The Impacts of Petrochemical Activity and Climate Change on Polycyclic Aromatic Hydrocarbon (PAH) Deposition to Lake Sediments of Northwestern Canada

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Abstract

With the rising demand for fossil fuels, Northern Canada has seen an unprecedented increase in petrochemical development. These developments are often associated with emissions of PAHs, a group of hydrophobic organic contaminants that are known to be carcinogenic and otherwise harmful to humans. Due to their hydrophobic nature, PAHs tend to bind to organic matter and can be produced through both anthropogenic and natural processes, making them ubiquitous in the environment. Therefore, in addition to impacts from petrochemical developments, changes to climatic conditions, such as increased forest fire disturbance and primary production also have the potential to alter delivery of organic carbon (OC) and PAHs to ecosystems. However, very little is known as to how the combined stressors of climate change and petrochemical development may affect environmental deposition of these contaminants. The concentrations and composition of parent and alkyl PAHs were analysed in radiometrically-dated sediment cores from lakes with one of four different types of petrochemical development in their catchments: (1) *in-situ* oil sands extraction (Cold Lake, AB); (2) open-pit oil sands extraction (Fort McMurray, AB); (3) abandoned conventional natural gas exploration (Mackenzie Delta Uplands, NWT); and (4) conventional gas and oil extraction (Cameron Hills, NWT). PAH deposition to lake sediments was also compared to climate reconstructions using climate proxies (diatom assemblages, inferred chlorophyll *a* and its diagenetic products, and Rock Eval carbon fractions as well as %OC). PAH sources were differentiated between potential pyrogenic and petrogenic origin over a period that extends to pre-industrial times using ratios of specific PAHs that can be traced to their potential source. Sediment cores from Cold Lake, AB showed concentrations of the sum of alkyl PAHs greater than those of parent PAHs, while all other cores show the reverse trend. A comparison of the % change of PAH concentrations from pre-development to post-development sediments between the four regions, showed that the greatest increase in concentrations of PAHs occurred in the Athabasca oil sands region. PAH profiles in the conventional regions have been historically dominated by mixed sources (pyrogenic PAHs from general background atmospheric inputs and petrogenic PAHs from the surrounding hydrocarbon-rich soils). While cores from the Fort McMurray area show a clear shift from pyrogenic sources (primarily wood and coal burning) in earlier sediments to petrogenic sources in more modern sediments, and the Cold Lake cores show some shifting sources to

those dominated by pyrogenic sources in modern sediments. Organic carbon was significantly correlated with the sum of parent PAHs in 2 out of the 6 NWT cores that were examined for climate change impacts, while all other PAH parameters (concentration and composition) do not correlate significantly with any of the climate proxies. Establishing background concentrations and sources of PAHs in aquatic ecosystems is essential for understanding the natural environmental variations in these contaminants. Moreover, as both petrochemical activity and impacts from climate change are predicted to intensify in the future, studies such as this one allow us to build a solid understanding of how PAH deposition to northern lakes has responded to the warming climate and whether PAHs have been altered as a result of petrochemical activity.

Résumé

Dernièrement, le climat a été soumis à des transformations qui à leurs tours ont causé des changements au niveau des écosystèmes. L'augmentation de la sévérité des impacts causés est anticipée, surtout dans les régions du nord. Certains de ces changements, tels que l'augmentation de la productivité primaire et du nombre de feux de forêts, ont le potentiel de modifier la source, la quantité et la circulation dans l'environnement du carbone organique et des contaminants organiques hydrophobes, ces derniers préférant se lier au carbone organique. En raison de sa richesse en hydrocarbures, le nord du Canada connait aussi un changement en termes d'utilisation du sol, spécifique au développement des ressources pétrolières souvent associé à l'émission d'hydrocarbures aromatiques polycycliques (HAPs). Les HAPs sont des contaminants organiques hydrophobes potentiellement cancérogènes aux humains. D'origine naturelle ou anthropique, ils sont omniprésents dans l'environnement, et les impacts du changement climatique et du développement des ressources pétrolières sur le dépôt des HAPs dans les sédiments des lacs sont mal connus. Des carottes de sédiments provenant de lacs choisis spécifiquement selon le type de développement pétrolier ayant lieu à proximité ont été datées de façon radiométrique, et les concentrations et la composition des HAPs ont été quantifiées et qualifiées. Les résultats ont été comparés à l'historique du développement pétrolier de chaque région. Les impacts de quatre types de développements pétroliers différents ont été étudiés, incluant (1) l'extraction *in-situ* des sables bitumineux (Cold Lake, AB) ; (2) l'extraction minière de surface (Forth McMurray, AB) ; (3) la prospection abandonnée de gaz naturel conventionnel (Mackenzie Delta Uplands, TNW) ; (4) l'extraction conventionnelle de ressources pétrolières (Cameron Hills, TNW). De plus, la variation des dépôts des HAPs dans les lacs du TNW a été comparée à la variation des changements climatiques, ces derniers reconstruits à l'aide d'indicateurs comme les colonies de diatomées, la chlorophylle-*a* déduite et ses produits de diagnèse, le pourcentage de carbone organique et sa fraction produite par les algues. En utilisant des ratios de composés spécifiques, les sources des HAPs ont été caractérisées selon leur origine (pyrogénique ou pétrogénique), et ce, sur une période allant de l'ère préindustrielle au présent. Les échantillons de sédiments de Cold Lake, AB ont révélés des concentrations élevées de HAPs alkylés en comparaison aux HAPs non-substituées, alors que les autres carottes ont indiqué une tendance inverse. Une comparaison des taux de changement des HAPs entre les

