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Reactive Transport Model of the Carbonate-Evaporite Elk Point Group Underlying the Athabasca Oil Sands

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ABSTRACT

A reactive transport model was constructed to investigative how the carbonate-evaporite aquifer system within the Devonian Elk Point Group in northeast Alberta has evolved over geologic time and to evaluate connectivity in the present system. The mineable area of the Athabasca Oil Sands overlies the Elk Point Group where regional dissolution of halite and anhydrite has eroded the Prairie Evaporite Formation from a thickness of approximately 260 m to as thin as 20 m. Groundwater chemistry changes along the flowpath towards the Athabasca River, reflecting the evaporite distribution. Water types change from bicarbonate- to sulphate- to chloride-types, and salinities increase exponentially toward saturation with respect to halite. The model was constructed as a 1-D domain in PHREEQC to simulate the regional dissolution of the evaporites, using hydraulic testing results, isotopic data, and the measured hydrochemistry to constrain the transport parameters. The dual porosity function in PHREEQC was needed to calibrate the model to the measured chloride distribution over the 90 km-long model domain, indicating the role of mass transfer from poorly-connected regions of the aquifer system. The simulated duration of the regional dissolution was on the order of 50,000 to 500,000 years, suggesting that the phases of dissolution postulated from geologic evidence from the pre-Cretaceous to the Holocene (Stoakes et al. 2014a) may have consisted of relatively shorter pulses of dissolution. Two types of accelerated anhydrite dissolution and dedolomitization are identified for the current conditions: anhydrite/gypsum removal by low-TDS, bicarbonate-dominated waters and increased anhydrite/gypsum solubility with higher salinity. Both of these processes appear to be occurring within approximately 20 km east of the Athabasca River. Reactive transport modelling of the regional system may help to identify local areas of karst development and vertical connection to open-pit mines in the overlying McMurray Formation.

Keywords: Reactive transport carbonate evaporite karst

INTRODUCTION

The mineable Athabasca Oil Sands Area (AOSA; Figure 1) is underlain by a carbonate-evaporite aquifer system with hydraulic heads >50 m above the planned base of the open-pit mines. A karst-type inflow of saline water to one of the mines in 2010 demonstrated the high degree of vertical connectivity to this aquifer system where hydraulic windows exist (Wozniewicz et al. 2014). Characterization of the Devonian strata and specifically the aquifer system within the Elk Point Group in the region has increased significantly since the 2010 karst inflow event.

This study presents a reactive transport model that provides an investigative tool to aid in attempts to identify regions with highly-transmissive karst and potential vertical connectivity between the Elk Point Group and overlying McMurray Formation. The model was also used to explore the evolution of the Elk Point aquifer system over geologic time and to postulate a mechanism for the present-day hydrochemical distribution. Data from previously published studies were used to calibrate the model, as the geochemical interpretation from recent field programs at several operating mines is ongoing. Dedolomitization is assessed at a regional scale, as it has been recognized as a key process resulting in enhanced permeability laterally within the Elk Point aquifer system and potentially also enhancing vertical connectivity (Wozniewicz et al. 2014).

BACKGROUND

Geologic Setting

The mineable AOSA is near the northeast edge of the Western Canadian Sedimentary Basin (Figure 1), where the Phanerozoic strata form a wedge over the Canadian Shield with maximum thickness of approximately 700 m. Devonian strata consisting primarily of marine carbonates regionally dip to the southwest (Figure 2) due to subsidence associated with formation of the Rocky Mountains. Cretaceous sediments including the McMurray Formation, which hosts the oil sands deposit, overlie a 200 million year gap in the geologic record referred to as the Pre-Cretaceous unconformity.

