



# Microbial interactions with naturally occurring hydrophobic sediments: Influence on sediment and associated contaminant mobility



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## ABSTRACT

The erosion, transport and fate of sediments and associated contaminants are known to be influenced by both particle characteristics and the flow dynamics imparted onto the sediment. The influential role of bitumen containing hydrophobic sediments and the microbial community on sediment dynamics are however less understood. This study links an experimental evaluation of sediment erosion with measured sediment-associated contaminant concentrations and microbial community analysis to provide an estimate of the potential for sediment to control the erosion, transport and fate of contaminants. Specifically the paper addresses the unique behaviour of hydrophobic sediments and the role that the microbial community associated with hydrophobic sediment may play in the transport of contaminated sediment. Results demonstrate that the hydrophobic cohesive sediment demonstrates unique transport and particle characteristics (poor settling and small floc size). Biofilms were observed to increase with consolidation/biostabilization times and generated a unique microbial consortium relative to the eroded flocs. Natural oil associated with the flocs appeared to be preferentially associated with microbial derived extracellular polymeric substances. While PAHs and naphthenic acid increased with increasing shear (indicative of increasing loads), they tended to decrease with consolidation/biostabilization (CB) time at similar shears suggesting a chemical and/or biological degradation. PAH and naphthenic acid degrading microbes decreased with time as well, which may suggest that there was a reduced pool of PAHs and naphthenic acids available resulting in their die off. This study emphasizes the importance that any management strategies and operational assessments for the protection of human and aquatic health incorporate the sediment (suspended and bed sediment) and biological (biofilm) compartments and the energy dynamics within the system in order to better predict contaminant transport.

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## 1. Introduction

The lower Athabasca River and many of its tributaries (e.g. Eells River) cut through the Fort McMurray Formation, the geological strata that constitute the Oil Sands deposit in northern Alberta, Canada. Visual observations of the lower Eells River shows occasional areas of oil sheens at the water–bank interface and the development of oil sheens when disturbing recently deposited sediment via samplers or footsteps. For the Eells River, Yergeau et al. (2012) showed a two order of magnitude increase in total petroleum hydrocarbons, total single-chain hydrocarbons, total aromatic

hydrocarbons, and an order of magnitude increase in the sum of USEPA 16 priority PAHs moving from headwaters to mouth. Whether this was an accumulation of natural PAHs with progression downstream or a site specific change due to geological variations was not determined (active mining in this basin has not yet commenced). High natural petroleum hydrocarbon content has translated to very hydrophobic sediment within this basin. Hydrophobicity measurements on the Eells River range from 34% (17 km upstream from mouth) (this study) to as high as 96% (near confluence with Athabasca River) (Droppo et al., 2015). It is known that hydrophobic micropores associated with the sediment particles/flocs themselves can retain significant amounts of organic contaminants while at the same time allowing for slow release (Cheng et al., 2012). Nonspecific hydrophobic interactions are

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acknowledged as an important mechanism for organic pollutant binding to natural organic matter (Chefetz et al., 2000). Further, it has been shown that biofilm productivity and biomass collected from the Athabasca River and tributaries (including the Ells River) can be reduced with exposure to natural bituminous (oil) compounds (Yergeau et al., 2013). The unique high oil content (relative to other rivers) of the sediment also provides for a selective community structure that can associate with such conditions (Droppo et al., 2015; Yergeau et al., 2012); often hydrocarbon consumers. While hydrophobic contaminant sorption/desorption with sediments (particularly soils) has been studied extensively, (e.g., Cheng et al., 2012; Karickhoff et al., 1979), the general hydrophobic characteristic of particle, regardless of the origin of the hydrophobicity, has received very little attention with respect to their influence on sediment dynamics and microbial interactions.

Droppo et al. (2015) have shown that microbial interactions with bed sediments for the Ells River plays a role in the stabilization of the sediments from erosive forces and influences eroded floc size, shape and density. Microbial interactions can increase the energy required to erode bed sediment (Gerbersdorf and Wierpecht, 2015) with Droppo (2009) showing bed sediments to be up to 10 times stronger with biofilms than without. Further, the elastic/plastic nature of eroded flocculated particles (provided by the attached microbial community/biofilm integration) can generate considerable strength relative to electrochemically bound flocs (Liss et al., 1996). The level of microbial activity within the water column has been shown to be influenced by the sediment dynamics within the system (Walters et al., 2014a, 2014b). Microbial mediation of sediments (bed and/or suspended) can also play a strong role in the biogeochemistry of the sediment and overlying water column [e.g., influence on redox reactions (Elliott et al., 2014; Elliott and Warren, 2014; Saulnier and Mucci, 2000)].

