Effects of Capping Strategy and Water Balance on Salt Movement in Oil Sands Reclamation Soils

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Abstract: The success of oil sands reclamation can be impacted by soil salinity depending on the materials used for soil reconstruction and the capping strategies applied. Using both a greenhouse-based column experiment and numerical modeling, we examined the potential pathways of salt migration from saline groundwater into the rooting zone under different capping strategies (the type and the thickness of the barrier layer) and water balance scenarios. The experimental results showed that there would be salinity issues in the cover soil within several growing seasons if there was a shallow saline groundwater table and if the soil was not properly reconstructed. The thickness of the barrier layer was the most significant factor affecting the upward movement of saline groundwater and salt accumulation in the cover soil. The suitable thickness of the barrier layer for preventing the upward movement of saline groundwater and salt accumulation in the cover soil for each material varied. A numerical simulation for a 15-year period further indicates that, when the cover soil was 50 cm of peat-mineral soil mix and when wet, dry, or normal climatic conditions were considered, the minimum barrier thickness to restrain salt intrusion into the cover soil in the long term was about 75 or 200 cm for coarse tailings sand or overburden barrier material, respectively. In view of the above, to minimize salt migration into the rooting zone and ensure normal plant growth, oil sands reclamation should consider salt migration when designing soil capping strategies.

Keywords: barrier material; barrier thickness; land reclamation; oil sands; saline groundwater; salinity; water balance

1. Introduction

In the Athabasca Oil Sands Region (AOSR) of Canada, surface mining for oil sands have completely altered the landscape from boreal forests and wetlands into mine pits, dumps, and tailings ponds. By 2017, the disturbed lands already covered an area of 895 km², which account for about 0.2% of Alberta’s boreal forest [1]. As regulated by the government, the disturbed lands need to be reclaimed. Taking restoration of ecological function and bio-diversity as a priority, all the reclamation practices are required to re-establish the functional boreal ecosystems with equivalent capability prior to disturbance [2]. Meanwhile, the mining and processing operations produce large volumes of mining by-products that must be disposed of properly [3]. Soil covers are used to cap disturbed lands to meet equivalent land capability. These reconstructed soil profiles usually have a top layer (also known as cover soil) that provides nutrients for vegetation and an underlying barrier layer (subsoil) to insulate the mining wastes beneath as well as provide more relatively clean media suitable for root growth [3,4].
The major waste materials requiring capping in oil sands reclamation are coarse tailings sand and overburden. Coarse tailings sand is produced during bitumen extraction, and overburden is the natural sediments between the oil sands layer and the soil horizons, which needs to be excavated before mining [3]. A large concern when dealing with these materials is that some of them contain high concentration of salts [5]. The salts in the coarse tailings are mainly sourced from the process aids such as caustic (NaOH) and sodium citrate for bitumen extraction. Overburden from the Clearwater Formation above the oil sands is a natural highly saline sediment. Salts released from the sediment after disturbance becomes another important source [5].

Due to the mobility of sodium, the capping strategy carries the risk that the salts in the underlying materials may migrate into the rooting zone and then negatively affect the vegetation. Soils are considered “unsuitable” for plants when the electrical conductivity (EC) reaches or exceeds 8 dS/m [6]. At these high salinity levels, the physiological functions of most boreal forest species would be restricted, and result in a failure of the reclaimed lands to meet the capability requirements [7]. In the AOSR, soil salinity has been one of the substantial issues that impacts the success of reclamation [8–11]. Improper barrier materials and insufficient barrier thickness were the common causes of soil salinization in oil sands reclamation [5,11–13]. Overburden and coarse tailings sand have opposite soil texture and physical properties. Fine-textured overburden has strong water holding capacity but weak infiltration ability. It preserves moisture and promotes plant growth but creates the risk of salinization due to capillarity. On the contrary, coarse-textured tailings sand has high hydraulic conductivity but poor water holding capacity. Salts can easily be flushed down with water flow, but the material may not be able to preserve enough water for plant growth [3,14,15]. Therefore, the barrier thicknesses were different when different barrier materials were used. According to a study at Syncrude Canada Ltd., where tailings sand was used as a barrier material, cover soils were salinized when a saline water table was within 1 m of the soil surface, at high risk of salinization when the saline water table was within 1.5 m, and low risk only when the saline water table was below 2 m [3].

Hydrology is another critical factor that influences soil solute transport and salinization [4,8,16,17]. Reclamation aims to rebuild the landscapes with a mosaic of uplands and wetlands [3,4,16]. Under these reconstructed undulating terrains, water tables varied at locations and fluctuated seasonally [8]. Soils in the discharge areas of the reconstructed watershed would always have high risk of salinization [3,11,18]. Water supply and precipitation also aggravate the situation because more salts would flow in with water from the other parts of the watershed. In the other areas, soil salts are expected to be gradually diluted and leached out. However, if deep percolation is not occurring, these soils can also be salinized due to the active hydrologic connections between shallow saline groundwater and the upper cover soils. Increased water supply and precipitation will also stimulate salinization at some locations because of a rise in the water table and enhancement of evaporation [17,19].