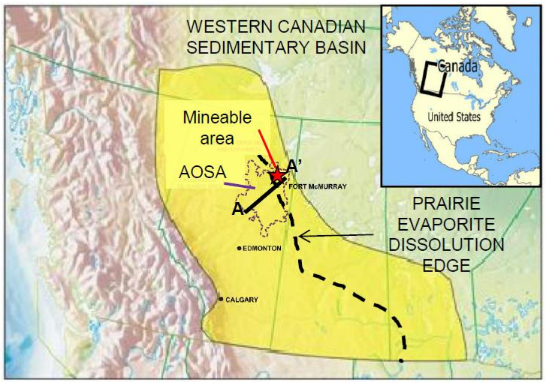

Figure 1 The mineable Athabasca Oil Sands Area

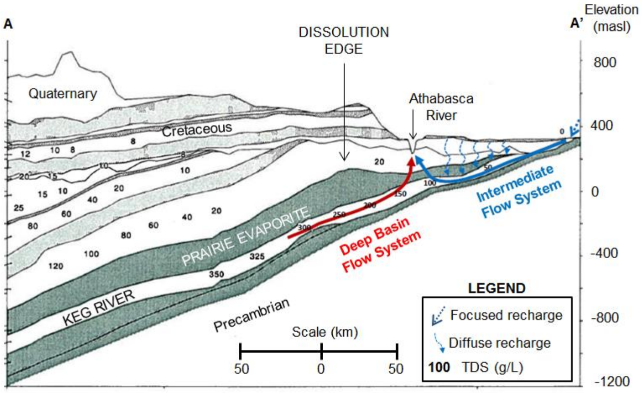

Figure 2 Cross section through Northeast Alberta

The Devonian Elk Point Group includes porous reef buildups of the Keg River Formation and an evaporative sequence in the Prairie Evaporite Formation, which on-laps the reefs. The Prairie Evaporite Formation was deposited as brining-upward sequences of dololaminites, anhydrite, and halite (Stoakes et al. 2014b). These two formations are overlain by approximately 60 m of primarily marine carbonates and shales, which are referred to here as the Upper Devonian.

Infiltrating meteoric waters to the eastern edge of the basin have dissolved the Prairie Evaporite Formation from an initial thickness of approximately 260 m west of the dissolution edge (Figure 1) to as thin as 20 m near the basin edge. This dissolution resulted in extensive collapse of the overlying strata and enhanced permeability within the Prairie Evaporite Formation.

Physical Hydrogeology

Dololaminites in the lower Prairie Evaporite Formation form a highly transmissive (10^{-3} to 10^{0} m^2/s) aquifer where they have been more extensively dedolomitized, as evident in regional responses to the 2010 inflow event (Wozniewicz et al. 2014). This event provided direct evidence of karst in the Devonian strata and of good connection in places between the Elk Point and the overlying McMurray Formation, neither of which had previously been unequivocally accepted (e.g., Alsands 1981). Reefs in the Keg River Formation form aquifers capable of sustained wastewater injection (1,733,000 m^3 over ten years; Husky 2011). Most multi-day hydraulic tests in the region have been conducted over both of these formations and have yielded transmissivities on the order of 10^{-5} m^2/s. The Prairie Evaporite Formation on-laps to the sides of reefs suggesting some degree of connection; hence, the two aquifers are collectively referred to as the Elk Point aquifer system.

The Elk Point aquifer system is highly heterogeneous, and flow paths at the sub-lease scale are thought to follow complex fracture networks with a wide range of connectivity. At the regional scale, the subcrop of the Elk Point Group beneath Quaternary sediments near the provincial border (Figure 3) provides focused recharge to the aquifer system. Groundwater flows from the subcrop generally westward toward the discharge zone at the Athabasca River, and the system also receives more diffuse vertical recharge through the overlying units (Figure 2).

