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Research article

Elucidating carbon sources driving microbial metabolism during oil sands reclamation

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ABSTRACT

Microbial communities play key roles in remediation and reclamation of contaminated environments via biogeochemical cycling of organic and inorganic components. Understanding the trends in *in situ* microbial community abundance, metabolism and carbon sources is therefore a crucial component of effective site management. The focus of this study was to use radiocarbon analysis to elucidate the carbon sources driving microbial metabolism within the first pilot wetland reclamation project in the Alberta oil sands region where the observation of H_2S had indicated the occurrence of microbial sulphate reduction. The reclamation project involved construction of a three compartment system consisting of a freshwater wetland on top of a sand cap overlying a composite tailings (CT) deposit. Radiocarbon analysis demonstrated that both dissolved and sediment associated organic carbon associated with the deepest compartments (the CT and sand cap) was primarily fossil ($\Delta^{14}C = -769$ to -955‰) while organic carbon in the overlying peat was hundreds to thousands of years old ($\Delta^{14}C = -250$ to -350‰). Radiocarbon contents of sediment associated microbial phospholipid fatty acids (PLFA) were consistent with the sediment bulk organic carbon pools (Peat: $\Delta^{14}C_{PLFA} = -257$‰; Sand cap $\Delta^{14}C_{PLFA} = -805$‰) indicating that these microbes were using sediment associated carbon. In contrast, microbial PLFA grown on biofilm units installed in wells within the deepest compartments contained much more modern carbon that the associated bulk carbon pools. This implied that the transfer of relatively more modern carbon was stimulating the microbial community at depth within the system. Correlation between cellular abundance estimates based on PLFA concentrations and the $\Delta^{14}C_{PLFA}$ indicated that the utilization of this more modern carbon was stimulating the microbial community at depth. These results highlight the importance of understanding the occurrence and potential outcomes of the introduction of relatively bioavailable carbon to mine wastes in order to predict and manage the performance of reclamation strategies.

1. Introduction

It is estimated that mining activities produce a total volume of 7125 Mt/year of tailings worldwide across all extractive industries (Mudd and Boger, 2013). Many reclamation landscapes designed to manage these materials involve waste materials high in organic compounds as well as sulphur and iron constituents that are important terminal electron acceptors for organic carbon degradation, so proper design and material placement with an understanding of the microbial biogeochemical cycling is required. Microbial biogeochemical cycling is an important component of the functioning of any ecosystem, including mine waste deposits. The extent and impact of this cycling in any given system is determined by the abundances, carbon sources and metabolisms of in situ microbial communities. For instance, microbial carbon cycling can remove organic contaminants from a system via mineralization during cellular metabolic activities (e.g. Megharaj et al., 2011). Numerous studies have assessed the role of these biodegradation processes in determining the fate and transport of organic compounds (Essaid et al., 2015). This microbial biogeochemical cycling can also be associated with the mobilization or generation of undesirable metabolic products, such as the hydrogen sulphide (H_2S) generated by sulphate reducing bacteria (SRBs) (Muyzer and Stams,

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2008). The sources and cycling of the inorganic reactants that are involved in redox cycling, such as sulphate concentrations in the case of the activities of SRBs, have likewise been well studied (Vile et al., 2003; Wu et al., 2013, 2011). However, in the latter cases, the organic carbon pool is often not well characterized, despite its role as the electron donor driving redox cycling.

The pool of organic carbon present in an environmental system is often complex and comprised of a wide range of compounds with a wide range of bioavailabilities. In many systems, such as soils, the biological cycling of organic compounds has been related to their age, with recent biogenic compounds cycling quickly and residual recalcitrant compounds cycling slowly based on the radiocarbon ages of soil carbon fractions and respired CO_2 (Trumbore, 2009). The development of techniques allowing direct assessment of the radiocarbon content of microbial cellular constituents such as phospholipid fatty acids (PLFA) enables a new perspective on such studies (Mahmoudi et al., 2013a, 2013b; Petsch et al., 2001; Slater et al., 2005; Whaley-Martin et al., 2016). Using this approach, the biogeochemical cycling of petroleum hydrocarbons has been shown to range from very rapid utilization and degradation by the in situ microbial community (Mahmoudi et al., 2013b) to very little degradation of the petroleum hydrocarbons due to the preferential utilization of natural organic matter (Mahmoudi et al., 2013a; Slater et al., 2005). Management of mine waste materials often involves high levels of both recalcitrant organic material and redox sensitive species that have been excavated from below ground. Since waste materials management often occurs at the ground surface, there is a considerable potential for exchange of materials and microbial communities derived from surface ecosystems with the wastes being managed, intentionally or not. This mixing has the potential to drive significant changes in biogeochemical cycling. In particular, the introduction of recently produced, bioavailable organic matter may stimulate microbial biogeochemical cycling of redox reactive species to a far greater extent than the recalcitrant organic matter associated with the mine waste materials. Such stimulation may drive generation of metabolites that require management, such as H_2S or mobilized metal constituents. Understanding the interplay between the surface environment surroundings and mine wastes is thus crucial in developing effective management strategies.