sédiments pré-développement et post-développement des quatre régions étudiées a démontré une grande augmentation de contaminants dans les sables bitumineux d'Athabasca. Historiquement, les régions à proximité de l'extraction conventionnelle ont été dominées par des HAPs de sources mixtes telles que les HAPs pyrogènes atmosphériques généraux et les HAPs pétrogéniques des sols riches en hydrocarbures. Alors que les carottes de sédiments de Fort McMurray ont indiquées un changement évident des sources pyrogènes (principalement la combustion de charbon et de bois) dans les sédiments plus anciens vers des sources petrogéniques dans les sédiments plus récents, les carottes de sédiments de Cold Lake ont indiquées que les sédiments modernes sont dominés par des sources pyrogènes d'HAPs. Aucune corrélation entre la concentration et composition des HAPs ne s'est avérée significative, excepté entre les concentrations des HAPs non-substituées et le carbone organique (%CO) pour 2 des 6 lacs du TNW, pour lesquels on a étudié les impacts des changements climatiques. Établir le contexte des concentrations et des sources d'HAPs générales et globales dans les écosystèmes aquatiques est essentiel pour comprendre la variation naturelle de ces contaminants dans l'environnement. Étant donné qu'une intensification de l'activité pétrolière et des impacts du changement climatique est attendue, ce type d'étude est nécessaire à la compréhension des conséquences du changement climatique sur les dépôts d'HAPs dans les lacs du nord, surtout lors de la gestion des risques que pose l'activité pétrolière.

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Cyndy Desjardins
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Glossary

AOS	Athabasca oil sands, AB
ASE	Accelerated solvent extraction
Bitumen	heavy oil
CAM	Cameron Hills, NWT
CAPP	Canadian Association of Petroleum producers
CCME	Canadian Council of Ministers of the Environment
CL	Cold Lake, AB
CIMP	Cumulative Impact Monitoring Program
GC-MS	Gas chromatography coupled to a mass spectrometer
HMW	high molecular weight
In-situ	Extraction of oil sands using steam to liquefy bitumen at depth
IPCC	International Panel on Climate Change
ISQG	Interim sediment quality guidelines
LMW	low molecular weight
MDU	Mackenzie Delta Uplands, NWT
NG sump	Dumping pit for waste from exploration for natural gas
NWT	Northwest Territories
OC	organic carbon
OG pump	Extraction of oil and gas by jack pump
Open-pit	Extraction of oil sands using open pit mines at the surface
PCA	principle components analysis
PCB	Polychlorinated Biphenyl
SOA	secondary organic aerosol
TOC	Total organic carbon
USEPA	United States Environmental Protection Agency

Chemical Glossary by increasing molecular weight

PAH	Polycyclic aromatic hydrocarbons
NAPH	Naphthalene
ACE	Acenaphthene
ACY	Acenaphthylene
FLU	Fluorene
ANT	Anthracene
PHE	Phenanthrene
FLA	Fluoranthene
PYR	Pyrene
BaANT	Benz[a]anthracene
CHR	Chrysene
BbFLA	Benzo(b)fluoranthene
BkFLA	Benzo(k)fluoranthene
BaPYR	Benzo[a]pyrene
IcdP	Indeno(1,2,3-cd)pyrene
DahA	Dibenz(a,h)anthracene
BghiP	Benzo(g,h,i)perylene
C1-Parent	The specified parent PAH with 1 alkyl group
C2-Parent	The specified parent PAH with 2 alkyl groups
C3-Parent	The specified parent PAH with 3 alkyl groups
C4-Parent	The specified parent PAH with 4 alkyl groups

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CHAPTER 1. GENERAL INTRODUCTION

1.1. Introduction

1.1.1. Multiple Stressors to Aquatic Ecosystems

The global population is growing at an increasing rate, and with modern civilization's demands for fuel, petrochemical resource development has expanded to reflect this growth. In addition, the last hundred years have witnessed significant climate change, widely acknowledged to be exacerbated by human activities. These combined stressors have the potential to severely alter ecosystems across the world. Of particular concern is what impact these multiple stressors may have on aquatic ecosystems and thus freshwater resources. In 2009 domestic consumption of water in Canada was about 274 litres of water per day per capita, one of the largest amounts of water consumed per capita in the world (Environment Canada 2015). In addition, Canada holds about 7% of the world's renewable freshwater supply (Environment Canada 2015), a disproportionally large amount compared to the size of its population (only about 0.5% of the global population) (Statistics Canada 2015). The country's many freshwater bodies cover an area of almost 900,000 km^2 (Environment Canada 2015), much of which resides in the northern part of Canada, which is currently undergoing major climatic and economic change.

1.1.2. Climate Change

Although all regions of the world are expected to experience climate change to varying degrees, climate models consistently predict the greatest impacts will occur in the northern regions of the world, a process known as polar amplification. Over the period from 1948 to 2010 Canada experienced a greater rate of warming than most other countries of the world, with the national average annual temperature increasing 1.6˚C (Government of Canada 2015), while the increase in temperature in the Arctic has been twice the global annual average since 1980 (AMAP 2011). Paleoecological reconstructions using lake sediment cores and other natural archives have shown that shorter winters in the north have altered lake benthic invertebrate communities, shifting

their dominance from benthic, pennate organisms such as *Fragilaria* and *Navicula*, to more centric and planktonic genera such as *Cyclotella* (e.g. Thienpont et al. 2013b), indicating enhanced thermal stratification in lakes. Additionally, by decreasing ice-coverage and increasing the algal growing season, shorter winters have stimulated increases in primary production in Arctic regions, indicated by rising levels of inferred chlorophyll-*a* and its diagenetic products in lakes (Thienpont et al. 2013a, Rühland et al. 2013, Michelutti et al. 2010). Increases in organic carbon are expected from the expansion in primary production, and could in turn alter the transportation and fate of hydrophobic organic contaminants that preferentially bind to organic matter, such as polycyclic aromatic hydrocarbons, which have been increasing in the environment as a result of human activities (Kurek et al. 2013, Kelly et al. 2009, Sojinu et al. 2010).