While it is known that sediment dynamics (erosion, transport and fate) can play a significant ecological role within river systems, influencing all trophic levels, geomorphological processes, and with possible significant socioeconomic implications (Grabowski et al., 2011; Chapman, 1988) knowledge of the factors controlling these sediment processes [particularly those of hydrophobic (oil associated) sediments] is relatively limited. An improved understanding of hydrophobic sediment dynamics and the role that the microbial community may play in this, will help better predict the fate and effect of upstream sources of contaminants to downstream impact areas. As such, this study links an experimental evaluation of sediment erosion with measured sediment-associated contaminant concentrations and microbial community analysis to provide an estimate of the potential for sediment to control the erosion, transport and fate of contaminants in the Ells River, Alberta, Canada. The specific objectives of this paper are 1) to describe the unique behaviour of hydrophobic sediments within a river system, 2) to improve our understanding of the microbial community structure associated with hydrophobic sediment and how its development may influence associated contaminant transport.

## 2. Methods

### 2.1. Field program

#### 2.1.1. Study river

The Ells River has a mean annual flow of  $9 \text{ m}^3 \text{ s}^{-1}$  and drains an area of 2450 square km (Environment Canada, 2011; Carson, 1990). The River drains into a variety of different soil types with the headwaters beginning in the Birch Mountains. These soils range from non to exceedingly stony, loams to clay and are predominately formed on calcareous till with the exception of the many bogs surrounding the river (Alberta Environment, 1982). The lower

portion of the river cuts through the Fort McMurray formation (Conly et al., 2002). This basin is only beginning to be developed for in situ bitumen extraction and represented a relatively undisturbed basin at the time of sampling.

#### 2.1.2. Bed sediment sampling for sediment dynamic assessment

A sediment-water mixture (1000 L) from the river was collected from a mid-stream location ( $57^\circ 14' 43.43'' \text{N}$ ;  $111^\circ 43' 57.62'' \text{W}$ ) on October 5, 2012 using a submersible pump with mild bed disturbance upstream of the pump to mobilize recently deposited sediment. The pump sampling was carried out at several locations within a 50 m length of the river. The sediment-water mixture was transported to the laboratory in a refrigerated ( $4^\circ \text{C}$ ) transport truck.

Additional bulk bed sediment (100 Kg) was collected in polyethylene 20L buckets by using a stainless steel shovel cleaned with acetone to scrape the top 1 cm of deposited sediment within a backwater area at the site. This additional sediment was used to form a cohesive bed surface within the flume (described below) during erosion experiments.

### 2.2. Characterisation of fine sediment dynamics

Erosion characteristics of fine sediments along with bio-sediment interactions of the Ells River were studied experimentally using a laboratory rotating annular flume (Krishnappan, 1993). The flume consists of a 5.0 m in mean diameter, 0.30 m wide and 0.30 m deep channel with a counter rotating top cover (ring) that fits just inside the flume ( $\sim 1.5 \text{ mm}$  gap on either side) and makes contact with the water surface within the flume (Krishnappan, 1993). The flows generated in the flume are close to two dimensional with a bed shear stress distribution across the width of the flume that is relatively uniform (Krishnappan and Engel, 2004). The flume calibration results of Krishnappan and Engel (2004) were used to predict the relationship between the bed shear stress and the rotational speeds of the flume.

#### 2.2.1. Erosion experiment

Ells River bed sediment was added to the flume (sieved at  $250 \mu\text{m}$  to remove organic plants and large material) which was then mixed at a high rate of speed for 30 min to thoroughly mix the sediment and water. The flume speed was then gradually reduced to a stop to allow the mixture to settle and consolidate/biostabilize (bed thickness approximately 2 cm). Three different consolidation/biostabilization (CB) periods were used in these experiments (3-, 6- and 9-days). Following a CB period, the flume and the lid were then set in motion and their speeds were incrementally increased in 60 min time intervals (bed shear intervals used were 0.028, 0.046, 0.072, 0.1, 0.134, 0.169 and 0.211 Pa). Suspended solid (SS) concentrations were estimated using a calibrated flush mount optical backscatter sensor (OBS) located on the outer side of the flume at mid depth. Additional sediment samples were collected every 10 min to measure the SS concentration variation as a function of time and for additional calibration of the OBS probe. The critical bed shear for erosion ( $\tau_c$ ) was defined as the bed shear stress at which the SS showed a well-defined increase in concentration. After 50 min of a given shear step applied, an additional 50 mL of water/sediment suspension was collected for biological analysis as per Section 2.6 and 2.7. To maintain water contact with the lid of the flume all sample volumes removed were immediately replaced with additional river water. Sediment-water samples were analysed for concentration of solids by a gravimetric method that consisted of filtering the sample ( $0.45 \mu\text{m}$  pre-weighed Millipore filter), and drying and weighing the residue.

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