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Assessing relationships between methane dynamics and chemical mass transport across the tailings-water interface of an oil sands end pit lake

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Summary

Mass loading from fluid fine tailings (FFT) into the overlying water cap of oil sands end pit lakes (EPLs) can affect water quality. Research focused on Syncrude's Base Mine Lake (BML) shows that advective-dispersive transport across the tailings-water interface(TWI) is a major source of dissolved constituents within the water cap. Initial numerical modelling suggest that enhanced mixing was occuring within the FFT near the TWI. Methanogenic degradation of diluent hydrocarbons added during bitumen extraction does result in methane (CH4) production and concomitant exsolution and ebullition of CH4 within the FFT. We propose that this CH_4 ebullition produces enhanced diffusion/dispersion within the FFT below the TWI altering geochemical gradients and altering seasonal mass loading to the overlying lake. In this study, we develop a modelling framework to explore relationships between CH_4 dynamics and chemical mass transport across this interface. First, we examine CH_4 dynamics within BML to assess potential for exsolution and ebullition of this biogenic gas. Second, we develop a numerical transport model that incorporates enhanced mixing and resulting chemical mass transport due to ebullition. Our study improves understanding of relationships between CH_4 dynamics and chemical mass transport across the tailings water interface, which in turn has implications for forecasting the long-term chemical evolution of the BML water cap.

Background

End pit lakes are a key reclamation strategy for oil sands mine closure (Kabwe et al., 2017). Approximately 30 EPLs are currently planned for the Athabasca oil sand region (AOSR), eight of which are to contain tailings under a shallow water cap (Kabwe et al., 2017). The initial cap water composition is dominated by oil sands process-affected water (OSPW), which contains elevated concentrations of dissolved salts, naphthenic acids, and hydrocarbons (Allen, 2008; Li, 2017; White and Liber, 2018). The long-term success of EPLs ultimately depends upon sustained cap water quality improvements resulting from *in situ* biogeochemcial processes and fresh water inputs. The geochemical gradients and rates of seasonal and annual mass loading from the tailings to the overlying water column will influence the rate and timing of water quality evolution within the EPLs (Alberta Energy Regulator, 2017; Dompierre et al., 2017).

Base Mine Lake is the first commercial-scale demonstration EPL in the Athabasca oil sands region of northern, Alberta. Approximatley 183M m^3 of FFT was pumped into the former West-In-Pit over 19 years leading up to BML commissioning in December 2012 (Dompierre et al., 2016). These tailings were initially capped with a combination of OSPW and freshwater pumped from a nearby reservoir. The water cap depth increased steadily from approximately 5 m to between 8 and 12 m from 2012 to 2018 due to FFT settlement. Internal mass loading from the FFT into the lake is driven advectively by the expressed pore-water associated with settlement; however, the geochemical gradients and seasonal fluctuations in loading are also influenced by the diffusion/dispersion that occurs within the FFT immediately below the TWI. Improving understanding of chemical mass transport processes and, therefore, internal mass loading is critical for forecasting BML cap water quality over time.

Methods

One-dimensional numerical models of advective-dispersive transport were used to constrain mass fluxes and geochemical gradients at the TWI (Dompierre and Barbour 2016; Dompierre et al., 2017). These models had to invoke an annual physical mixing event over the upper 1 m of FFT to simulate observed profiles of conservative species (i.e. Chloride, stable isotopes of water). Although these models could be calibrated against the observed profiles, several lines of evidence suggested that the mixing was likely associated with CH_4 ebullition. This hypothesis has potentially important implications for numerical modelling since CH_4 ebullition is likely seasonally variable yet continuous process. Here, we explore whether mixing within the upper FFT results from the generation, exsolution and ebullition of methane during natural *in situ* biodegradation of diluent hydrocarbons (Foght et al., 2017; Rudderham 2019; Stasik and Wendt-Pothoff, 2015). Specifically, we determine the potential for methane exsolution and ebullition within BML through field measurements and thermodynamic modelling. We then develop numerical models of conservative mass transport using appraoches to mimic mixing by CH_4 ebullition.