1.1.3. Petrochemical Development

In addition to stress from changing climatic conditions, aquatic ecosystems are under stress from anthropogenic land-use change. One such change is the development of petrochemical resources. Beginning in the early 20th century, energy and fuel requirements of an increasingly industrialized global civilization were being satisfied more and more by oil and gas over coal, leading to greater petrochemical resource development. In Canada, especially in the remote northern regions of the country, the past five decades have seen a tremendous boom in petrochemical development. Canada has the third largest oil reserve in the world, close to 97% of which is in the unconventional resources of the Alberta oil sands (CAPP 2014). The oil and gas industry is the single largest private investor in Canada, accounting for fully 20% of Canada's largest stock exchange (the Toronto Stock Exchange), and is responsible for 550, 000 jobs nationally (CAPP 2014). The three major oil sands deposits that make up the Alberta oil sands (the McMurray, Peace River and Cold Lake deposits) are overlain by 140,000 km^2 of land (Energy Alberta 2015), an area larger than the total landmass of Greece. Concerns have been raised over the impact this scale of land-use change, and the potential contamination that comes with industrial development, will have on ecosystems near petrochemical developments, especially aquatic ecosystems and thus freshwater resources. In particular, petrochemical developments are associated with the release of polycyclic aromatic hydrocarbons (PAHs) into the environment, compounds that are

known to be carcinogenic, mutagenic and can cause general disruption to biological function (e.g. Baird et al. 2004, Boffetta et al. 1997, Camus et al. 2003, Carls et al. 2010). Establishing environmental baselines for these chemicals and determining how petrochemical development may be affecting the environmental inventory of PAHs remains a research priority.

1.2. Polycyclic aromatic hydrocarbons (PAHs)

1.2.1. General Description of PAH chemistry and formation

PAHs are a large group of organic contaminants comprising two or more aromatic rings, 16 of which are listed by the United States Environmental Protection Agency (USEPA) as Priority Pollutants. They have been detected in many environmental compartments, including lake and river sediments, animal tissue, soil, air and water, and can bioaccumulate in animal tissue (Thienpont et al. 2013a, Hall et al. 2012, Gabos et al. 2001, Galarneau et al. 2014, Amdany et al. 2014, De Laender et al. 2011). For example, one study (De Laender et al. 2011) analysed several organic contaminants in Arctic species from different trophic levels, including blue mussels (benthic, sedentary filter feeders), arctic cod (who lead a pelagic lifestyle feeding mainly on pelagic invertebrate prey) and polar bears (top predators). During the period from 1985 to 2009, the body burden of PAHs in these animals were found to have increased, in some cases up to 30-fold, while the body burden of all other organic contaminants included in the study (PCBs, HCHs, DDTs and HCB) were stable or in decline during the same period (De Laender et al. 2011). The authors suggested that this trend largely reflects efforts to regulate the use of PCBs, HCHs, DDTs and HCB. Being exclusively anthropogenic, concentrations of these contaminants have subsequently declined since the 1970's when many industrialized countries legislated for severe limits on their use. PAHs on the other hand have both natural and anthropogenic sources (most importantly as the by-product of fuel combustion), so emissions continue and are increasing in some food webs (De Laender et al. 2011).

Many PAHs and their homologues are known to be mutagenic, carcinogenic or otherwise disruptive to the general physiology of humans and other organisms (Baird et

al. 2004, Boffetta et al. 1997). For example, Carls et al. (2010) found that pink salmon embryos (*Oncorhynchus gorbuscha*) exposed to PAHs from a crude oil spill experience several detrimental physiological effects including delayed development, edema, anemia and increased mortality, compared to unexposed embryos. As such, improving our understanding of the magnitude of PAH deposition and the pathways of PAHs to the environment has become an important area of research.

PAHs are generally formed during incomplete combustion of any organic material, making them ubiquitous in the environment. PAHs can be further classified as pyrogenic or petrogenic depending on sources, which are linked to the conditions at the time of their formation. Pyrogenic PAHs form during relatively high temperature combustion at atmospheric pressures. When combustion commences, the organic material is converted into smaller molecular fragments that are extremely unstable (i.e. free radicals), a process sometimes called cracking. The free radicals formed as a result of cracking then react with each other establishing the first aromatic ring, which then goes on to react with very small hydrocarbon molecules through a number of chemical pathways, not all of which are presently or entirely understood. Through these pathways, however, the original aromatic ring becomes a multi-ring system that is much more stable than its precursors. As combustion proceeds all the way to soot production, with lighter PAHs reacting to form larger heavier PAHs, the heavier PAHs eventually act as organic precursors to the production of the soot particles themselves. Indeed, early work by Prado and Lahaye (1982) demonstrated that as the production of soot particles increased, the amount of PAHs decreased over time.

Petrogenic PAHs are formed over a geological timescale under lower temperature geothermal conditions, such as those during petrogenesis. These conditions favour the formation of substituted, or alkylated, PAHs, and are found in the petrochemicals (i.e. the oil and gas) themselves. They are typically, although not exclusively, emitted from point sources during events such as ground seeps or oil spills. In contrast, pyrogenic PAHs usually form the unsubstituted, or parent, PAHs and are typically emitted from more diffuse sources such as forest fires or traffic emissions. PAHs in surface sediments can also be formed during digenesis by microorganisms from biological precursors, but are generally considered to be highly localized sources, and are therefore thought not to

contribute a significant amount of PAHs to the general inventory in most aquatic systems (Lima et al. 2005).