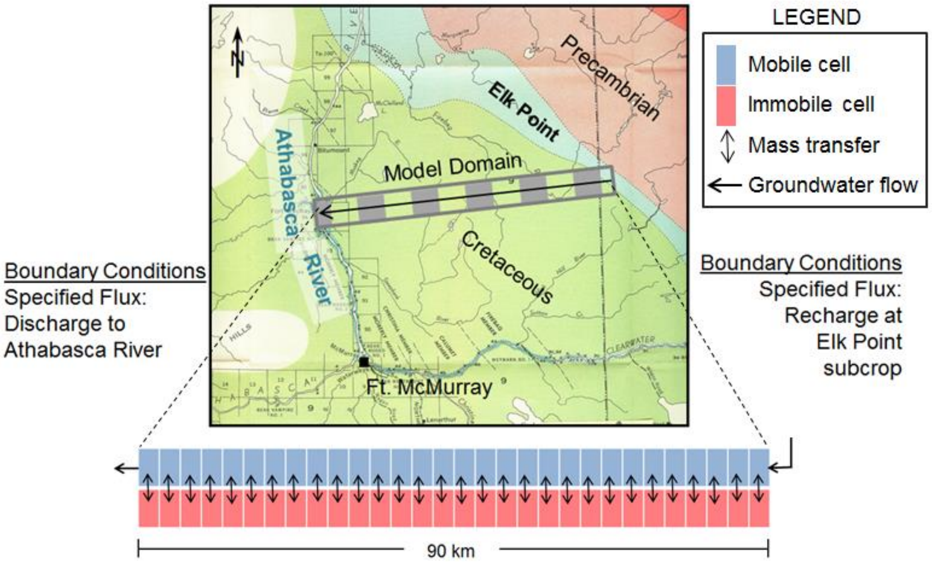

Figure 3 Geologic map: units subcropping beneath Quaternary sediments (Norris 1963), adapted with regional flow directions in the Elk Point and the 1-D domain for the reactive transport model.

GEOCHEMISTRY OF THE ELK POINT AQUIFER SYSTEM

The regional distribution of evaporite minerals in the Prairie Evaporite Formation (Figure 4) provides a primary control on the hydrochemistry within the Elk Point aquifer system. Generally in the region, halite is still present and dissolving between the dissolution edge and the Athabasca River. Within approximately 20 km to 30 km east of the Athabasca River halite has been removed and anhydrite is dissolving, while in the eastern part of the region both halite and anhydrite have been removed leaving an extensively collapsed region with dolomite the primary mineral (Figure 4; Schneider and Grobe 2013).

A preliminary model was constructed in PHREEQC (v. 3.06; Parkhust and Appelo 2013) to begin investigating the relationship between dissolution of the Prairie Evaporite over geologic time and the present day hydrochemistry. A batch model domain of one litre was filled with an approximate composition of Prairie Evaporite Formation prior to dissolution, based on publically-available geologic logs: 40% dolomite ($CaMg(CO_3)_2$), 20% halite (NaCl), 20% anhydrite ($CaSO_4$), 20% calcite ($CaCO_3$), and 10% porosity. The model domain was then successively flushed with pure water equilibrated with atmospheric CO_2 (pCO_2 10^{-2} atmospheres [atm]) and allowed to equilibrate to these minerals and CO_2 at 10°C at each step. The results of this preliminary modelling exercise are shown in Figure 5.

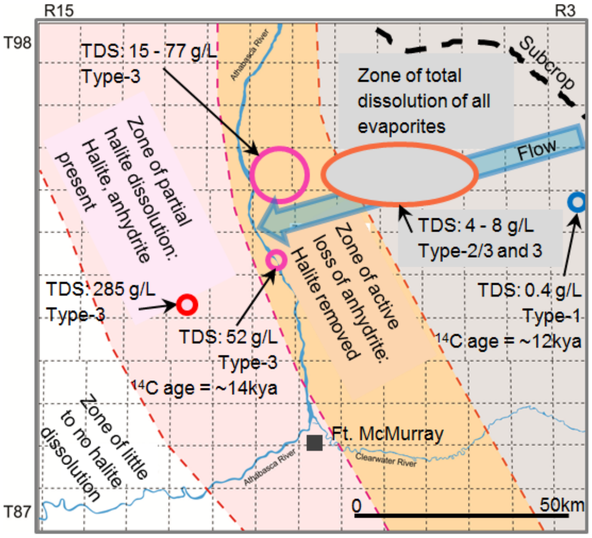

Figure 4 Prairie Evaporite dissolution map (adapted from Schneider and Grobe 2013); samples from Elk Point wells and saline a saline spring (1 to 9 wells per oval).