A better understanding of salt migration under different capping strategies and different hydrologic conditions will help develop better land reclamation practices to deal with soil salinity. However, the effectiveness of these capping strategies has not been fully verified. Many questions on how reclamation materials and their configuration affect soil water movement and salt accumulation remain unanswered. Moreover, the hydrologic impacts on potential salt migration also need further study. Given the above, this paper examined the potential pathways of salt migration into the rooting zone to affect plants using a greenhouse-based column experiment and numerical simulation, and tried to address three key questions: (1) how does the texture of the barrier material affect salt movement? (2) what thickness of the barrier material will minimize salt intrusion into the rooting zone in reconstructed soils in the presence of a shallow saline groundwater table? and (3) how does water balance affect salt movement in reconstructed soil profiles?
2. Materials and Methods

2.1. Column Experiment

2.1.1. Experimental Design

A greenhouse-based column experiment was set up at Alberta Innovates—Technology Futures (now InnoTech Alberta) in Vegreville, AB, Canada in summer 2013. It became operational in October the same year and was destructively sampled in May 2015 after 3 simulated growing seasons (October 2013–January 2014, April 2014–September 2014, and January 2015–April 2015). A completely randomized block design was used in the experiment with barrier material type, barrier thickness, and water balance at 2 levels (simulated dry vs. simulated rainy condition for the oil sands mining region in Canada).

The two barrier materials with contrasting soil textures, TS and OB, and the cover soil material, peat-mineral soil mix (PMM), were all sourced from the Canadian Natural Resources Limited Horizon mine, north of Fort McMurray, Alberta, Canada. The materials all had low salinity and alkalinity (Table 1). TS and OB had low organic carbon contents (<0.5 g kg\(^{-1}\)), and PMM was a mixture of peat and mineral soil with high porosity and organic carbon content (>5 g kg\(^{-1}\)). The materials were packed according to pre-determined bulk densities (Table 1) in columns (45 cm in diameter and 180 cm in height) with the following layers (Figure 1): (1) The top layer (cover soil): all columns were packed with 50 cm of PMM as cover soil. (2) The barrier layer: TS and OB were packed under the cover soils as barrier materials. Their thicknesses varied from 0, 20, 50, to 100 cm in different treatments. And (3) The bottom layer: The same TS was placed under the barrier materials to keep all columns to the same height (180 cm) (Figure 1). This layer was submerged into saline water (3 g L\(^{-1}\) of NaCl) to mimic the salinity threat from underneath in the field such that the water table depth ranged from 50 cm to 150 cm (Table 2). Water levels in the columns were controlled at the interface between the barrier and the bottom layers using Mariotte bottles, and the amounts of saline water added to the bottles were recorded. Two more controls, which had cover soil and TS or OB barrier layer but without saline groundwater, were also included as part of the experiment (Figure 1 and Table 2). With 3 replications for each treatment, a total of 54 columns were set up.

In each column, four jack pine (Pinus banksiana) seedlings were planted at the beginning of the experiment. Water balance treatment was started in October 2013, 3 months after the seedlings were planted. Precipitation was mimicked using tap water sprinkled on the top of the columns. Average water supplies during the entire experiment were controlled at 260 and 385 mm per growing season for dry and wet conditions, respectively. During the growing season, air temperature in the greenhouse was maintained at 22 °C for 16 h of day and at 16 °C for 8 h of night to simulate the summer condition in the oil sands region.

Figure 1. Cont.
Figure 1. Column design with treatments of barrier material, barrier thickness, and water balance. The positions of GS3 sensors are illustrated by the symbol .

Table 1. Initial soil properties in the column experiment.

| Materials | Texture            | Bulk Density (g cm$^{-3}$) | EC (dS m$^{-1}$) | pH   | Porosity (cm$^3$ cm$^{-3}$) |
|-----------|--------------------|----------------------------|------------------|------|-----------------------------|
| PMM       | Sandy clay loam,   | 0.89                       | 1.42             | 7.52 | 0.66                        |
| TS        | Sand               | 1.37                       | 2.59             | 8.14 | 0.48                        |
| OB        | Silty clay         | 1.65                       | 1.55             | 7.53 | 0.30                        |

1 TS, OB, and PMM are abbreviations for tailings sand, overburden, and peat mineral-soil mix, respectively.

Table 2. A list of treatment codes and details of the design.

| Treatment  | Barrier Material | Barrier Layer Thickness (cm) | Saline GW Depth (cm) | Water Balance $^3$ |
|------------|------------------|------------------------------|-----------------------|--------------------|
| CK_TS+     | TS               | 50                           | No saline GW          | +                  |
| CK_TS−     | TS               | 50                           | No saline GW          | −                  |
| CK_OB+     | OB               | 50                           | No saline GW          | +                  |
| CK_OB−     | OB               | 50                           | No saline GW          | −                  |
| NC+        | NC               | 0                            | 50                    | +                  |
| NC−        | NC               | 0                            | 50                    | −                  |
| 20TS+      | TS               | 20                           | 70                    | +                  |
| 20TS−      | TS               | 20                           | 70                    | −                  |
| 50TS+      | TS               | 50                           | 100                   | +                  |
| 50TS−      | TS               | 50                           | 100                   | −                  |
| 100TS+     | TS               | 100                          | 150                   | +                  |
| 100TS−     | TS               | 100                          | 150                   | −                  |
| 200OB+     | OB               | 20                           | 70                    | +                  |
| 200OB−     | OB               | 20                           | 70                    | −                  |
| 500OB+     | OB               | 50                           | 100                   | +                  |
| 500OB−     | OB               | 50                           | 100                   | −                  |
| 1000OB+    | OB               | 100                          | 150                   | +                  |
| 1000OB−    | OB               | 100                          | 150                   | −                  |

1 CK, NC, TS, OB, “+”, and “−”, represent control, no barrier layer, tailings sand, overburden, wet and dry conditions, respectively. 2 Saline GW depth: saline groundwater depth. 3 Irrigation intensities were 385 and 260 mm per growing season on average for the wet (+) and dry (−) conditions, respectively.
2.1.2. Instrumentation and Regular Monitoring