In urban centres most PAHs are anthropogenically derived from domestic energy production, industrial emissions, and most importantly from vehicle emissions (Lima et al. 2005). PAHs in more remote areas usually come from a mix of sources including long-range transport (LRT) from urban and industrial areas and from forest fires. For example, studies showed that PAHs deposited in snow and ice in remote alpine regions in the Alps faithfully reflected the different phases and general pace of post-17th century industrialization across Europe (Gabrieli et al. 2010). General global trends have showed increases in PAHs continued until the 1970s then began to decrease subsequent to this, due to increased restrictions on industrial emissions as well as improvements in technology leading to more efficient internal combustion engines in automobiles (Lima et al. 2005). More recently it has been suggested that global PAH concentrations have begun increasing in the past decade, however this time due to general global population growth and the increasing industrialization of economies that historically used very little fossil fuels (Lima et al. 2005).

1.2.2. PAH Behaviour in Aquatic Ecosystems

PAHs enter aquatic environments through wet/dry deposition, or through run-off from the surrounding land. Deposition occurs through adsorption of PAHs in the gas phase to particles on the water's surface or to atmospheric particles that are then deposited to the water column. PAHs in the gas phase can also cross the air-surface water interface and be subsequently dissolved in the water column. They can also be deposited through precipitation laced with PAHs. Once in the water column, PAHs can be transformed or degraded through several different chemical and biological processes, including photooxidation, biotransformation and biodegradation. The half-lives of the PAHs undergoing these different processes largely depends on the physical structure of the PAH (i.e. the number of rings, the degree of linearity, the number of alkyl structures attached to the ring system). For example, the USEPA (1976) found that the photooxidation half-life of anthracene (a compound with 3 benzene rings) in water is 0.1-4.4 years, while benzo(a)pyrene, a much bulkier PAH with 5 benzene rings, has a

photooxidation half-life in water ranging from 8.6 days to just over a year, depending on environmental conditions (Smith et al. 1978). However, volatilization from water has the opposite trend related to the size and weight of the compound; a compound with three benzene rings, such as anthracene will volatilize quicker from water (about 17 hours), than a four-ringed structure such as pyrene (115 hours to 3.2 years) (Southworth 1979). In addition, the transformation and degradation processes will depend in no small part on whether the PAH is sorbed to particulate matter and what the composition of the particle is. For example, Zelenyuk et al. (2013) observed in a controlled lab experiment that the presence of PAHs in the gas-phase at the time of formation of secondary organic aerosols (SOA) actually causes the PAHs to be incorporated into the SOA, prolonging the evaporation time of the SOA and protecting the PAH from photo and chemical oxidation in the atmosphere, potentially allowing particulate-bound PAHs to travel further before being deposited to the water column.

PAHs can be found in many environmental compartments (air, water, soil, vegetation, sediments) (Thienpont et al. 2013a, Hall et al. 2012, Camus et al. 2003, Carls et al. 2010, Sojinu et al. 2010, Galarneau et al. 2014), but the ultimate fate of PAHs in aquatic ecosystems is largely considered to be lake and river sediments. Being hydrophobic and lipophilic to varying degrees, it is generally assumed that the transportation and fate of PAHs are influenced by the amount of organic carbon in a given system. General models (MacKay and Paterson 1991) have suggested that PAH burial in sediments should increase with the increases in organic carbon that are expected in aquatic ecosystems, especially in northern lakes, as a result of increased primary production from a warming climate. In an effort to better understand the transport mechanism during runoff events that connects PAHs from the surrounding land to deposition in aquatic ecosystems, Zheng et al. (2013) conducted a controlled experiment in which they collected runoff from PAH-laced soil being bathed by simulated rainfall of clean water. The soils they used contained similar amounts of clay, black carbon and PAHs, while total organic carbon (TOC) in one soil was double that found in the other. Enrichment ratios of the clean sediment in the runoff collection vessel suggested that organic carbon, and not clay particles, seems to be the primary vehicle of PAHs from soil to sediment during runoff events, although these findings have yet to be verified in field

studies. Jeremiason et al. (1999) conducted a field study on the impacts of increased autochthonous carbon (phytoplankton-derived) to PAH accumulation in sediments at Lake 227 in the Experimental Lakes Area in northwestern Ontario, which has a well-documented history of eutrophication. Sediment mass-accumulation almost doubled while the flux of organic carbon (OC) to the sediments increased 3-fold over the history of the sediment core. However, while PAH levels initially spiked post-P-addition, they subsequently declined and continued to do so towards the surface of the core, despite the general increase in OC. The authors suggested that, in addition to the high efficiency of carbon recycling in the lake, the initial spike in PAHs and their subsequent general decline to below background levels in sediments post-eutrophication are likely due to shifts in phytoplankton. In more recent times, the sediments are dominated by a phytoplankton community structure that is abundant in species that have lower bioconcentration factors, thus fewer PAHs are scavenged from the water column and subsequently deposited to the sediments (Jeremiason et al. 1999).

1.2.3. Sourcing PAHs

The ability to establish baseline concentrations of PAHs, as well as distinguish natural sources in aquatic ecosystems is vital if we are to understand the impact that petrochemical development and climate warming may be having on PAH deposition. Due to the paucity of direct measurements of PAH levels in parts of northwestern Canada, an effective way to track PAHs through space and time is to analyse dated lake sediment cores. The success of chronological reconstructions of the past physical, chemical and biological conditions of lakes using sediment cores has been well documented (Michelutti et al. 2010, Thienpont et al. 2013a, Jeremiason et al. 1999), including applications for tracking PAHs through time (Kurek et al. 2013, Jautzy et al 2013). As previously mentioned, sediments are very stable sinks for PAHs, which generally resist migration in sediments. Consequently, PAH concentrations in sediment core records faithfully archive historical trends in PAH deposition in aquatic environments.