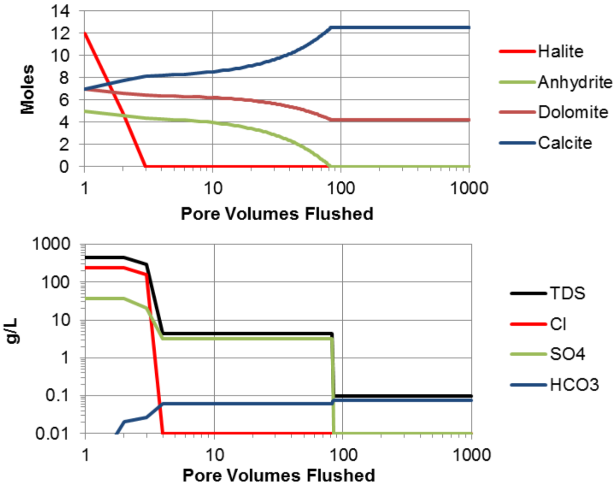

Figure 5 Results from a preliminary modelling exercise: successive flushing with fresh waters of a one litre domain representing the Prairie Evaporite Formation.

Three distinct zones were simulated with the preliminary model, similar to the regional dissolution map (Figure 4). The simulation illustrates that halite in the Elk Point Group was dissolved after only a few pore volumes had been flushed by infiltrating fresh recharge waters, such that regions with halite remaining and salinity of >300 g/L TDS can be assumed to have had little contact with fresh waters. An intermediate region was simulated having anhydrite remaining but halite removed and waters dominated by sulphate and salinity of approximately 5 g/L TDS (Figure 5). Where on the order of one hundred pore volumes or more had been flushed in the preliminary model, the anhydrite was removed leaving only dolomite, calcite, and bicarbonate-dominated waters with salinity on the order of 0.1 g/L.

Water types and salinities in samples collected from the Elk Point aquifer system in the region are shown on Figure 4 (Alsands 1981, ARC 1975, Cenovus 2011, Golder 2002, Gue 2012, and Husky 2011). Water type classifications developed here are listed in Table 1. The samples exhibit an evolution along the east to west flowpath that reflects the mineralogy and is generally similar to the results from the preliminary modelling exercise. Near the focused recharge zone at the Elk Point subcrop, a sample had salinity of 0.4 g/L TDS and was dominated by bicarbonate. Approximately 45 km to 60 km downgradient of the subcrop, salinities range from 6 g/L to 8 g/L TDS with dominant anions of sulphate and chloride. Within 15 km of the Athabasca River, salinities in the Elk Point increase to 77 g/L TDS with chloride the dominant anion (Figure 4). West of the river a chloride-dominated sample with salinity of 285 g/L TDS has been collected, and similar samples have been collected from wells farther to the west.

Table 1 Water types in the Elk Point aquifer system, northeast Alberta

Water Type	Dominant Anion	Origin of Water	Salinity	Residence (m=million; k=thousand)	Occurrence
Type 1	Bicarbonate	Meteoric	< 2 g/L	< 10k yrs	Near subcrop
Type 2	Sulphate	Meteoric	3 to 5 g/L	< 10k yrs	~20 to ~50 km east of Athabasca R.
Type 3	Chloride	Meteoric	5 to 300 g/L	5k to 40k yrs	East of dissolution edge
		Connate	> 250 g/L	> 1m yrs (?)	West of dissolution edge
Intermediates	HCO_3, SO_4, Cl	Meteoric	1 to 10 g/L	< 40k yrs	East of Athabasca R.