Soil dielectric permittivity (ε), bulk EC (EC_b), and temperature data in the materials were recorded hourly during the experiment by multi-functional Decagon GS3 probes. In each column, two probes were installed in the cover soil (PMM) at 20 and 40 cm depths, the other probes were installed in OB or TS, and all of them were above the groundwater level (Figure 1). Soil volumetric water contents (VWC) were calculated from ε values using one of the following calibration equations specific for the media:

\[
\text{TS: } VWC = 4.0559 \times \varepsilon - 2.244 \quad (R^2 = 0.99) \tag{1}
\]

\[
\text{OB: } VWC = 20.035 \times \ln(\varepsilon) - 15.844 \quad (R^2 = 0.93) \tag{2}
\]

\[
\text{PMM: } VWC = 0.0032 \times \varepsilon^3 - 0.2336 \times \varepsilon^2 + 6.1369 \times \varepsilon - 0.959 \quad (R^2 = 0.99) \tag{3}
\]

In the study, EC_b was first converted to the EC of pore water (EC_p), and then the EC of saturated paste (EC_sp), and total dissolved solids (TDS) were calculated using VWC, EC_p, bulk density and porosity [20].

2.1.3. Final Sampling and Data Analysis

At the end of the third growing season, soil columns were destructively sampled. Soils were sampled by layers, and the final salt concentrations in the cover soil and barrier materials were determined and compared with the initial values in Table 1. Plant growth and root distribution under different treatments were assessed. The treatment effects on water dynamics and salt accumulation in the soil profiles and plant response after 3 growing seasons were analyzed using ANOVA and conditional inference tree (CIT) tests [21].

2.2. Numerical Modeling

2.2.1. Model Description

Numerical modeling was conducted to expand our research duration and understand how soil salinity evolves over the longer term and tried to find out the minimum safe barrier thickness to prevent salt intrusion into the rooting zone for the PMM and TS and PMM and OB designs, respectively.

A soil water flow and solute transport model, HYDRUS-1D, which was designed to analyze water and solute movement in unsaturated, partially saturated, or fully saturated porous media [22] was used to complete this part of the work. It included the following aspects: (1) Developing soil hydraulic and solute transport parameters by interpreting/simulating water and salt dynamics in the columns; (2) evaluating the long-term performance of different capping configurations under various climatic conditions; and (3) proposing recommendations for the design of reconstructed soil profiles when using PMM and TS or PMM and OB combinations that will minimize salt migration into the rooting zone.

2.2.2. Parameterization and Model Validation

Possible ranges of the parameters required by HYDRUS-1D were firstly synthesized from previous studies: the field saturated hydraulic conductivities for PMM ranged from 103.7 to 1123.2 cm day\(^{-1}\) [23] or 86.4–691.2 cm day\(^{-1}\) [3], K_s for TS ranged from 3.5 to 138.2 cm day\(^{-1}\) [11] or 17.28 cm day\(^{-1}\) [24], and K_s for OB ranged from 4.1 to 172.8 cm day\(^{-1}\) [23] or 0.3–69.1 cm day\(^{-1}\) [3]. Bulk soil diffusion coefficients estimated for each material varied from 1.1 \times 10^{-14} to 1.4 \times 10^{-10} m^2 s^{-1} for the PMM, and from 5.0 \times 10^{-12} to 6.5 \times 10^{-11} m^2 s^{-1} for the OB [5].

With the historic data reported in references as detailed above, a Marquardt-Levenberg type parameter estimation technique was applied to determine the parameters based on our experimental data [22]. Soil bulk density (BD), residual water content (Q_r), saturated water content (Q_s), α and n in the van Genuchten soil water retention function, saturated hydraulic conductivity (K_s), and solute transport parameters such as longitudinal dispersivity (Disp), and molecular diffusion coefficient in free water (Diff_w), were all estimated using the above method.
The upper and lower boundaries for water movement modeling were set up as an atmospheric boundary with surface layer and constant water content. The water content at the lower boundary equaled its saturated water content (water potential equals 0). For solute transport, constant flux concentration and variable concentration boundary conditions were used as the upper and lower solute transport conditions. Among them, irrigation, evaporation, transpiration, salt concentration in the irrigation water, and salt concentration in the recharging groundwater were temporally variable. Irrigation water in the model used the real data recorded. Salt concentration in the irrigation water was taken as 0, and salt concentration at the lower boundary was time-variable, and its values were set according to the monitored EC data from GS3 probes. Potential evapotranspiration (PET, mm) was estimated based on temperature at soil surface (T_{soil}) using the following equation [25]:

\[ \text{PET} = 0.254 \times 1.07^{1.8T_{soil}} \]  

(4)

Potential transpiration (PT, mm) was calculated as:

\[ \text{PT} = \left(0.3 + 0.4 \frac{t}{t_{r-max}}\right) \text{PET} \]  

(5)

where \( t \) is the time (day) after the seedling was planted, and \( t_{r-max} \) is the time (day) that the root grows to its max length. Evaporation was the difference between PET and PT.