PAHs are emitted as complex mixtures, the composition of which is known to be linked to their source, therefore an effective method for sourcing PAHs is to examine the abundance of certain PAH compounds relative to each other, known as diagnostic ratios

(Tobiszewski & Namiesnik 2012). In order to account for some of the disparity in behaviour of the different PAH compounds due to structural differences, diagnostic ratios are often made up of isomeric pairs of PAHs (i.e. compounds having the same molecular weight but different configuration of atoms). These ratios are not absolutes, and should be used as guidelines only, as some compounds, despite being isomeric pairs, have been shown to degrade differently under certain conditions. However, when evaluated together with historical records of oil and gas development, they can be useful when trying to interpret general trends in sources of PAHs over time. For this reason diagnostic ratios are a good tool to assess how petrochemical developments may affect PAH deposition in their vicinity.

1.3. Study Regions

Prime examples of major petrochemical developments in Canada include the gas fields of the Mackenzie Delta, the oil and gas of the southern Northwest Territories, and the unconventional resources of the Alberta Oil Sands, which currently produce 1.9 million barrels per day (CAPP 2014).

1.3.1. Mackenzie Delta, NWT

The Mackenzie Delta uplands, located in the Taiga Plains region of the Northwest Territories in Canada, are experiencing a period of rapid economic development and climate change. Natural gas exploration began in the early 1960's, and over 150 drilling mud sumps have been dug since that time. These drilling sumps are essentially dumping pits for the disposal of drilling fluids, cuttings and other waste produced during the drilling of the exploratory borehole, which are then backfilled, capped with excavated material, and abandoned once full or exploration is complete. The Mackenzie Delta drains one of the largest watersheds in North America and is an important area for hunting and fishing by the indigenous Gwich'in and Inuvialuit peoples. As such, the consequences of the exploration for and exploitation of petrochemicals on the ecosystems in this region have been called in to question. A report by Kannigan and Kokelj (2010) has shown that the practice of dumping and capping drilling waste into sumps is no longer a viable option for drilling waste containment and storage as the surrounding

permafrost, meant to be a permanent solution for containment, has become unstable and is thawing as a result of climate change. A recent study by Thienpont et al. (2013a) found that these compromised drilling sumps are in fact leaking into nearby lakes and ponds, causing elevated chloride levels (a major component of drilling fluids) compared to nearby reference lakes not impacted by sump-failure. The study showed shifts in invertebrate communities to ones dominated by saline-tolerant species consistent with the timing of natural gas exploration in the area and construction of the sumps. In addition to warming-mediated thawing permafrost, lakes in this area have seen increasing duration of ice-free periods, indicated by changes in benthic assemblages from those dominated by benthic cold-tolerant species to ones dominated by planktonic species and increases in primary production beginning in the second half of the 19th century or first half of the 20th century (Thienpont et al. 2013a, 2013b). These changes can in turn increase the pool of organic matter (OM) and impact the transportation and fate of hydrophobic organic contaminants, such as PAHs, that preferentially bind to OM. Although impacts from petrochemical development and changing climatic conditions to biological communities (including increases in primary production) has been explored in this region, virtually nothing is known about how these two major stressors may be affecting the amount and sources of PAHs deposited to aquatic ecosystems in the area.

1.3.2. Cameron Hills, NWT

South of Great Slave Lake in the Northwest Territories, near the territorial border with Alberta, the Cameron Hills lie in the south-central Taiga Plains in the mid-boreal region and are underlain by Cretaceous marine shale with occasional pockets of glacial-fluvial materials. Petrochemical development in this area is generally less intensive than that in the Mackenzie Delta regions, and has occurred more recently, beginning in the 1970s and 80s. Deposits here are termed conventional deposits, which are near-liquid forms and are therefore usually brought to the surface with jack pumps. The oil and gas products are then pumped through pipelines to a processing plant near Bistcho Lake in Alberta, to be upgraded and distributed. To the best of our knowledge, no study of PAHs in the Cameron Hills, in the river or surrounding lakes, has been published in a peer-reviewed journal.

1.3.3. Athabasca Oil Sands, AB

Oil sands are a different type of bitumen deposit compared to the conventional deposits described above, and are found in Alberta as roughly 10-12% bitumen, ~83-85% sand and ~4-5% water (Wang et al. 2014). A large proportion of the oil sands found in Alberta are in the McMurray Formation, which covers about 46,000 km^2, and can be found at depths of up to a couple hundred meters. Much of the research done on the impacts of petrochemical development on PAH deposition in Canada has focused on the Athabasca region north of Fort McMurray, where the oil sands are within 100m of the surface (Alberta Innovates 2011) and are thus surface-minable through large open-pit mines. For example, Kurek et al. (2013) found that concentrations of PAHs in dated lake sediment cores began to increase shortly after open pit mining began in the mid 1960's, and continued in a rising trend towards the surface of the cores. In fact, in the lakes closest to the open-pit mining PAH levels exceeded interim sediment quality guidelines (CCME 1999) for several compounds. Additionally, using diagnostic ratios of PAHs, they were able to detect a shift from pyrogenic sources of PAHs (likely from wood and coal burning) in pre-development sediments to a more petrogenic signal in surface sediments. Similarly, Jautzy et al. (2013) found significant increases in PAHs and shifting profiles of pyrogenic to petrogenic sources in sediment cores from the same region. Both studies attribute the bulk of these profiles to the scattering of bitumen particles through wind action as the oil sands are scooped into dump trucks to be transported off-site, for separation, processing and distribution.