The main geochemical processes, based on the mineralogy, hydrochemistry, and preliminary modelling, are: halite (NaCl) dissolution, anhydrite ($CaSO_4$) dissolution, dolomite ($CaMg(CO_3)_2$) dissolution, and calcite ($CaCO_3$) dissolution/precipitation. Ion exchange in the Elk Point in the region is thought to be less significant than the main mineral interactions listed above, but is significant enough to characterize the samples east of the dissolution edge as having meteoric origin, using the Index of Bases Exchange (IBE) formulation of Collins (1975). Samples west of the dissolution edge have IBE greater than the diagnostic cut-off for connate water defined by Collins (1975) of 0.129, indicating deep basin water with residence time of millions of years or more.

Isotopic signatures further characterize origins of waters in the Elk Point. Deep basin waters sampled in the Edmonton area have $\delta^{18}O$ and $\delta^{2}H$ signatures of warm recharge and a high degree of water-rock interaction (Simpson 1999). Samples from Elk Point wells and saline springs east of the dissolution edge in the region have meteoric $\delta^{18}O$ and $\delta^{2}H$ signatures of cold water recharge, with $\delta^{18}O$ values ranging from -19.2‰ to -24.6‰ (Cenovus 2011, Gue 2012, Wallick and Dabrowski 1982). These values and ^{14}C groundwater age estimates of approximately 10,000 to 30,000 years indicate that Elk Point waters east of the dissolution edge represent a mixture of meltwater from the Laurentide ice sheet, which melted in the region approximately 10,000 years ago and has a $\delta^{18}O$ signature of -25‰ to -28‰, and warmer water similar to present-day recharge (-17.7‰ to -19‰) (Grasby and Chen 2005; Gue 2012).

Karst development in the region is caused by halite dissolution, anhydrite/gypsum dissolution, and dedolomitization. These processes accelerate carbonate dissolution, which is otherwise controlled by pH of the water and specifically carbonic acid sourced from atmospheric CO_2. Dedolomitization causes a large increase in the dolomite dissolution rate (e.g., Palmer 2007; Escoria et al. 2013) due to dissolution of both dolomite and anhydrite. It causes a process referred to as the common-ion effect, with ideal stoichiometry (Appelo and Postma 2005):

$$1.8CaSO_4 + 0.8CaMg(CO_3)_2 \rightarrow 1.6CaCO_3 + Ca^{2+} + 0.8Mg^{2+} + 1.8SO_4^{2-}.$$

Anhydrite and dolomite dissolution both produce calcium ions, driving precipitation of calcite.

1-D REACTIVE TRANSPORT MODEL

A 1-D reactive transport model was constructed in PHREEQC to attempt to simulate salinities above 5 g/L TDS where halite has been completely or almost completely removed, and to provide an investigative tool to potentially identify areas with higher dissolution rates. The model domain represents a 90 km flow path from the Elk Point subcrop to the Athabasca River, using 30 mobile and 30 immobile cells (Figure 3).

The initial conditions represent the Elk Point prior to dissolution, based on publically-available logs from the region: 40% halite, 40% dolomite, 10% anhydrite, and 10% calcite; 6% porosity; and 1 mol/L dissolved organic carbon (oil and bitumen). Boundary conditions are specified flux into the east end of the domain yielding a groundwater velocity of 10 m/yr through the model domain, and an equivalent discharge rate at the west end. Recharge was simulated as pure water equilibrated to atmospheric CO_2 with pCO_2 of 10^{-2} atm. No transverse flow boundaries are necessitated by the 1-D model geometry: an important limitation, which assumes zero vertical recharge through the overlying Cretaceous sediments and Upper Devonian into the Elk Point aquifer system. This limitation is discussed further below.

A high degree of simulated heterogeneity was required to approximately match the hydrochemistry in samples along the flow path. Dispersion was simulated using a dispersivity of 1000 m for the regional flowpath. The dual-porosity function in PHREEQC was also required: the domain was divided into 30 cells through which the simulated groundwater flows laterally, referred to as mobile cells, and 30 adjacent cells referred to as immobile cells (Figure 3). Rate-limited mass transfer between immobile and mobile cells is allowed, simulating a store-and-release function in the poorly-connected regions of the domain similar to the concept of Harvey and Gorelick (2000). The mobile and immobile cells initially have very high salinity due to equilibration with halite; as the mobile cells are flushed with meteoric recharge, Na and Cl in particular diffuse from the immobile cells into the mobile cells.