Soil water dynamics were validated by VWCs at depths. Soil salt transport was validated by comparing the observed and simulated TDS in pore water at different depths in the column. Mean error (ME), mean absolute error (MAE), and root mean squared error (RMSE) were used to evaluate the fitness of the model.

\[ \text{ME} = \frac{1}{n} \sum_{1}^{n} (x_m - x_s) \]  

(6)

\[ \text{MAE} = \frac{1}{n} \sum_{1}^{n} \left| x_m - x_s \right| \]  

(7)

\[ \text{RMSE} = \left( \frac{1}{n} \sum_{1}^{n} (x_m - x_s)^2 \right)^{0.5} \]  

(8)

where \( n \) is the number of validation points, \( x_m \) is the experimental data, and \( x_s \) is the simulated data [26].

2.2.3. Scenarios for Salinization Risk Assessment

The validated HYDRUS-1D model was run for a 15-year period under different barrier material, barrier thickness and water balance scenarios. In the simulation, barrier materials used PMM and TS or PMM and OB combinations; barrier thickness of TS or OB was set as 50, 75, 100, 150, or 200 cm; and wet, dry and normal climatic conditions were considered. The normal climatic condition in the simulation used real weather recordings from 2001 to 2015 at the weather station called Fort McMurray A (weather.gc.ca); and the wet and dry conditions were set to have 130% or 70% of the normal precipitation with other meteorological parameters the same as the normal condition.

For each scenario, modeling was performed only for the growing season (1 May to 31 October). It was assumed that there was no evaporation loss and no loss in the form of snow in winter. Snow accumulation in winter would melt in the first 15 days at the beginning of the growing season. The PET was calculated using Equation (4). The PT was set as half of PET, and the root distribution in different materials was based on the final root distribution data in the column experiment. The lower boundary conditions were the same as those in the simulation for the column experiment. Groundwater levels were maintained at the bottom of the barrier layer. Saline groundwater was the only external salt source for the system and TDS in the groundwater was 3 g L\(^{-1}\).
3. Results and Discussion

3.1. Experimental Results

3.1.1. Treatment Effects on Soil Water Dynamics

During the experiment, barrier materials or thicknesses showed apparent effects on soil water contents above the groundwater ($p < 0.01$, ANOVA), while the water balance treatment showed slight effects (Figure 2 and Table 3). Cover soils were generally moister when there was no barrier layer, or the barrier thickness was thin ($\leq 50$ cm). As the barrier thickness increased, VWCs in the cover soils generally decreased regardless of the barrier material. Highest VWCs in cover soils (at 20 and 40 cm depths) were found in treatments without a capping layer; these values were significantly higher than most of the other treatments.

Figure 2. Average soil volumetric water contents in the soil profile during the experiment. Different letters in a row indicate significant differences ($p < 0.05$, ANOVA) among treatments at the same depth, which were derived by comparing all the values across the NC, OB and TS trials.
Table 3. Statistical results of the treatment effects on soil water, EC and plant after 3 growing seasons.

| Soil VWC (cm$^3$ cm$^{-3}$) | NC | OB | TS | 0 cm | 20 cm | 50 cm | 100 cm | CK | − | + |
|-----------------------------|----|----|----|------|-------|-------|-------|----|---|---|
| 20 cm                       | 0.46 | 0.31 | 0.29 | 0.46 | 0.34  | 0.30  | 0.28  | 0.28 | 0.31 | 0.33 |
| 40 cm                       | 0.59 | 0.37 | 0.35 | 0.59 | 0.49  | 0.35  | 0.28  | 0.28 | 0.38 | 0.38 |
| 60 cm                       | 0.23 | 0.16 | 0.27 | 0.19 | 0.17  | 0.15  | 0.20  | 0.20 |
| 90 cm                       | 0.24 | 0.21 | 0.30 | 0.19 | 0.20  | 0.20  | 0.23  | 0.24 |
| 140 cm                      | 0.23 | 0.38 | 0.33 | 0.22 | 0.29  | 0.29  |

| Soil EC (dS m$^{-1}$) | NC | OB | TS | 0 cm | 20 cm | 50 cm | 100 cm | CK | − | + |
|-----------------------|----|----|----|------|-------|-------|-------|----|---|---|
| 20 cm                 | 6.00 | 2.79 | 2.42 | 6.00 | 7.24  | 0.85  | 0.71  | 0.88 | 3.00 | 2.97 |
| 40 cm                 | 4.76 | 3.27 | 2.83 | 4.76 | 6.88  | 2.66  | 1.00  | 0.90 | 3.12 | 3.43 |
| 60 cm                 | 5.78 | 3.82 | 7.71 | 7.26 | 2.66  | 1.29  | 4.34  | 5.69 |
| 90 cm                 | 6.59 | 3.72 | 8.44 | 4.39 | 2.18  | 5.56  | 5.53  |
| 140 cm               | 8.13 | 7.45 | 9.94 | 7.89 | 7.83  |

| Plant (g) | AGB $^1$ | 117.25 | 255.52 | 182.86 | 117.25 | 265.20 | 226.67 | 205.00 | 197.33 | 196.05 | 235.47 |
| TR $^2$   | 42.31 | 90.55 | 69.65 | 42.31 | 81.93 | 88.44 | 78.55 | 79.30 | 73.35 | 83.10 |

$^1$ AGB: aboveground biomass in a column after the experiment; $^2$ TR: total root in a column after the experiment.