1.3.4. Cold Lake, AB

Once the oil sands deposits are below about 100 m from the surface, such as the deposits found in the Cold Lake region of Alberta, open-pit mining is no longer a feasible method for extracting the oil sands. Instead, a method using steam assisted gravity drainage (SAGD) is employed. Steam is typically pumped down to the deposit to separate the bitumen and oil sands *in-situ*, and to liquefy the bitumen slightly, which is then pumped to the surface and transported off-site for upgrading and processing. Comparatively few studies have looked at the impacts of *in-situ* oil sands extraction,

despite the fact that 80% of the known oil sands reserves in Alberta are not within the reach of open-pit mining and thus require *in-situ* methods for extracting the bitumen. Korosi et al. (2013) looked at PAH profiles in dated sediment cores from two lakes near an oil field near Cold Lake, AB in order to assess the temporal pattern of PAH deposition in the area. In addition, they examined surface soil and pine needle samples in order to investigate the spatial variation in PAH deposition in the area. Their analysis showed that concentrations in some lakes near *in-situ* operations are increasing, and that the timing of the increase is consistent with the history of oil sands extraction in the area. Sources for increased PAHs in the region were not investigated, however, which is a knowledge gap that is addressed in this thesis. Furthermore, it is unclear how the impacts of oil sands extraction compare to that of conventional oil and gas development on PAH deposition trends.

1.4. Thesis Objectives

The main goal of this thesis is to reconstruct PAH deposition histories to areas in the Northwest Territories (NWT) where little is known about PAH deposition, and to compare them to more extensively studied areas south of the NWT in Alberta. My aim was to conduct both spatial and temporal analyses on PAH depositional patterns in different parts of northwestern Canada in order to assess the impact that the combined stressors of petrochemical development and climate change may be having on the sources and fate of PAH emissions into aquatic ecosystems. In addition I was able to identify key compounds of interest in each region that may be used as chemical markers in future efforts to monitor or regulate PAH emissions from petrochemical developments in these areas.

Chapter 2 compares the historical PAH concentration profiles across lakes near four major types of petrochemical developments in northwestern Canada to see whether the amount of PAHs deposited to the environment has been exacerbated by petrochemical development, and whether the scale of impact to PAH concentrations is the same across all four types of developments. Since industrial activity is known to be a source of PAHs, I also looked at changes in the composition profiles across the four regions that would indicate shifting sources of PAHs over time to see if petrochemical developments may be

affecting PAH sources, and whether the different types of developments may be affecting the sources of PAHs differently

Chapter 3 examines historical PAH concentration profiles from two regions in the NWT that have experienced different climate change scenarios, against well known climate proxies for lake sediments, including inferred chlorophyll-*a* and diatom community assemblages, to assess whether an increase in organic carbon (through warming induced increases in primary production) and other warming-mediated shifts in aquatic ecosystems may be increasing PAH burial in sediments as general models might suggest (Mackay and Paterson 1991). In addition, climate change may affect the conditions under which PAHs deposited to the environment are transported to aquatic systems (e.g. increased precipitation and runoff from the surrounding land), or degraded within aquatic ecosystems (e.g. increasing chances of photooxidation through an increase in ice-free periods). Previous studies have suggested that the behaviour of individual PAHs throughout these processes will depend in large part on their structures, and thus are likely to be degraded or transported differently under different conditions, leading to shifts in PAH composition in sediments over time. As such, PAH composition is compared to climate proxies to see if changing conditions in northern lakes may be affecting the composition of PAHs being deposited to sediments. These combined analyses allowed me to characterize general PAH-climate interactions that are as yet largely unknown.

Chapter 4 summarizes general conclusions, and suggests future directions.

CHAPTER 2. Comparative histories of polycyclic aromatic hydrocarbons in lakes near petrochemical activity in northwestern Canada

2.1. Abstract

Radiometrically-dated lake sediment cores were analyzed for both unsubstituted (parent) and substituted (alkyl) polycyclic aromatic hydrocarbons (PAHs) to compare deposition histories near four major types of petrochemical developments: (1) open-pit oil sands extraction (Athabasca oil sands, AB); (2) *in-situ* oil sands extraction (Cold Lake, AB); (3) abandoned conventional natural gas mud-sumps (Mackenzie Delta uplands, NWT) and (4) pump-jack extraction of conventional oil and gas sources (Cameron Hills, NWT). The sediment cores taken near open-pit oil sands mining indicate a shift from pyrogenic sources (primarily wood and coal burning) in pre-development sediments to more petrogenically sourced PAHs in modern sediments, as well as large increases in concentrations of PAHs that are consistent with the onset of open-pit mining in the area. Cores near *in-situ* oil sand extraction operations showed evidence of increasing concentrations and shifting sources, while all conventional sites showed no clear changes in either sources or concentrations coincident with the timing of petrochemical development in these regions. These results provide the first regional comparison of PAH profiles near key types of petrochemical developments in northwestern Canada, and will assist in quantifying and characterizing emissions from this major industry.