The simulated flow velocity of 10 m/yr was chosen based on corrected radiocarbon ages (Figure 4; Cenovus 2011, Gue 2012) and 1-D Darcy flux using hydraulic conductivity from multiple-day hydraulic tests (Alsands 1981, Golder 2002, Husky 2011), average hydraulic gradient of 0.002, and porosity of 10%. Both approaches yielded flow velocity estimates ranging from 1 to 10 m/yr. Flow velocities may have been lower at early stages of the dissolution of the Prairie Evaporite Formation. During the later glacially-driven stages of dissolution, flow velocities could have been an order of magnitude higher or more due to an increased pressure gradient imposed by the receding glacier.

Reactive transport is simulated in PHREEQC by solving equilibrium equations and shifting reaction products from each cell to the next cell along the flowpath, as calculated according to the flow velocity. At each time step in the model (representing approximately 300 yr), the water in each cell (3,000 m length) equilibrates with aqueous and mineral phases using the ThermoChimie database in PHREEQC, which utilizes the specific ion interaction theory (SIT) approach. The SIT approach estimates ion activities based on ionic strength and is hence suitable for solutions with high salinity; the Pitzer approach was not used due to its limited database in PHREEQC. Flow and transport of the reaction products are then simulated by input of the recharge to the eastern-most mobile cell and shifting the water in each mobile cell to the next mobile cell, followed by dispersion, rate-limited mass transfer from the immobile cells, and re-equilibration.

MODEL RESULTS

Dissolution Fronts and Simulation of Present Conditions

A halite dissolution front was simulated advancing from east to west over geologic time, followed by an anhydrite dissolution front (Figure 6). Dissolution fronts were delayed in the immobile model cells, which represent poorly-connected regions in or adjacent to the Elk Point aquifer system.

Figure 7 shows the simulated distribution of salinity and major ions along the flowpath for the present conditions, compared to concentrations in samples (Alsands 1981, ARC 1975, Cenovus 2011, Golder 2002, Gue 2012, and Husky 2011). Present conditions were simulated by 50,000 years of active freshwater recharge. At that time in the simulation, halite was completely removed from the mobile cells and partially remained in immobile cells within 10 km of the Athabasca River. The dissolution front for anhydrite, modelled as gypsum, is simulated to have advanced approximately 15 km (Figure 6), with anhydrite still present in the rest of the model domain.

The modelled concentrations of major ions and TDS in the Elk Point aquifer system (solid lines in Figure 7) approximately match the majority of the concentrations from samples, where available along the regional flowpath. The modelled distribution of water types generally matches those measured in samples: Type-1 in the east, Type-2/3 in the central part of the region, and Type-3 in the western part of the flowpath near the Athabasca River.

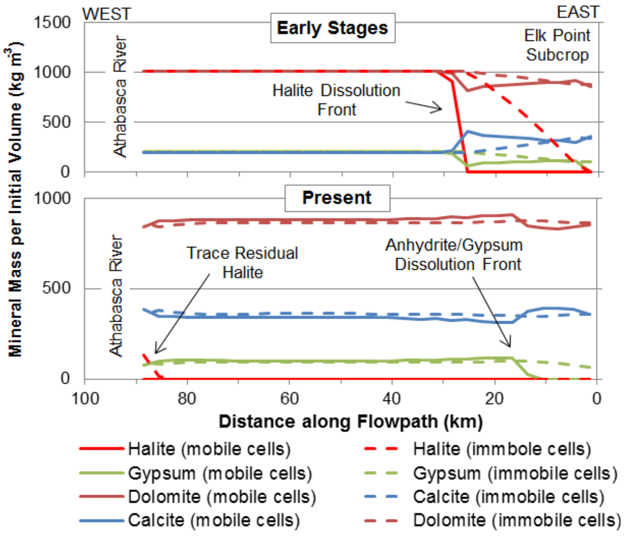

Figure 6 Model results: mineral mass and dissolution fronts along the flowpath, at early stages in the evolution and for the simulated present conditions.