Intense up-flux of saline groundwater means a high risk of rooting zone salinization, and the salts coming up with capillary water have been considered an important cause of the salinity in the rooting zone in oil sands reclamation [24,27]. Since soil moisture regimes in the columns remained almost the same at the beginning and the end of saline water application, the amount of saline water added to the Mariotte bottles generally equaled the net amount of upward capillary water. It was the gross capillary water minus deep percolation and represented the intensity of capillarity. In the experiment, the largest net up-flux of groundwater was found in treatments with thin layers of OB barrier material (20OB−, 46.25 cm yr$^{-1}$ on average, and 20OB+, 42.81 cm yr$^{-1}$ on average); and strong capillary effects were also found in treatments without a barrier layer (NC+ and NC−), with only 20 cm of TS (20TS− and 20TS+), and with 50 cm of OB and simulated wet condition (50OB+) (Figure 3). The CIT analysis further indicated that barrier thickness was the most significant factor affecting the intensity of capillarity (as the root node in the CIT model in Figure 4). There was on average 39.21 cm yr$^{-1}$ of saline groundwater up-flux into the unsaturated zone when the barrier layer was less than 20 cm, regardless of the barrier material type and the amount of precipitation (irrigation). It was almost as much as the amount of water irrigated during the same period. When capped with 50 cm, saline groundwater up-flux was reduced to 20.14 cm yr$^{-1}$, and when the barrier layer increased to 100 cm it was further reduced to 5.57 cm yr$^{-1}$. Nevertheless, it should be noted that the capillary connections between cover layer and saline groundwater were not completely cut off even with as much as 100 cm of barrier material; especially when fine-textured OB was used, there was still significant upward capillary water movement (18.41 cm yr$^{-1}$ for 100OB− and 12.61 cm yr$^{-1}$ for 100OB+). Water balance treatments did not lead to statistically significant differences in capillary water up-flux, although the treatments with lower irrigation amounts generally consumed more saline water (Figure 3).

3.1.2. Treatment Effects on Soil Salinity

Cover soil is the main layer for root growth of the seedlings especially the following years after reclamation. Preventing the intrusion of salts from below is an important function of the barrier layer, and salts accumulated in this layer could also possibly get into the cover soil and eventually influence the success of reclamation. In the column experiment, salts accumulated quickly in some soil profiles (Figure 5). Salinization occurred in both cover soil (PMM) and the barrier layer (TS or OB) after three growing seasons (Figure 6). Barrier thicknesses showed strong influence on soil EC at all monitored depths, water balance didn’t show significant effects on soil salinity in most cases, while the effects of barrier material were more complicated (Figure 6 and Table 3).
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**Figure 3.** Saline groundwater up-flux during 3 growing seasons. Different letters indicate significant differences ($p < 0.05$, ANOVA) among treatments, which were derived by comparing all the values across the NC, OB and TS trials.

**Figure 4.** The CIT model for saline groundwater supply (Node 2 = 39.21 cm yr$^{-1}$, Node 4 = 20.14 cm yr$^{-1}$, and Node 5 = 5.57 cm yr$^{-1}$).
In the cover soils, salinization developed quickly after the initiation of the experiment when the cover soils were directly placed above the saline groundwater or the barrier layer was thin (20 cm). 50 cm of barrier layer had successfully prevented salt concentration increase in cover soil; however, when using OB, this thickness should be more than 50 cm to prevent salinization. Data from EC probes installed at 20 and 40 cm depths indicated that salt concentrations in the cover soils in these treatments all increased from less than 2 dS m\(^{-1}\) to 4 dS m\(^{-1}\) or more, from the beginning to the end of the experiment. These saline conditions could have stressed the plants [7,10,28]. In the barrier layers, OB tended to accumulate more salts than TS. Salinization in this layer could be largely mitigated when using 50 cm or more of TS as barrier material; however, the EC level still increased with 100 cm of OB as barrier material. When the barrier layer was not thick (≤50 cm), salt concentrations increased faster in OB than in TS at 60 and 90 cm depths, especially in its upper part (at the 60 cm depth, below the cover soil-barrier material interface, Figure 5). When the barrier thickness increased to 100 cm, although the upper part might not be strongly affected, the lower part still had a salinity problem when OB was used. Based on the experimental data, the minimum barrier thickness recommended to prevent the increase of salt concentration in the cover soil would be between 20 cm and 50 cm for TS and more than 50 cm for OB.

Figure 5. Temporal variations of soil EC\(_{sp}\) in the cover soils and barrier layers monitored by the probes.
Figure 6. Salinization in cover soils after 3 growing seasons. Different letters in a row indicate significant differences ($p < 0.05$, ANOVA) among treatments at the same depth, which were derived by comparing all the values across the NC, OB and TS trials.

The CIT analysis on the final TDS in the cover soil also confirmed that barrier thickness was the most significant factor affecting salt accumulation in the cover soils (as the root nodes) (Figure 7), which was consistent with the result of CIT analysis on cumulative saline groundwater up-flux. Strong correlations were found between soil salinity and the amount of saline water up-flux especially in the
upper part of the soil profiles. At the end of the experiment, the coefficients of determination between groundwater up-flux and soil EC$_{sp}$ at 40, 60, 90, and 140 cm depths were 0.74, 0.58, 0.51, and 0.38, respectively. It indicates that not only the soils close to saline groundwater but also the upper cover soils could be affected by upward water and salt movement when a shallow water table is present, and the salinization process in reclaimed soils could develop very quickly and have marked effects on cover soil salinity in just several growing seasons.

![Diagram](image)

**Figure 7.** The CIT model for average TDS in the cover soil (PMM) (average TDS are 0.078%, 0.037%, and 0.021% in Node 2, 4, and 5 branches, respectively).

