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Comparative histories of polycyclic aromatic compound accumulation in lake sediments near petroleum operations in western Canada☆

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ABSTRACT

We examined the historical deposition of polycyclic aromatic compounds (PACs) recorded in radiometrically-dated lake sediment cores from a small, conventional oil and gas operation in the southern Northwest Territories (Cameron Hills), and placed these results in the context of previously published work from three other important regions of western Canada: (1) the Athabasca oil sands region in Alberta; (2) Cold Lake, Alberta; and (3) the Mackenzie Delta, NT. Sediment PAC records from the Cameron Hills showed no clear changes in either source or concentrations coincident with the timing of development in these regions. Changes were small in comparison to the clear increases in both parent and alkyl-substituted PACs in response to industrial development from the Athabasca region surface mining of oil sands, where parent PAC diagnostic ratios indicated a shift from pyrogenic sources (primarily wood and coal burning) in pre-development sediments to more petrogenically-sourced PACs in modern sediments. Cores near *in-situ* oil sand extraction operations showed only modest increases in PAC deposition. This work directly compares the history and trajectory of contamination in lake ecosystems in areas of western Canada impacted by the most common types of hydrocarbon extraction activities, and provides a context for assessing the environmental impacts of oil and gas development in the future.

1. Introduction

Canada has the third largest proven oil reserve in the world, >95% of which is located in the Alberta oil sands (CAPP, 2016). Canada is also the fifth largest natural gas producer globally (US Energy Information Administration, 2014). Examples of potentially recoverable reserves include large, proven natural gas deposits in the Mackenzie Delta of the western Arctic (Northwest Territories), oil and gas deposits in northern British Columbia and the southern Northwest Territories, and the well-known Alberta oil sands deposits, which produced 2.4 million barrels of oil per day in 2015 (CAPP, 2016). Projections suggest that, while growth in conventional petroleum developments is stabilizing in Canada, growth in unconventional developments in the oil sands will increase to 3.7 million barrels per day by 2030 (CAPP, 2016). One of the largest public concerns regarding the rapid growth of the petroleum industry in western Canada (and indeed globally) is the potential release of contaminants to the environment, which may result in deleterious ecological effects that threaten the provisioning of essential ecosystem services and negatively affect ecosystem health. In particular, oil and gas developments have been associated with the release of polycyclic aromatic compounds (PACs), a class of organic contaminants, some of which are known to be mutagenic,

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carcinogenic, or otherwise disruptive to living organisms, including humans (Boffetta et al., 1997; Baird, 2004; Camus et al., 2003; Carls and Thedinga, 2010).

PACs are a diverse group of organic compounds that contain two or more fused aromatic rings, which are formed during the incomplete combustion of organic material. PACs can broadly be categorized as pyrogenic (formed during high-temperature combustion) or petrogenic (produced over geological time scales under geothermal conditions) in origin. These compounds can enter the environment through both natural processes and anthropogenic activities, making them ubiquitous in natural systems. The predominant pyrogenic source of PACs associated with petroleum developments is the burning of fossil fuels, while oil spills, pipeline leaks and petroleum coke (petcoke) dust are important anthropogenic sources of petrogenic PACs (e.g. Zhang et al., 2016; Yan et al., 2016). PACs have been detected in a wide range of environmental compartments in Western Canada including lake and river sediments (Kelly et al., 2009; Kurek et al., 2013a), soil (Gabos et al., 2001; Irvine et al., 2014), air (Hung et al., 2005; Genualdi et al., 2009; Irvine et al., 2014), and water (Kelly et al., 2009; Hall et al., 2012), and have been shown to bioaccumulate in animal tissues (Wayland et al., 2008; Carls and Thedinga, 2010; De Laender et al., 2011; Ohiozebeau et al., 2017). Their pervasiveness, and the existence of both natural and anthropogenic sources, makes it challenging to assess whether or not petroleum exploration and extraction activities are increasing the burden of PACs in the environment.