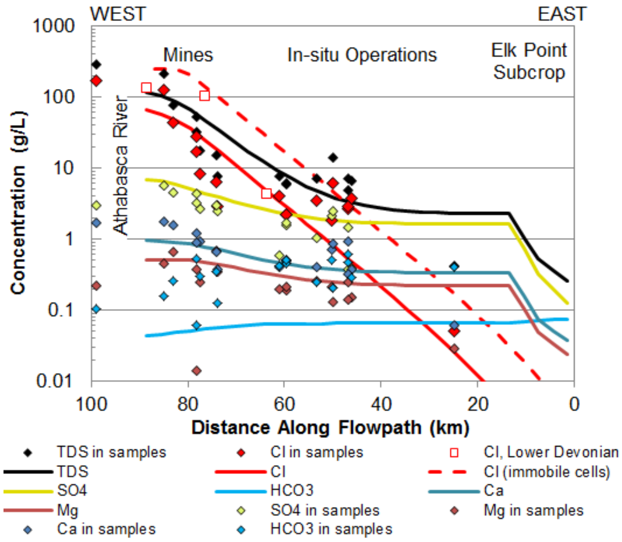

Figure 7 Model results for present conditions, as the 50,000 year simulation: major ions modelled (lines) and in Elk Point (◈) and Lower Devonian (□) samples.

Halite dissolution resulted in removal of more than half of the mass in the model. This large loss in volume was accommodated by regional collapse (not modelled). Anhydrite dissolution and dedolomitization resulted in increases in porosity of <1% to approximately 5%, based on the reactive transport model results.

Areas with Higher Degree of Connectivity

Rates of dissolution of halite, anhydrite, and dolomite were examined with the reactive transport model. Simulated rates of dissolution were highest as the halite dissolution front moved through the region: on the order of 1,000 g m^{-3} yr^{-1} for halite and in excess of 100 g m^{-3} yr^{-1} for anhydrite and dolomite. As the evolution of flow velocities over geologic time is unknown, modelled dissolution rates are presented for comparison only. The flow rate in the model assumes the halite dissolution products are advanced forward. In the actual system, the halite forms an aquiclude that was being dissolved entirely at the eastern edge and would likely have forced the dissolution products upward, possibly along collapse features at the dissolution front, further enhancing these vertical connections.

Dissolution rates for anhydrite and dolomite at the anhydrite dissolution front (Figure 6) were on the order of 10 g m^{-3} yr^{-1}. Dissolution rates were lower over much of the model domain, except within 20 km of the Athabasca River where they ranged from 0.5 g m^{-3} yr^{-1} to 10 g m^{-3} yr^{-1}. Simulated dissolution rates indicate two types of enhanced anhydrite/gypsum dissolution and dedolomitization: input of Type-1 water (~15 km on Figure 6) and where gypsum solubility is higher due to increased salinity (elevated SO_4 at ~70 to 90 km on Figure 7).

A minority of samples in the area of the mines have Cl concentrations an order of magnitude lower than the regional trend simulated with the model (Figure 6). These low Cl concentrations, which correspond with lower SO_4 and higher HCO_3, indicate greater vertical recharge in those areas. Input of vertical recharge in the eastern area of the mines could provide a driver for circulation of freshwater to development of karst. Mine operators should carry out geochemical modelling at the scale of the lease to further investigate higher dissolution rates.

Model Sensitivity

Model sensitivity was investigated for these parameters:

- Groundwater velocity;
- Mass transfer rate;
- Initial mineral composition; and
- Initial and boundary pH.