### 3.1.3. Plant Response

Plant growth could be influenced by soil moisture, nutrient availability as well as salinity. In the experiment, plants showed apparent differences in both aboveground biomass and total root biomass after 3 growing seasons. Among the three treatment factors, barrier material was the most significant factor affecting plant growth (Figure 8 and Table 3). Treatments using OB as barrier material generally had higher aboveground biomass than those using TS, and treatments without barrier layer had the lowest biomass. Water balances also showed positive effects on plant growth, except in those treatments without a barrier layer. In a period of three growing seasons, the barrier thickness exhibited slight effects on plants if the barrier layer was not missing. High aboveground biomass was found in treatments with thin barrier layer (≤50 cm), especially in the OB treatments. The experiment results also clearly showed that roots preferred to grow in OB rather than in TS, and the growth was also facilitated by water condition (Figure 8). Therefore, high root biomass amounts were found in treatments with OB barrier material and high irrigation.

Within three growing seasons, plant response was not consistent with the soil water and salinity results. Soil columns using OB material or thin barrier layer still accumulated high biomass, although they tended to accumulate more salts. The possible reason is that salt concentration in the rooting zone didn’t increase high enough due to the limit of experimental time and it couldn’t offset the benefit brought by soil moisture (Table 3). Therefore, the treatments should be validated over a longer period. A numerical simulation was done to further evaluate whether the soils become salinized and “unsuitable” for plants over a longer period (15 years).
3.2. Modeling Salinity Evolution in Reclaimed Soils

3.2.1. Validation of the Soil Water and Salt Movement Model

Since soil hydraulic properties could change over time dramatically, especially in the first few years after reclamation [29], we used hourly experimental data more than one year after the column setup for the hydraulic and salt transport parameter inversion. The period covers days (12 September 2014–7 October 2014), and has two irrigation events during the period. Soil hydraulic and salt transport parameters used in the model are listed in Table 4.

Table 4. Hydraulic and solute transport parameters 1 for the reclamation materials used in the HYDRUS-1D model.

| Material 2 | BD 3 (cm³ cm⁻³) | Qr 4 (cm³ cm⁻³) | Qs 4 (cm³ cm⁻³) | α 5 | n 6 | Ks (cm hr⁻¹) | Disp (cm) | Diffw (cm² hr⁻¹) |
|------------|-----------------|-----------------|-----------------|-----|----|--------------|------------|--------------|
| PMM        | 0.89            | 0.18/0.25 4     | 0.66            | 0.44| 1.89| 5.44         | 10         | 0.01         |
| TS         | 1.37            | 0.08            | 0.48            | 0.071| 2.70| 4.00         | 50         | 0.01         |
| OB         | 1.65            | 0.17            | 0.30            | 0.022| 1.55| 0.10         | 10         | 0.01         |

1 BD, Qr, Qs, α, n and Ks are the parameters of bulk density, residual water content, saturated water content and saturated hydraulic conductivity in the van Genuchten functions; and Disp and Diffw are longitudinal dispersivity and molecular diffusion coefficient in free water, respectively. 2 TS, OB, and PMM are abbreviations for tailings sand, overburden, and peat mineral soil mix, respectively. 3 Calculated based on the dry soil weight packed in the columns. 4 0.18 cm³ cm⁻³ for profiles with only PMM material, and 0.25 cm³ cm⁻³ for profiles reclaimed with PMM and TS/OB.

The experimental data of the NC+ treatment between October 2013 and May 2015 recorded by GS3 probes were used for validating the parameters of PMM (single layer); data of the 50TS+ and 100OB+ treatments during the same period were used for validating the parameters of PMM and TS, or PMM and OB constructed in layers, respectively. The temporal changes of measured and simulated VWC and TDS in pore water in different treatments during the whole experiment are shown in Figure 9, and the validation results from 100th day after the start to the end are shown in Table 5. In the first
100 days, there were some poor agreements between the measured and simulated values because of the instability of the soil structure [29]. After that, most of the simulated values were in good agreement with the measured data. All simulated VWC values in different treatments had low ME ($-0.02, 0.03$), MAE ($0.01, 0.03$), and RMSE ($0.01, 0.04$), indicating that the model can be a promising tool for predicting water movement. The prediction of TDS in pore water had low ME, MAE and RMSE and was generally acceptable as well. However, the errors in treatments with TS as barrier material were relatively large, and this was probably due to the hydrophobic nature of this material [30].

Figure 9. Cont.
Figure 9. Comparison of the experimental soil moisture and salt concentration with the simulated data using parameters in Table 2 (NC+ treatment) (• average data of the treatment recorded by GS3 probes, —simulated data from HYDRUS-1D).

Table 5. Comparison of simulated and experimental soil moisture content and TDS in the soil profile.

| Soil Property | Depth (cm) | PMM | PMM + TS | PMM + OB |
|---------------|------------|-----|----------|----------|
| VWC 1 (cm³ cm⁻³) | 20        | 0.00 | 0.02 | 0.02 | -0.01 | 0.03 | 0.03 | -0.02 | 0.03 | 0.03 |
|               | 40        | 0.01 | 0.02 | 0.02 | 0.02 | 0.02 | 0.03 | 0.00 | 0.01 | 0.02 |
|               | 60        | -0.02 | 0.02 | 0.02 | 0.00 | 0.01 | 0.01 |
|               | 90        | 0.02 | 0.02 | 0.02 | 0.00 | 0.01 | 0.01 |
|               | 140       | 0.03 | 0.03 | 0.04 | -0.02 | 0.02 | 0.02 |
| TDS 2 (g L⁻¹) | 20        | -0.01 | 0.34 | 0.40 | 0.98 | 0.98 | 1.03 | -0.21 | 0.34 | 0.41 |
|               | 40        | -0.53 | 0.54 | 0.73 | 1.20 | 1.20 | 1.37 | -0.35 | 0.50 | 0.68 |
|               | 60        | 0.30 | 1.63 | 1.99 | -0.24 | 0.40 | 0.53 |
|               | 90        | -1.18 | 1.18 | 1.23 | -0.22 | 0.22 | 0.35 |
|               | 140       | -0.49 | 0.49 | 0.54 | -0.22 | 0.26 | 0.32 |

