



Microcosm assessment of the biogeochemical development of sulfur and oxygen in oil sands fluid fine tailings



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ABSTRACT

Bitumen recovery from Alberta oil sands generates fluid fine tailings, which are retained in tailings ponds where solids settle and release process water. The recovered water is recycled for bitumen extraction, while the resulting tailings are incorporated into various landforms for reclamation, with one option being conversion of tailings basins to viable end pit lakes. Tailings ponds commonly host diverse microbial communities, including SO₄-reducing prokaryotes. The highly reducing nature of the hydrogen sulfide produced by these prokaryotes may impact the biogeochemical cycling of key nutrients. However, the behavioral dynamics of hydrogen sulfide production in ponds containing fluid fine tailings remain to be clearly explained. In this study, microcosms are used as analogues of the sediment–water interface of a tailings pond undergoing reclamation to determine sulfide generation patterns and the behavior of O₂. In the microcosms, hydrogen sulfide fluxes correlated positively with biotic activity, reaching levels of over $2 \times 10^3 \text{ nmol cm}^{-2} \text{ s}^{-1}$, leading to Fe sulfide formation. Depth-related hydrogen sulfide profiles in the microcosms were comparable to those encountered *in situ*, in Syncrude's West In-Pit, an active tailing pond. Oxygen diffusion across the fluid fine tailing sediment–water interface was controlled to different degrees by both biotic and abiotic processes. The results have implications for quantitatively estimating the impact of hydrogen sulfide production, O₂ availability, and biogeochemical cycling of key nutrients important for the success of life in fluid fine tailings-affected ecosystems. This paper shows that this production of hydrogen sulfide may be a self-limiting process, which will begin to decrease after a period of time.

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1. Introduction

The Athabasca oil sands in northeastern Alberta, Canada comprise the third largest hydrocarbon reserve in the world next to Saudi Arabia and Venezuela, estimated at ~1.7 trillion barrels of recoverable bitumen (Chalaturnyk et al., 2002; Alberta Energy, 2013). Currently, ~1.3 million barrels are extracted daily for commercial use (Alberta Energy and Utilities Board, 2007; Alberta Energy, 2013), and production is anticipated to grow following new advances in extraction technologies, coupled with global decline of conventional oil reservoirs (Williams, 2003). With new projects added annually, production is expected to reach 3 million barrels/day by 2018 (Alberta Energy, 2013).

After surface mining, bitumen is separated from the oil sands ore by a warm water process, resulting in tailings consisting of 20–30 wt% solids (mostly clay and sand), ~3 wt% residual bitumen, and alkaline water (Chalaturnyk et al., 2002). Producers comply with a zero-discharge policy, by transferring tailings to on-site

tailings ponds, where the sand quickly settles out and the resulting clay suspension is allowed to dewater through density-dependent settling, which produces fluid fine tailings (FFT). The released water is either recycled for further bitumen extraction or discharged and used for on-site landscape reconstruction (Siddique et al., 2006). The consolidated FFT is destined for integration into reclaimed landforms, and one option being considered is conversion of existing tailings ponds into future end-pit lakes (EPLs). The tailings pond water cap has a unique chemical composition, consisting mostly of recycled process water containing elevated Na⁺ (termed brackish) concentrations, SO₄²⁻ and Cl⁻ anions, naphthenic acids and NH₃. Current evidence suggests natural attenuation mechanisms result in the removal or degradation of naphthenic acid compounds and NH₃, as well as a general shift in the water chemistry toward less saline natural water composition as the system ages (Scott et al., 2005). The underlying chemical factors contributing to the processes operating during maturation between the FFT sediment and overlying water in tailing pond basins, however, still require explanation.

In order to develop a sustainable aquatic ecosystem in locations such as reclaimed EPLs, it is essential to have a detailed

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understanding of the long term impacts that FFT will have on both the physical environment and living organisms as these systems develop. This study addresses these issues by focusing on the biotic and abiotic processes contributing to the development of key elemental cycles (e.g. S, Fe, N, and C) that may perturb or stabilize these reclaimed environments. Assessing the short and long term biogeochemical drivers, as well as production rates of highly reactive dissolved species, like HS^- , will be essential to predicting the establishment of these elemental cycles. Faunal abundance and biomass in natural aquatic ecosystems correlate positively with sediment O_2 demand (SOD) (Tahey et al., 1994), which is comprised of biological O_2 demand (BOD) and chemical O_2 demand (COD) (Wang and Reed, 1984; Gelda et al., 1995). COD dominates epibenthic processes found in young marshes created with oil sands process material (OSPM) (Gardner Costa, 2010).

In this context, the SOD function of the FFT will directly influence dissolved O_2 (DO) concentrations in the water column, and thus, can be affected by a combination of biological (i.e., bacteria – BOD) and chemical processes (COD) occurring at the FFT sediment–water interface. The principal source of BOD is the biological respiration associated with bacterial decay processes at the FFT sediment–water interface. This biological activity can influence the release of chemically reduced compounds via bacterial respiration, such as iron, sulfide, methane and ammonia. These soluble compounds will exert a rapid kinetic O_2 demand as the reduced materials undergo oxidation (Gelda et al., 1995; Matzinger et al., 2010).