Simulations were run with flow velocity one order of magnitude lower and higher than the base case of 10 m/yr by varying the number of shifts as an analogue to flow rate. The higher velocity (100 m/yr), possible during glacial melt, corresponded with a reasonable match to the major ions in samples at a duration of 3,000 years, with dissolution fronts travelling an order of magnitude faster, and dissolution rates approximately 30% faster. Isotopic values and geologic evidence, however, necessitate a longer duration such that a high velocity could have occurred only for a relatively short time. With the lower velocity (1 m/yr), the match to major ions in samples was poor except when a reduced mass transfer rate was used. Dissolution fronts moved an order of magnitude slower, but peak dissolution rates were approximately the same as for the base case. While this slower velocity may have prevailed at earlier phases of the geologic evolution, an average velocity at this rate would necessitate a high proportion of vertical recharge to result in detection of modern carbon. Such a case may result in decreasing groundwater ages along the flowpath, which is not measured in samples.

The distribution of sodium and chloride, in particular, were sensitive to the mass transfer rate. A change of less than half an order of magnitude from the base case 5×10^{-13} mol s^{-1} yielded poor matches. The model results were not sensitive to the initial halite, dolomite, or calcite amounts (within +/- 50% of base case), but the timing of the anhydrite/gypsum dissolution front was sensitive to the initial anhydrite amount. Although carbonate reactions can be highly sensitive to pH, changing the recharge boundary pH or the initial conditions from pH 7 to either 6 or 8 yielded essentially no change in the results. The model also was not sensitive to the initial pCO2 value, and was not equilibrated with CO_2 (although the preliminary model was).

The simulated duration of active groundwater flow through the Elk Point and movement of dissolution fronts are sensitive to the selection of present-day groundwater velocity, and hence represents a lower estimate on the duration and an upper bound on dissolution front velocity, each with uncertainty of approximately an order of magnitude. Dissolution rates at the regional scale were less sensitive to the flow velocity. The model was not sensitive to whether halite was remaining in the poorly-connected model cells or completely removed: elevated chloride can be explained by either diffusion or active interaction with residual halite.

CONCLUSIONS

Karst inflow from the carbonate-evaporite Elk Point aquifer system to the overlying mines of the Athabasca Oil Sands Area is a major geohazard for the operators in the region. The following conclusions from investigative geochemical modelling with a 1-D reactive transport model are highlighted:

- The hydrochemistry within the Elk Point is directly related to mineral dissolution.
- Poorly-connected regions provide an important store-and-release function affecting the regional distribution of salinity in the Elk Point.
- Removal of anhydrite by Type-1 (TDS <1 g/L, HCO_3-dominated) water is associated with accelerated dedolomitization occurring in the eastern part of the region and possibly in areas of increased vertical recharge.
- Anhydrite dissolution and dedolomitization are also accelerated within approximately 20 km of the Athabasca River where salinity is greater than approximately 10 g/L, yielding increased anhydrite and gypsum solubility.
- The modelled dissolution rates suggest that dissolution of the Prairie Evaporite appears to have occurred as shorter periods of active dissolution (total duration 50,000 to 500,000 years) within phases of karst development in the Jurassic, Cretaceous, Tertiary, and Quaternary (Stoakes et al. 2014a).

The modelling approach used in this regional study has several limitations: the 1-D domain assumes zero or constant vertical recharge such that focused vertical recharge was only identified by mismatch to the model; mineral dissolution rates may not be applicable to shorter time scales, as kinetics were not included; an aggregate flow system was simulated rather than realistic geometries of flowpaths; and geologic information at the lease-scale to sub-lease scale were not included. This modelling approach is used as an investigative tool to improve the understanding of the evolution of the aquifer system and to aid in identification of areas of higher connectivity, rather than comprehensively modelling the system.

ACKNOWLEDGEMENTS

This work was initiated with direction from Hugh Abercrombie. Comments from Valerie Bertrand and an anonymous reviewer are greatly appreciated.

NOMENCLATURE

Cl = chloride

HCO_3 = bicarbonate

SO_4 = sulphate

TDS = total dissolved solids

$\delta^{18}O$ = isotopic ratio for oxygen-18 to oxygen-16

$\delta^{2}H$ = isotopic ratio for hydrogen-2 to hydrogen-1

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