2.2. Introduction

Increasing global population growth and the slow shift to renewable energy sources has kept the demand for fossil fuels high. Petrochemical companies have responded by increasing their production efforts to meet this demand. Canada has the third largest proven oil reserve in the world, with over 97% of its crude oil reserves tied up in the unconventional resources of the Alberta oil sands (CAPP 2014), and is the fifth largest natural gas producer in the world (USEIA 2014). Projections suggest that, while growth in conventional petrochemical developments is stabilizing, the unconventional developments will see a 2.5 fold increase in daily production from 2013 to 2030 (CAPP

2014). Furthermore, over 90% of the current production and future growth in Canadian petrochemical industries is in the western part of the country (CAPP 2014). Notable examples include the gas fields of the Mackenzie Delta, the oil and gas of the southern Northwest Territories, and the unconventional resources of the Alberta Oil Sands, which currently produce 1.9 million barrels per day (CAPP 2014). One of the largest public concerns regarding the rapid growth in petrochemical developments is the potential for contamination of aquatic ecosystems that provide freshwater resources to nearby communities. In particular, oil and gas developments are often associated with the release of polycyclic aromatic hydrocarbons (PAHs), a class of organic contaminants that can be mutagenic, carcinogenic or otherwise disruptive to humans and other organisms (Baird et al. 2004, Boffetta et al. 1997, Camus et al. 2003)

2.2.1. General Description of PAHs

PAHs are a large group of organic contaminants that contain two or more fused aromatic rings, and can enter the environment through both natural processes and anthropogenic activities, making them ubiquitous in the environment. In water they are generally hydrophobic, thus they adsorb to particulate matter. PAHs typically enter aquatic environments through precipitation or dry deposition directly to the water column, or potentially through run-off from the surrounding land, and, for the most part, settle in sediments thereafter. PAHs have been detected in many environmental compartments, including lake and river sediments, animal tissue, soil, air and water (Thienpont et al. 2013a, Hall et al. 2012, Camus et al. 2003, Carls et al. 2010, Sojinu et al. 2010, Galarneau et al. 2014) and can bioaccumulate in animal tissue (De Laender et al. 2011). Improving our understanding of the magnitude of PAH deposition and the pathways of PAHs to the environment is a research priority.

2.2.2. Study Objectives

The aim of this study was to address two main research objectives: (1) to determine whether parent and alkyl PAH concentrations have changed since the onset of oil and gas development in northwestern Canada, and compare the impact between the four methods of development (use of natural gas sumps for drilling waste containment,

pump-jacks for conventional oil and gas extraction, *in-situ* extraction of oil sands using high-pressure steam injection and open-pit mining of oil sands.); and (2) to determine whether a shift in PAH composition has occurred since the onset of petrochemical development that would indicate a major change in PAH source. We accomplish these objectives by measuring and comparing PAH deposition histories in dated lake sediment cores from regions in northwestern Canada that employ different methods of extracting oil and gas resources, including conventional and unconventional developments. In addition we assess changes in PAH sources using diagnostic ratios and identify PAH compounds that contribute most to the different regions using multivariate indirect ordinations and an analysis of the relative abundance of individual PAH compounds across the different regions.

2.2.3. PAH formation and chemistry

PAHs are generally formed during the incomplete combustion of organic material and can be broadly categorized as pyrogenic or petrogenic in origin. Pyrogenic PAHs come from diffuse sources as a result of the burning of biomass, including fossil fuels, favouring the formation of unsubstituted (also known as "parent") PAHs. Petrogenic PAHs are usually emitted from point sources (e.g. oil spills, ground seeps) and generally form under geothermal conditions, over geological time scales that favour the formation of alkylated compounds. PAHs are emitted to the environment as mixtures and the PAH composition of an environmental sample is considered to be reflective of its predominant sources. One method for determining major sources of PAHs to the environment is the examination of the abundance of certain PAH compounds relative to each other, known as diagnostic ratios. In order to account for the disparity in chemical and physical behaviour due to structural differences, ratios are often made up of PAH isomers thus have similar chemical behaviour, such as flouranthene (FLA) and pyrene (PYR) (molar mass=202.25 g) (CCME 1999). They can also be made up of a parent PAH and its alkylated counterparts, for example fluorene (FLU) and the alkylated fluorenes.

2.2.4. Sourcing PAHs

Canada's diverse petrochemical resources will have different background PAH signatures that can be detected in environmental samples. In addition they require different scales and methods of exploration and exploitation, which have the potential to impact the amount and composition of PAHs deposited to the environment differently over time. Lake sediment cores can be used to reconstruct historical inputs of PAHs to lakes over time, including changes in diagnostic ratios, providing a means to assess changes in concentrations, composition and sources of PAHs to aquatic ecosystems that may have occurred over time, especially following the onset of industrial activity. The examination of diagnostic ratios in dated lake sediments, along with other factors that may help inform the interpretations of these ratios, such as historical records of petrochemical development, make paleoenvironmental reconstructions of PAH diagnostic ratios a useful tool for differentiating pyrogenic versus petrogenic sources over time. In addition to the ability to determine general sources of PAHs, sediment cores also allow us to understand how PAHs are actually entering aquatic ecosystems over time and how petrochemical developments may or may not be affecting their deposition.

2.2.5. Global and Canadian PAH deposition trends near petrochemical activity

Previous studies have tracked trends in sediment PAH profiles that are consistent with the general onset of the industrial revolution, including in the Mediterranean (Gabrieli et al. 2012) the Niger River Delta (Sojinu et al. 2010) and the Barigui River in Brazil (Machado et al. 2014). In contrast, much less work has been done on tracking the impacts specifically from petrochemical developments to PAH deposition and most has focused on soil and marine or river sediments. For example a recent study reported much higher concentrations and different composition of PAHs in topsoil near a coking facility as compared to soils near a pharmaceutical and detergent factory (Yuan et al. 2014). Likewise, Sojinu et al. (2010) found that PAHs were elevated in soil and river sediments in the Niger Delta near areas of petrochemical activity. The authors also found a dominant petrogenic signal, which they ascribed to disturbance from oil exploration efforts and leaking oil pipes (Sojinu et al. 2010). Fewer studies have focused on the impacts of petrochemical developments explicitly to lakes. Recent studies have shown

that the scale of land-use change caused by open-pit mining has affected the deposition of PAHs to lakes in the Athabasca region (Kurek et al. 2013, Jautzy et al. 2013) and that some lakes in the Cold Lake region are showing increases in PAH deposition whose timing is consistent with the onset of *in-situ* oil sands extraction in the area (Korosi et al. 2013). Virtually nothing is known as to how conventional exploration and extraction methods might be impacting PAH deposition to aquatic ecosystems, nor how these compare to the impacts from oil sands extraction. To the best of our knowledge, this study presents the first comprehensive direct comparison of temporal trends in PAHs across the major methods of petrochemical extraction employed in northwestern Canada, providing a critical context for assessing hydrocarbon emissions of petrochemical development.