Current data suggest various microbial consortia responsible for driving distinct redox-dependent biogeochemical processes have developed in established FFT settling basins (Penner and Foght, 2010; Ramos-Padrón et al., 2011). These complex associations include anaerobic heterotrophs, sulfate-reducing prokaryotes (SRP), and methanogens (Clemente and Fedorak, 2005; Siddique et al., 2006, 2007; Penner and Foght, 2010; Ramos-Padrón et al., 2011). Substantial evidence also indicates that S cycling will be crucial in governing successional processes in EPL settings, and seems to be a dominant determinant of the spatial organization of the various microbial communities (Penner and Foght, 2010; Ramos-Padrón et al., 2011; Chi Fru et al., 2013) linked to sulfide production and methanogenesis (Holowenko et al., 2000; Salloum et al., 2002).

However, little is known about the specific rates of generation and consumption of key oxidants and reductants in EPLs, such as O_2 , SO_4^{2-} and HS^- or the factors responsible for controlling the biogeochemical dynamics of these redox-sensitive species. This is especially true for HS^- generation, as this can stress eukaryotic organisms (Williford et al., 2009). The HS^- production might critically limit microbial and metazoan colonization in the overlying water column, where natural communities are expected to develop following the reclamation of the previously mined areas. In addition, HS^- plays a key role in regulating trace metal solubility in many aquatic environments, especially Fe, which precipitates to form Fe sulfides (Schoonen, 2004). Iron is important both as an essential trace element for microbial growth (Archer and Johnson, 2000; Church et al., 2000), and as a terminal electron acceptor and donor during microbial metabolism (Weber et al., 2006; Crowe et al., 2007; Chi Fru et al., 2012). Thus, HS^- generation linked to SRP individuals within EPLs may play a fundamental role in both Fe cycling and microbial growth.

As mentioned above, O_2 availability will be a factor determining the speed and success of microbial colonization and sustainability in EPLs, especially in laying the foundation for establishing aquatic conditions that can sustain a complete food web. In addition, rates of O_2 diffusion into the FFT will determine the degradation rates of recalcitrant hydrocarbons. Hydrocarbons are much more biodegradable under oxic conditions (Leahy and Colwell, 1990).

However, specific groups of microorganisms, such as some members of the SRP can degrade hydrocarbons under anoxic conditions (Rueter et al., 1994). In particular, naphtha (a residual solvent remaining in the FFT after froth treatment) and organic acids present in the original oil sands ore that are concentrated during the extraction process can be biodegraded both in the absence and presence of O_2 (Scott et al., 2005; Siddique et al., 2006, 2007, 2011). Therefore, the development of redox gradients in the FFT, as a function of O_2 availability and demand, will likely facilitate various biogeochemical processes at different rates.

This study investigates the behavior of S and O_2 at the FFT–oil sand process water (OSPW) interface, in order to assess these primary chemical components in EPL settings over short and long term time periods, using a combination of microcosms. Previous work (Chi Fru et al., 2013) demonstrated the ability of this approach to faithfully simulate *in situ* development of microbial community structures similar to those observed in the existing West In-Pit lake (WIP) storage basin. This study compares the development of a defined redox front in both field and laboratory settings for the WIP material, which may influence the geochemical cycling of elements within the pond. Specifically, the experiment poses and answers the question of whether these processes are predominantly regulated by biotic or abiotic driven activity, with respect to S and O_2 .

2. Materials and methods

2.1. Microcosm setup

Approximately 200 L of fresh FFT and 150 L of OSPW samples were collected from the West In-Pit (WIP) tailings pond operated by Syncrude Canada Ltd., approximately 40 km north of Fort McMurray. The tailings pond is part of the In-pit facility (formerly mined out area) that was commissioned in 1995. The tailings pond was isolated from the tailings process system in 2012 and is being considered for reclamation as an end pit lake containing water-capped FFT as part of Syncrude's reclamation strategy. The FFT sample consists of 33 wt% solids (e.g., sand, clay, etc.), 65 wt% water with residual 1.97 wt% bitumen (Table 1), and less than 0.06 wt% soluble organics (not shown). The samples were collected 10 M below the mudline and are considered to be FFT based on water content. Samples collected were stored and shipped at 4 °C. A subset of FFT samples (100 L) and OSPW (75 L) was gamma irradiated at 28 kGy for 24 h, at the McMaster Institute of Applied Radiation Services (McIARS), Hamilton, Ontario, Canada to serve as a sterile control.

A 2×2 factorial study design was used to distinguish the effects of atmosphere (oxic vs. anoxic environments) and microbial activity (biotic vs. abiotic microcosms) on the evolution of S and O_2 ($n = 10$ replicate microcosms per treatment). Microcosms were constructed from ethanol-sterilized 4 L Camwear® containers purchased from Cambro Manufacturing Company. The anoxic microcosms were fitted with covers equipped with O-ring airtight seals and two push-plug valves for headspace gas sampling and purging, if required (Fig. 1).

The FFT was thoroughly mixed using a power portable drill to which was attached a sterilized Teflon-coated stirring paddle. A 2000-g aliquot of FFT was added to each microcosm and overlaid with 1000 mL of OSPW. Anoxic microcosms were then flush cycled with ultra pure N_2 gas for 5 min, and cycled additionally when set inside the gas (95% N_2 , 5% H_2 mixture) chamber. Oxic microcosms were incubated on shelves with unmodified lids without seals. All microcosms were kept in darkness at 22 °C. Individual microcosms were destructively analyzed after 0, 6, 24, 36, and 52 weeks ($n = 2$ per treatment).

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