1 Volumetric water content. 2 Total dissolved solid in pore water.
3.2.2. Modeling of Long-Term Soil Salinity Evolution

Soil water and salt dynamics in a series of capping designs which used TS or OB as barrier material from 0 to more than 2 m thick were simulated under normal, wet and dry conditions. The final salt distribution in profiles and the cumulative water fluxes at lower boundary of the soil profiles were used to identify the reliability of the design.

When PMM and TS were used as reclamation materials, soil profiles using 50 cm of TS as a barrier layer would be seriously salinized after 15 years under the dry condition (green lines in Figures 10 and 11), especially in the lower part of the PMM and the lower part of TS. Under the normal precipitation condition, it is still under saline risk at the bottom of the PMM layer. Using this capping strategy, there would be continuous net upward water flux from the bottom under the dry condition, and periodic up-flux under the normal condition. This confirmed again that design with a thin barrier layer would risk having salts migrate into the rooting zone (Figure 11). When the barrier thickness was over 75 cm, the salinity level in the cover soil was always lower than the critical ECsp for plant growth (4 dS m−1). From this simulation, it can be summarized that the minimum barrier thickness to restrain salt intrusion into the rooting zone for the PMM and TS combination would be 50 cm and 75 cm.

When OB was used as a barrier material, soil capillarity was more intensive, and more salt accumulated over the 15-year simulation period. Layers with high salt concentration or EC values formed in the soil profile, and the thinner the barrier layer, the closer was the cover soil to the saline layer. When the thickness of the barrier layer (OB) was 100 cm, the salts would intrude into the cover soil, especially under dry condition. The continuous upward flux also indicated that this design would have potential risk under dry and normal conditions (Figure 11). When the barrier thickness was increased to 150 cm, the ECsp in the cover soil might not reach a problematic level. Therefore, the recommended minimum safe barrier thickness to restrain salt intrusion into the rooting zone for the PMM and OB combination would be 50 cm and 200 cm, which was more than double of the thickness of PMM and TS combination.

Figure 10. Cont.
Figure 10. Final salt distribution in profiles under different capping strategies after 15 years of simulation (blue line, normal condition; red line, wet condition; green line, dry condition).

Figure 11. Cont.
Figure 11. Cont.
with the OB barrier material than with the TS due to the greater capillarity of the OB material, although when a shallow water table is present. Further, the risk of salinization in the rooting zone was higher with the OB barrier material than with the TS due to the greater capillarity of the OB material, although the OB material facilitated plant growth within the first a few growing seasons after reclamation. If less than 20 cm of TS or less than 50 cm of OB was used as the barrier layer, salt concentration in the cover soil could increase dramatically under the simulated saline groundwater conditions and some weather conditions. A further conclusion drawn from the 15-year modeling work was that when 50 cm of PMM was used as a cover soil and the water table is at the bottom of the barrier layer, the minimum barrier thickness to prevent salt intrusion into the rooting zone in long term was 75 cm for TS and 200 cm for OB as a barrier material.

4. Conclusions

After a comparison of the effects of different capping strategies and different hydrologic conditions on salt movement in oil sands reclamation soils, we conclude that barrier thickness was the most important factor that should be considered to minimize salt migration from below into the rooting zone when a shallow water table is present. Further, the risk of salinization in the rooting zone was higher with the OB barrier material than with the TS due to the greater capillarity of the OB material, although the OB material facilitated plant growth within the first a few growing seasons after reclamation. If less than 20 cm of TS or less than 50 cm of OB was used as the barrier layer, salt concentration in the cover soil could increase dramatically under the simulated saline groundwater conditions and some weather conditions. A further conclusion drawn from the 15-year modeling work was that when 50 cm of PMM was used as a cover soil and the water table is at the bottom of the barrier layer, the minimum barrier thickness to prevent salt intrusion into the rooting zone in long term was 75 cm for TS and 200 cm for OB as a barrier material.

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References
1. Alberta. Government of. Oil Sands Mine Reclamation and Disturbance Tracking by Year, 2017-12-31 ed.; Government of Alberta: Edmonton, AB, Canada, 2019.
2. Leskiw, L.A. Land Capability Classification for Forest Ecosystems in the Oil sands Volume 1-Field Manual; Cumulative Environmental Management Association, Ed.; Cumulative Environmental Management Association: Fort McMurray, AB, Canada, 2003.
3. Barbour, L.; Macyk, T. Soil Capping Research in the Athabasca Oil Sands Region; Syncrude Canada Limited: Fort McMurray, AB, Canada, 2010.

