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Metal bioaccumulation and biomarkers of effects in caged mussels exposed in the Athabasca oil sands area

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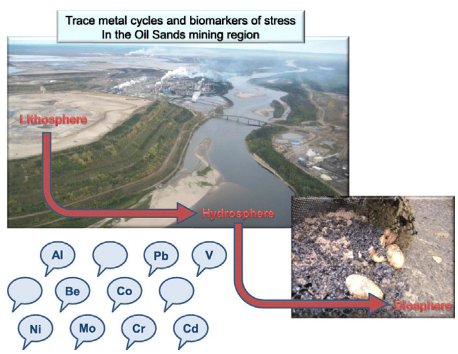
HIGHLIGHTS

- Significant oils sands mining-related metal exposure
- Metal partitioning largely influenced by hydrology and flows level.
- Bioavailability of trace metals and bioaccumulation in mussels
- Potential ecotoxicological risk to biota and the aquatic environment
- Environmental cost associated to mining activity.

GRAPHICAL ABSTRACT



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ABSTRACT

The Athabasca oil sands deposit is the world's largest known reservoir of crude bitumen and the third-largest proven crude oil reserve. Mining activity is known to release contaminants, including metals, and to potentially impact the aquatic environment. The purpose of this study was to determine the impacts of oil sands mining on water quality and metal bioaccumulation in mussels from the Fort McMurray area in northern Alberta, Canada. The study presents two consecutive years of contrasting mussel exposure conditions (low and high flows). Native freshwater mussels (*Pyganodon grandis*) were placed in cages and exposed in situ in the Athabasca River for four weeks. Metals and inorganic elements were then analyzed in water and in mussel gills and digestive glands to evaluate bioaccumulation, estimate the bioconcentration factor (BCF), and determine the effects of exposure by measuring stress biomarkers. This study shows a potential environmental risk to aquatic life from metal exposure associated with oil sands development along with the release of wastewater from a municipal treatment plant nearby. Increased bioaccumulation of Be, V, Ni and Pb was observed in mussel digestive glands in the Steepbank River, which flows directly through the oil sands mining area. Increased bioaccumulation of Al, V, Cr, Co, Ni, Mo and Ni was also observed in mussel gills from the Steepbank River. These metals are naturally present in oil sands and generally concentrate and increase with the extraction process. The results also showed different pathways of exposure (particulate or dissolved forms) for V and Ni resulting from different river water flows, distribution coefficient (K_d) and BCF. Increasing metal exposure downstream of the oil sands mining area had an impact on metallothionein and lipid peroxidation in mussels, posing a potential environmental risk to aquatic life. These results confirm the bioavailability of some metals in mussel tissues associated with detoxification of metals (metallothionein levels), and oxidative stress in mussels located downstream of the oil sands mining area. These results highlight a potential ecotoxicological risk to biota and to the aquatic environment downstream of the oil sands mining area, even at low metal exposure levels.

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1. Introduction

The potential resource value of oil sands was first observed and documented centuries ago (Gosselin et al., 2010). The occurrence of bituminous oil sands and oil film either on river shore or surface water was undoubtedly a natural source of petroleum products (Lamb, 1970). Moreover, sediment core analysis shows that the anthropogenic footprint of mining activities has increased significantly in recent decades and exploded in the 21st century in tandem with increased oil demand and production (Kurek et al., 2012). Commercial production, which began in the mid-1900s, has impacted some of the major components of the surrounding ecosystems (air, soil and water) (Lynam et al., 2015; Korosi et al., 2013; Barrow et al., 2015), causing a high potential environmental risk to the regional environment (Swart and Weaver, 2012) and the global footprint (Hoag, 2013). Environmental pressures on the surrounding ecosystems and global system may further increase in coming decades given the estimated oil reserves of the Canada's oil sands (~170 billion barrels of crude raw bitumen), the world's third largest proven crude oil reserves (Alberta Energy, 2015). Transition from traditional oil resources to oil sands development has increased the financial, energy and environmental costs because unconventional oil reserves are more complex to extract (Murphy, 2014).

Recent studies showed the impact of oil sands development on the aquatic ecosystem. Ross et al. (2012) identified naphthenic acids (NAs) in natural surface water and groundwater and highlighted potential bitumen contamination from oil sands process water tailings ponds. Korosi et al. (2013) observed an increase in polycyclic aromatic hydrocarbon (PAH) concentrations in lake sediments corresponding to the onset of commercial bitumen production. No significant enrichment of mercury (Hg) or other metals have been observed in sediment cores in the Athabasca oil sands region (Skierszkan et al., 2013; Neville et al., 2014). In contrast, significant loadings of Hg and other metals in the snowpack were observed surrounding the oil sands mining area related to atmospheric deposition (Kirk et al., 2014). Further, there is no evidence that Hg concentration in fish increases with oil sands activity (Evans and Talbot, 2012). However, high levels of Hg in colonial water bird eggs sampled downstream of the oil sands mining area were highly correlated with NAs and indicated similar bitumen sources (Hebert et al., 2011). Mining process water contains large amount of NAs and other organic compounds (Brown and Ulrich, 2015). Metal concentrations in the Athabasca River and its tributaries, such as Cu and Zn (Alausa and Akinyemi, 2015), as well as Sb, As, Be, Cd, Cr, Cu, Pb, Hg, Ni, Se, Ag, Ti and Zn (Kelly et al., 2010), increase significantly near or downstream of the oil sands mining area. In addition to metal concentrations in water, more information is needed on metal bioavailability and potential ecotoxicological risk to aquatic organisms exposed to contaminants related to oil sands mining.