PACs are hydrophobic and tend to partition to the sediments in aquatic ecosystems, where they are generally stable over long timescales (Simcik et al., 1999; Van Metre and Mahler, 2005, 2010). Consequently, lake sediment cores provide historical archives of PAC inputs to lakes over time, and can be used to determine whether the concentration, composition and predominant sources of PACs has changed following the onset of industrial activities. As long-term monitoring data are rarely available, and where available typically postdate the onset of industrial activities, paleolimnological techniques are often one of the few methods of accurately hindcasting the environmental contamination of anthropogenic activities. In a global context, paleolimnological studies have recorded increases in concentrations of PACs following industrial development, including in the Mediterranean (Azoury et al., 2013), in the Niger River Delta (Sojinu et al., 2010), and in the Barigui River in Brazil (Machado et al., 2014), as well as in North America (Slater et al., 2013). More specifically, in Canada, numerous recent studies have used lake sediment cores to identify changes in the burden and sources of PACs in lakes proximate to, downstream and/or downwind of operations in the Athabasca oil sands region (AOSR), where intensive surface mining of bitumen is occurring (Hall et al., 2012; Jautzy et al., 2013; Kurek et al., 2013a). These studies have shown that the onset of oil sands mining activities in the AOSR led to notable increases in PAC concentrations in lakes as far as 90 km from major operations, and that one of the main sources of PACs is petcoke dust (Zhang et al., 2016). Other paleolimnological studies have been conducted in western Canada near *in-situ* bitumen mining operations in the Cold Lake region of the Alberta oil sands (Korosi et al., 2013, 2016a), and near exploratory hydrocarbon drilling operations in the Mackenzie Delta uplands of the Northwest Territories (Thienpont et al., 2013).

Using similar paleolimnological methods, these studies of the environmental impacts of oil and gas exploration and extraction in western Canada (Fig. 1) have recorded varying degrees of PAC contamination, demonstrating that the role petroleum developments play in releasing PACs to aquatic ecosystems is complex, and varies among regions. The degree and potential for deleterious environmental impacts related to the release of PACs to the environment will depend on the type of extraction, its scale, and the local geographic setting, amongst other variables. In this study, we present new results on the concentration and composition of PACs in five sediment cores from the Cameron Hills region of the southern Northwest Territories, where until recently, a small, conventional oil and gas extraction facility operated. Similar conventional operations are common throughout western Canada, and contamination from these small, but numerous, installations may be an underestimated source to the ecosystem, despite their small footprints. In this study we use previously published research from lakes near to unconventional extraction operations in Alberta, and exploration wells in the Mackenzie Delta to place the Cameron Hills into a broad range of potential contamination in western Canada. In addition, the new analyses and comparisons presented here characterize the range of variability that exists in the magnitude and composition of altered PAC depositional histories inferred from sediment cores in areas of petroleum development in western Canada. This represents the first comprehensive comparison of temporal trends in PACs between major methods of oil and gas development in this economically important region of Canada, and provides an important context for assessing the role of future resource extraction as a source of environmental contaminants.

2. Materials and methods

2.1. Description of study regions

2.1.1. Cameron Hills, Northwest Territories

The Cameron Hills, located in the southern Northwest Territories near the border with Alberta (Fig. 1), is the location of a small conventional oil and gas installation, employing pumpjack extraction technology. A previous study that measured PAC burdens in stream benthic macroinvertebrates in the Cameron Hills region found that, although PAC concentrations were typically similar between upstream and downstream sites, there was evidence of elevated alkyl naphthalenes in two downstream locations (Korosi et al., 2016b). Sediment cores were collected from five lakes in the Cameron Hills region to explore the potential contribution of small, conventional hydrocarbon developments to the environmental burden of nearby aquatic ecosystems. Lakes CamH1 (60.05°N, 117.51°W), CamH3 (60.14°N, 117.57°W) and CamH5 (60.03°N, 117.48°W) (Korosi et al., 2016b) had at least one working or capped conventional gas or oil well within their catchments at the time of sampling. Two additional lakes (CamH2 (60.22°, 117.80°W) and CamH7 (60.14°N, 117.38°W); unofficial names, Korosi et al., 2016b) without oil and gas installations were selected as reference lakes. Sediment cores were collected from the central basins of each lake in September 2012 using a Glew gravity corer (Glew, 1989), sectioned at 0.5 cm intervals in the field (Glew, 1988) and kept frozen until analysis, following laboratory methods described below.

2.1.2. Athabasca oil sands region, Alberta

Kurek et al. (2013a) examined the depositional history of PACs in sediment records of several small, headwater lakes located within ~35 km of site AR6 in the Athabasca oil sands region (AOSR). The area around AR6, often marked as the heart of the upgrading and surface mining activities in the AOSR (Kelly et al., 2009), is an area of intensive extraction, as well as the location of two major bitumen upgrader facilities (Syncrude and Suncor operations), and close to a third upgrader to the north (CNRL Horizon). Potential sources of PACs in the AOSR included fossil fuel emissions from upgraders, refineries and vehicles. In addition, exposed bitumen layers can contribute petrogenic PACs from dust, as can petroleum coke (petcoke) storage piles disturbed and distributed by wind. Current