The large volume of oil sands processed by extraction plants in Alberta's oil sands mines results in some of the largest tailings facilities in the world (COSIA, 2014), holding a volume of tailings waste exceeding 700 million m^3 (Dominski, 2007). These significant volumes highlight the critical need to understand the biogeochemical process associated with the material. Reclamation of oil sands tailings is made more challenging than in other resource sectors because the waste exists as fluid fine tailings (FFT) with very slow sedimentation and consolidation rates (COSIA, 2014). One approach used to manage FFT is to amend it with gypsum to reduce the double diffusive layer around the fine sized (<44 μm) clay minerals that are present and then combine with sand. After dewatering, the resulting more consolidated mixture of sand, residual bitumen, clay fine and gypsum is referred to as composite tailings (CT).

Syncrude is currently undertaking the first pilot scale wetland reclamation project in the Alberta Oil Sands Region. A goal of this project was to construct the initial conditions to allow the development of a fen wetland above a deposit of CT over time. This wetland watershed was constructed on a sand-capped CT deposit and is a permanently reclaimed area that will contribute to the final closure landscape as committed to in regulatory approvals (Wytrykush et al., 2012). This pilot watershed research facility will provide data to support future large-scale wetland reclamation projects and address challenges associated with tailings reclamation providing important insight for the management of oil sands tailings specifically and for reclamation efforts more broadly. Early in the wetland construction process (2009) H_2S gas was episodically detected associated with surface dewatering wells, suggesting the occurrence of microbial sulphate reduction within the underlying CT and/or sand cap materials. Hot water extraction of sand removes 88–95% of bitumen (Masliyah et al., 2004) and the residual bitumen organic carbon present in CT is highly recalcitrant, so that microbial sulphate reduction was assumed to be limited by access to labile organic carbon. However, the observation of H_2S generation raised the question whether introduction of younger, relatively more labile organic carbon from the developing wetland was stimulating microbial sulphate reduction within the system (Reid and Warren, 2016). The objectives of the current study were to elucidate the carbon sources being utilized by the in situ microbial communities within the fen/CT system. Specifically, we investigated whether inputs of more modern, more bioavailable organic carbon sources from the surface environment were driving increased microbial activity and stimulating H_2S production.

This study focussed on both un-reclaimed (no reclamation soil cover placed) and reclaimed (reclamation soil cover placed) CT. For the purposes of this study the reclamation project was divided into three “compartments”. The deepest compartment was the ~35 m of CT that had been deposited in Syncrude's East In Pit. The intermediate compartment was a ~10 m “sand cap” that had been placed over the CT deposit. The uppermost compartment consisted of a 0.5 m layer of clay overlain by a 0.5 m layer of peat salvaged from mine advancement. The peat layer was planted with local plant species and flooded in order to establish the initial conditions for peat forming wetlands to develop over time (Fig. 1). Characterization of microbial carbon sources and abundances was achieved by collecting samples that represented each compartment (surface peat, sand cap, and CT) of the system using a range of approaches. This included surface collection of solid matrix materials where accessible, installation of biofilm units in monitoring wells within the sand cap and CT compartments, and direct drilling to sample the CT deposit at an adjacent unreclaimed site. The abundance of microbial biomass within each compartment was determined via phospholipid fatty acid (PLFA) analysis. PLFA degrade within days to weeks after cell death (Harvey et al., 1986; White et al., 1979) and thus represent the viable bacterial and microeukaryotic community at a site. Concurrent radiocarbon values of potential carbon sources (total organic carbon (TOC), residue after solvent extraction (extracted residue: EXT-RES), and dissolved organic carbon, (DOC)), combined with compound specific radiocarbon analysis (CSRA) of PLFA enabled determination of microbial carbon source utilization by these microbial communities. This approach is based on the fact that petroleum hydrocarbons are millions of years old and thus contain no significant ^{14}C ($\Delta^{14}C = -1000‰$) while organic carbon recently produced from the atmosphere will have modern levels ($\Delta^{14}C$ = ~55‰ (Turnbull et al., 2007)). Since $\Delta^{14}C$ is normalized to a $\delta^{13}C$ of −25‰ during data handling to remove the effects of biosynthetic fractionation (Stuiver and Polach, 1977), the $\Delta^{14}C$ of microbial phospholipids can be directly compared to potential carbon sources to apportion the extent of their utilization. This approach has been used to directly identify microbial carbon sources during intrinsic (Ahad and Pakdel, 2013) and engineered bioremediation (Cowie et al., 2010, 2009), as well as the lack of significant degradation of recalcitrant petroleum hydrocarbons in the presence of more labile carbon sources (Mahmoudi et al., 2013a; Slater et al., 2005). Recently, this technique has been used to identify the role of inputs of relatively modern, bioavailable organic carbon in driving release of arsenic into shallow groundwater in Bangladesh (Whaley-Martin et al., 2016). However, to our knowledge, this is the first time that it has been applied to address carbon sources driving microbial cycling at a site where mine