We conducted field sampling and measurement campaigns at BML from 2015 through 2019. We collected and analyzed samples to determine dissolved ion (e.g., chloride) and gas (i.e., CH_4, carbon dioxide) concentrations within FFT pore water and across the TWI (Rudderham, 2019). We also measured *in situ* fluid and total dissolved gas pressures. We performed thermodynamic modelling of theoretical solubility and determined degree of saturation to assess potential for CH_4 exsolution (Duan and Mao, 2006). We performed dissolved atmospheric noble gas analysis to provide confirmation that CH_4 ebullition occurs within the upper FFT (Jones et al., 2014). We then incorporated ebullitive mixing into previous advective-dispersive transport models as a spatially variable enhanced diffusion coefficient that reflected seasonal changes in CH_4 solubility and ebullition rates.

Results

Our field sampling and thermodynamic modelling revealed a methanogenic zone positioned within the FFT approximately 1 to 3 m below the tailings-water interface. Dissolved CH_4 concentrations approach or reach theoretical saturation within this zone. Total dissolved gas pressures were also observed to exceed *in situ* fluid pressures over these depths. Depletion of relatively light (i.e., Ne, Ar) to heavy (i.e., Xe, Kr) dissolved atmospheric noble gases also indicated that ebullition has occurred over the same depth interval (Jones et al., 2014). Thermodynamic modelling results show that CH_4 solubility within the FFT (i) increases over time during FFT settlement and (ii) varies with seasonal temperature cycles. Although CH_4 ebulltion at BML has been observed year round, these combined results indicate that mixing due to ebullition is likely strongest from August to October as FFT temperatures rise and are declining gradually over time.

Numerical models that incoroporate seasonally variable mixing due to ebullition were better able to simulate the observed depth profiles of concervative tracers. The amount of mixing required was correlated with the degree of CH_4 saturation and estimated ebullition rate for a given location. During periods of greater ebullition (i.e., August through October), the chemical mass flux rate across the tailing-water interface increased due to the enhanced ebullitive mixing.

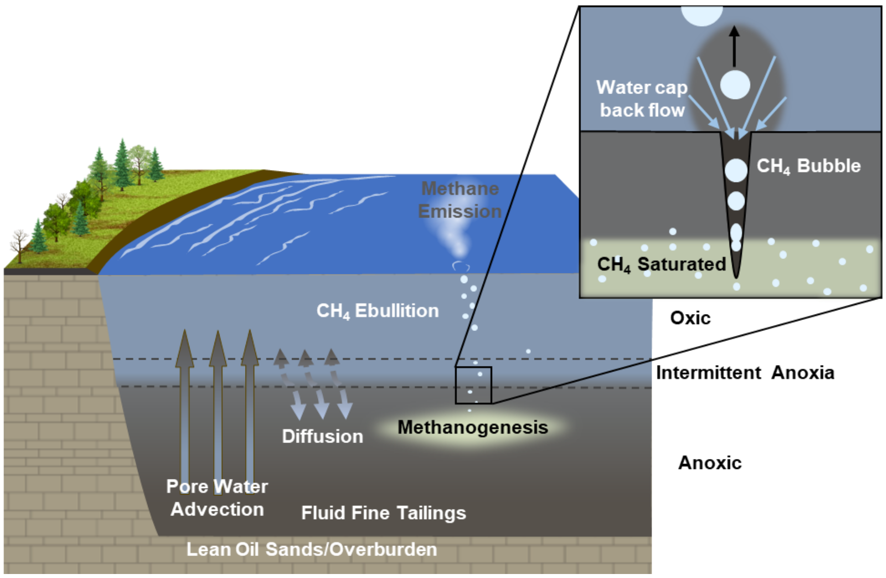

Figure 1. Conceptual model of processes affecting chemical mass transport across the tailings-water interface of Base Mine Lake.

Conclusions

Our results provide critical insight into processes that influence seasonal variations in geochemical gradients and chemical mass transport across the TWI of the first commercial scale oil sand EPL (Figure 1). These transport processes are directly influenced by CH_4 production and ebullition. Although the long-term influence of CH_4 ebullition within BML is not yet known, our findings suggest that limiting methanogensis within EPLs may expedite improvements in cap water quality.

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