In situ exposure of caged freshwater mussels is often used to evaluate potential sources of metal contamination and to assess the body burden of pollutants in aquatic ecosystems and wastewater effluent (Gagnon et al., 2006; Gillis et al., 2014a). Freshwater mussels filter large volumes of water and suspended particles and consequently have a large potential of metal exposure and bioaccumulation from either the water column or suspended particulate matter. Metal concentrations in mussel tissues typically increase as function of metal exposure, uptake mechanism (absorption or ingestion), metal forms and bioavailability (Yeager-Armstead and Yeager, 2007). Jebali et al. (2014) showed various levels of metal bioaccumulation (Cd, Pb, Zn, Mn and Fe) in the digestive gland, gills and muscle of mussels as a function of their ability to accumulate chemical compounds. One effect of the exposure of freshwater bivalves to metals is the production of metallothionein (MT), a metal-binding protein synthesized by the organism that is involved in the sequestration and elimination of divalent heavy metals and metalloids (Falfushynska et al., 2013). Metallothionein is also involved in the binding of reactive oxygen species such as nitric oxide, which arises from inflammation. Increased metal bioavailability may also increase oxidative stress and lead to substantial lipids damage in mussel tissues (Gagnon et al., 2014). Metal exposure in mussels can also affect the condition and health of the organism but without resulting in higher metal concentrations in mussel tissues (Otter et al., 2015).

The aim of this study was to evaluate, in situ, the bioavailability of metals in caged native mussels (*Pyganodon grandis)* to assess potential exposure to metals from oil sands mining and to evaluate the environmental factors impacting metal bioavailability. The native freshwater mussel *Pyganodon grandis* is frequently used as a sentinel species to assess metal contamination from mining areas and ore smelters (Masson et al., 2010; Cooper et al., 2013). Mining activities can significantly affect the environment and wildlife, releasing toxic and other metals to the water system, even at low toxic levels. The potential ecotoxicological risk and the impact of metal exposure on mussel metabolism were also investigated. Concentrations in water and metal partitioning were evaluated to support bioavailability data. Mussel caging deployment was done in two consecutive years to assess reproducibility and to address the temporal variability in metal bioavailability under different hydrological conditions.

2. Material and methods

2.1. Site description

The Athabasca oil sands mining area is located in the Athabasca River watershed, in northern Alberta (Canada), about 15 km north of the municipality of Fort McMurray (population of ~65,000 in 2016, and about 25,000 miners). The main geology of the area, the McMurray Formation, lies on an angular unconformity that truncates Devonian strata. The stratigraphic facies originated from deposited and accumulated fluvial, estuarine and coastal plain sediments, and was then impacted by sea level rise during the early Cretaceous (Hein et al., 2000). Eastern strata contain limestone and calcareous shale of the Waterways Formation, while western strata contain younger carbonate rocks of the Woodbend Group (Gingras and Rokosh, 2004).

Mussel exposure sites in 2012 were located along the Athabasca River (which flow mainly from south to north in this region) from upstream of the oil sands mining area (AT3) to downstream sites (AT4 and AT5), and at the mouth of the Steepbank River (SB4) in the oil sands mining area (Fig. 1). The first two letters of the site refer to the name of the river, and the last number refers to sites position from upstream to downstream. Cages were installed on riverbanks at a water depth of approximately 1 m. In the Athabasca River (~350 m wide), turbidity ranged from 13.9 to 21.6 NTU (Environment Canada (EC), 2016), and the bottom substrate ranged from bare bedrock (AT3) to a mixture of fine materials, mainly silt and gravel (AT4 and AT5). In the Steepbank River (~10 m wide), turbidity was lower 7.5 NTU (EC, 2016), and the river bottom was shallow and composed of coarse sediments, mainly sand (up to 80%) and silt surrounded by a film of bitumen. At the mouth of the Steepbank River (SB4) and other Athabasca River tributaries, flow was sufficiently high to avoid a mixing zone with the Athabasca River water. The reference site for both years, where mussels were previously collected for this exposure study, was Long Lake (LO1) (54°34′50″ N, 113°38′46″ W) in Alberta Provincial Park. It is located ~280 km southwest in a remote area (outside of the oil sands area and other direct sources of pollution). Long Lake is eutrophic and bottom sediments at the exposure site were a mixture of sand, fines and organic materials.

In 2013, mussel exposure was expanded to four sites along the Athabasca River, from upstream of the oil sands mining area (AT2 and AT3) to downstream (AT4 and AT5), and to four sites along the Steepbank River, from upstream and outside of the oil sands mining area (SB1) to downstream (SB2, SB3 and SB4) (Fig. 1). The new site along the Athabasca River (AT2) consisted of a mixture of gravel and cobble, and was located close to Fort McMurray and upstream from the municipal