (260 to 275 cm bgs slotted screen; 279.10 to 297.03 mASL; Fig. B-1). Thus, a vertical concentration gradient existed within the saturated zone of the tailings sand, or at least across the saturated zone from the water table to deep piezometer screen intake (saturated layer thickness ranging 52 to 224 cm). Vertical concentration gradients indicated by *EC* were most pronounced along the east side of the upland (Fig. B-1b and d), where the bottom layers had up to twice the *EC* of the upper layers, as in the 2015 season. In all seasons, with the exception of the east recharge basin (Fig. B-1d), the downward movement of water, indicated by a negative i_{vert}, was lowest (positive or near zero) during the wettest periods and seasons, and highest (negative) during dry periods, thus commonly decreased over the season (Fig. B-1a, b and c). Corresponding to this increased downward flux of water, *EC* gradients diminished, with deep piezometer *EC* indicating a convergence on the *EC* values measured by the well.

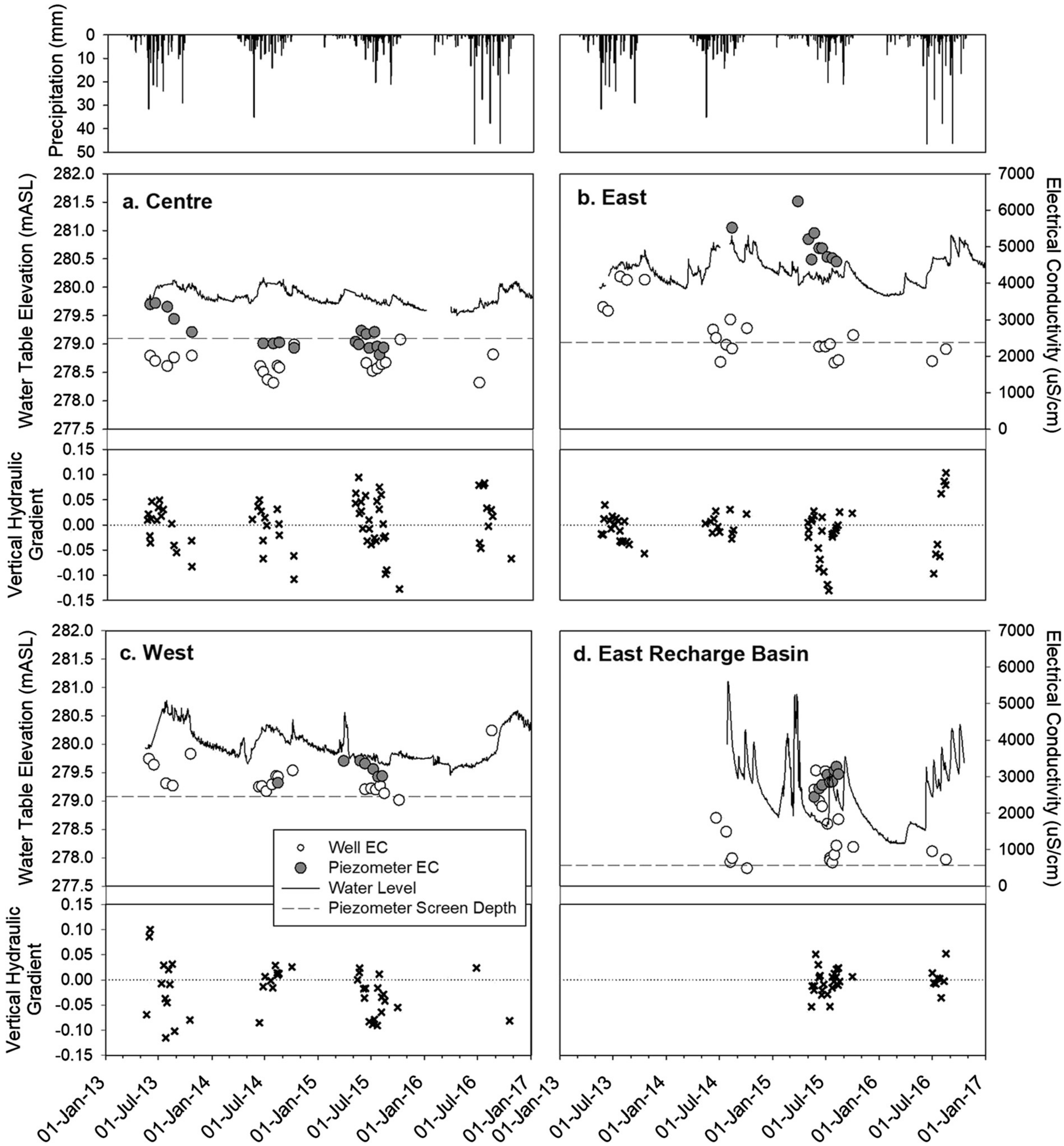

Fig. B-1. 2013 to 2016 water table (black lines) and electrical conductivity (white for well and grey for piezometer) for a) center, b) east, c) west and d) east recharge basin areas of the upland. i_{vert} are from the water table indicated by the monitoring well to the deep piezometer, with a negative value being a downward flow direction.

We were unable to explain the apparent rise in Na^+ concentration in the tailings sand aquifer during dry periods based on fundamental geochemical processes, such as by desorption from the solid phase or flushing of solutes from the vadose zone. Rather, we believe the decrease in concentration following substantial rainfall (>10 mm day^{-1}) is an artifact of using wells that sample preferentially from the top of a diluted and geochemically stratified saturated zone. The addition of relatively fresh water infiltrating through the LFH layer, and percolating down through hydrophobic sands (Ketcheson, 2015) towards the water table, could occur as finger flow (Bauters et al., 1998) or through macropores (Guebert and Gardner, 2001), both which bypass most of the matrix in the unsaturated zone, thus limiting salt leaching. This occurs on a magnified scale in the recharge basins, which recharge a substantial amount of freshwater through a very small part of the upland area (5% fractional area). The relatively fresh recharge water overlies the resident (older) groundwater. The data show (Fig. B-1) upland areas with the highest degree of freshwater recharge (i.e. east; Fig. B-1b and d) had the strongest concentrations gradients (greatest stratification). Conversely, upland areas with the lowest degree of freshwater recharge had lower concentration gradients (i.e. center and west; Fig. B-1a and c). Following prolonged periods without rain, this geochemical stratification diminishes through lateral groundwater flow down-slope and through other mixing processes (transverse dispersion and diffusion). Without this freshwater layer in the upper portion of the saturated region, water samples extracted from the wells yield a higher Na^+ concentration. We believe this is why there is an apparent increase in concentration evident in dry periods (Figs. 4, 5 and 6), rather than due to an input of solute mass into the porewater.

Geochemical stratification in the tailings sand aquifer has direct implication for the amount of Na^+ transmitted as during periods of rainfall, when stratification occurs, the petroleum coke underdrain is fed by this upper portion of relatively lower Na^+ groundwater as evident by a lower J (Table 3). Conversely, in dry periods, without geochemical stratification the petroleum coke is fed by water sources deeper within the aquifer water column which contains higher Na^+ concentrations, resulting in a higher J (Table 3). The effects of geochemical stratification was not anticipated when designing Nikanotee Fen but its importance on Na^+ migration warrants investigation in future research.

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