Figure 11. Cumulative water fluxes at the lower boundary of soil profiles under different capping strategies and climate conditions (blue line, normal condition; red line, wet condition; green line, dry condition. The line above zero indicates a net groundwater up-flux, and the line below zero indicates a net discharge of soil water to groundwater).
4. Kessel, E.; Ketcheson, S.; Price, J. The distribution and migration of sodium from a reclaimed upland to a constructed fen peatland in a post-mined oil sands landscape. *Sci. Total Environ.* **2018**, *630*, 1553–1564. [CrossRef] [PubMed]

5. Kessler, S.; Barbour, S.L.; van Rees, K.C.J.; Dobchuk, B.S. Salinization of soil over saline-sodic overburden from the oil sands in Alberta. *Can. J. Soil Sci.* **2010**, *90*, 637–647. [CrossRef]

6. Committee, Alberta Soils Advisory. *Soil Quality Criteria Relative to Disturbance and Reclamation*; Soils Branch, Alberta Agriculture: Greensboro, NC, USA, 1987.

7. Howat, D. Acceptable Salinity, Sodicity and pH Values for boreal Forest Reclamation; Environmental Sciences Division: Edmonton, AB, Canada, 2000.

8. Biagi, K.M.; Oswald, C.J.; Nicholls, E.M.; Carey, S.K. Increases in salinity following a shift in hydrologic regime in a constructed wetland watershed in a post-mining oil sands landscape. *Sci. Total Environ.* **2019**, *653*, 1445–1457. [CrossRef] [PubMed]

9. Purdy, B.G.; Ellen Macdonald, S.; Lieffers, V.J. Naturally saline boreal communities as models for reclamation of saline oil sand tailings. Restor. Ecol. **2005**, *13*, 667–677. [CrossRef]

10. Renault, S.; Zwiazek, J.; Fung, M.; Tuttle, S. Effects of oil sand tailings on plant species of the boreal forest. *Environ. Pollut.* **2000**, *107*, 357–365. [CrossRef]

11. Leatherdale, J.; Chanasyk, D.; Quideau, S. Soil water regimes of reclaimed upland slopes in the oil sands region of Alberta. *Can. J. Soil Sci.* **2012**, *92*, 117–129. [CrossRef]

12. Olatuyi, S.O.; Leskiw, L.A. Long-term changes in soil salinity as influenced by subsoil thickness in a reclaimed coal mine in east-central Alberta. *Can. J. Soil Sci.* **2014**, *94*, 605–620. [CrossRef]

13. Purdy, B.G.; Ellen Macdonald, S.; Lieffers, V.J. Naturally saline boreal communities as models for reclamation of saline oil sand tailings. Restor. Ecol. **2005**, *13*, 667–677. [CrossRef]

14. Spennato, H.M.; Ketcheson, S.J.; Mendoza, C.A.; Carey, S.K. Water table dynamics in a constructed wetland, Fort McMurray, Alberta. *Hydrol. Process.* **2018**, *32*, 3824–3836. [CrossRef]

15. Volik, O.; Petrone, R.M.; Hall, R.I.; Macrae, M.L.; Wells, C.M.; Elmes, M.C.; Price, J.S. Long-term precipitation-driven salinity change in a saline, peat-forming wetland in the Athabasca Oil Sands Region, Canada: A diatom-based paleolimnological study. *J. Paleolimnol.* **2017**, *58*, 1–18. [CrossRef]

16. Ketcheson, S. Hydrology of A Constructed Fen Watershed in A Post-Mined Landscape in the Athabasca Oil Sands Region; University of Waterloo: Waterloo, ON, Canada, 2016.

17. Leatherdale, J.; Chanasyk, D.; Quideau, S. Soil water regimes of reclaimed upland slopes in the oil sands region of Alberta. *Can. J. Soil Sci.* **2012**, *92*, 117–129. [CrossRef]

18. Hothorn, T.; Hornik, K.; Zeileis, A. Unbiased recursive partitioning: A conditional inference framework. *J. Comput. Graph. Stat.* **2006**, *15*, 651–674. [CrossRef]

19. Šimůnek, J.; Sejna, M.; Saito, H.; Sakai, M.; van Genuchten, M.T. *The Hydrus-1d Software Package for Simulating the Movement of Water, Heat, and Multiple Solutes in Variably Saturated Media*; Version 4.17; Department of Environmental Sciences, University of California Riverside: Riverside, CA, USA, 2013.

20. Boese, C.D. *The Design and Installation of a Field Instrumentation Program for the Evaluation of Soil-Atmosphere Water Fluxes in a Vegetated Cover over Saline/Sodic Shale Overburden*; University of Saskatchewan: Saskatoon, SK, Canada, 2003.

21. Anderson, M.P.; Woessner, W.W. *Applied Groundwater Modeling*; Academic Press: San Diego, CA, USA, 2002; pp. 343–372.

22. Dobchuk, B.S.; Shurniak, R.E.; Barbour, S.L.; O’Kane, M.A.; Song, Q. Long-term monitoring and modelling of a reclaimed watershed cover on oil sands tailings. *Int. J. Min. Reclam. Environ.* **2013**, *27*, 180–201. [CrossRef]
28. Naeth, M.; Chanasyk, D.; Burgers, T. Vegetation and soil water interactions on a tailings sand storage facility in the Athabasca oil sands region of Alberta Canada. *Phys. Chem. Earth Parts A/B/C* **2011**, *36*, 19–30. [CrossRef]

29. Meiers, G.P.; Barbour, S.L.; Qualizza, C.V.; Dobchuk, B.S. Evolution of the hydraulic conductivity of reclamation covers over sodic/saline mining overburden. *J. Geotech. Geoenviron.* **2011**, *137*, 968–976. [CrossRef]

30. Diamantopoulos, E.; Durner, W.; Reszkowska, A.; Bachmann, J. Effect of soil water repellency on soil hydraulic properties estimated under dynamic conditions. *J. Hydrol.* **2013**, *486*, 175–186. [CrossRef]

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