2.3. Materials and Methods

2.3.1. Field Methods and Study Site Description

Study lakes were strategically chosen for the type of oil and gas development found within their catchments. In the Mackenzie Delta uplands (MDU) (Fig. 2.1), Lakes I17 (68.95˚N, 113.45˚W) and I20 (68.97˚N, 133.52˚W) (unofficial names, Table 2.1) are downslope from leaking natural gas drilling sumps (dumping pits for drilling waste, including borehole cuttings and drilling fluid). While located in proximity to these sump lakes, Lakes C23 (68.00˚N, 133.51˚W) and C1A (68.66˚N, 133.76˚W) (unofficial names, Table 2.1) are both upslope from any natural gas developments and are considered reference lakes for this region. In the Cameron Hills (CAM) (Fig. 2.1), lakes CamH1 (60.05˚N, 117.51˚W), CamH3 (60.14˚N, 117.57˚W) and CamH5 (60.03˚N, 117.48˚W) (unofficial names, Table 2.1, Fig.2.2) have at least one working or capped conventional gas or oil jack-pump within their catchments (see Table 2.1 for exact numbers and status of instalments), while the two regional reference lakes (CamH2 (60.22˚N, 117.80˚W) and CamH7 (60.14˚N, 117.38˚W)) (unofficial names, Table 2.1, Fig.2.2) are upstream of any petrochemical developments in the area. Lakes Ethel (54.31˚N, 110.21˚W) and Hilda (54.31˚N, 110.25˚W), in the Cold Lake (CL)(Fig. 2.1) region, are located south and southeast of major *in-situ* oil sands extraction activity, downwind of the prevailing winds

in the area. The three study lakes in the Fort McMurray/Athabasca oil sands (AOS) (Fig. 2.1) region are northwest (NW35 (57.08˚N, 111.59˚W)), northeast (NE20 (57.07˚N, 111.33˚W)) and southeast (SE22 (56.54˚N, 111.08˚W)) of AR6, an area of intensive open-pit oil sands extraction that also houses two bitumen up-graders (Syncrude and Suncor), with a third upgrader found further north of the area (CNRL Horizon). Sediment cores were taken from the central basin for all study lakes using a Glew gravity corer (Glew 1989). Lakes in the Mackenzie Delta uplands were sampled in August 2009 and July 2010, Cold Lake in July 2010, Athabasca in March 2011, and Cameron Hills in September 2012. All sediment cores were sectioned on-site into 0.25-1.0 cm intervals using a Glew vertical extruder (Glew 1989) and kept frozen until analysis.

2.3.2. Laboratory Methods

Sediment radiochronologies were determined using standard ^{210}Pb radioisotopic methods on an Ortec High Purity Germanium Gamma Spectrometer (Oak Ridge, TN, USA), and the constant rate of supply (CRS) model (Appleby 2002) for all lakes except CamH7 where the constant flux/constant sedimentation (CFCS) model was used (Robbins et al. 1978), as it was found to have a better fit than the CRS model. Sediments from select core intervals were freeze-dried and enclosed in epoxy resin, then equilibrated over a period of at least 2 weeks. Certified Reference Materials obtained from International Atomic Energy Association (Vienna, Austria) were used for efficiency corrections, and results were analysed using ScienTissiME (Barry's Bay, ON, Canada).

Wet sediment samples for PAH extraction were manually homogenized and mixed in roughly a 50:50 mixture with Hydromatrix®, and 1:2 mixture (by weight) with activated copper to remove water and sulphur, respectively, from the samples. The samples were then spiked with ^{13}C-standard of the US EPA Priority 16 PAHs, and extracted using accelerated solvent extraction (ASE, Dionex) at 100˚C with Hexane:Dichloromethane (DCM), followed by 50%Acetone:50%Hexane. The non-polar extract (PAHs, Hexane and DCM) was separated from the polar extract (water and acetone) by manually transferring the non-polar extract to a Turbovap vessel containing 3 ml of 2,2,4-Trimethylpentane, while the polar phase was washed three times with 10ml of hexane each time and then discarded. The non-polar phase (i.e. the phase containing PAHs) was then evaporated down to 1ml in a water bath of 23˚C under a steady stream of

clean, dry nitrogen gas. PAHs were separated from interfering compounds by solid-phase extraction (SPE) on SPE cartridges packed with silica gel (45µm, 0.8 cm^3/g, 475 m^2/g). The cartridges were first conditioned with 5ml of Acetone, 10ml of Pentane and 10ml of 2,2,4-Trimethylpentane. Samples were transferred on to the cartridges with 1ml of 2,2,4-Trimethylpentane, and allowed to bind to the SPE packing over a period of 15 minutes. The samples were then washed with 2.5ml of Pentane and eluted off the column with 7ml Methylene Chloride (DCM)/Pentane (2:3, v/v). The eluent was evaporated down to 1ml again in a water bath of 23˚C using a gentle stream of clean, dry nitrogen gas, and analyzed by gas chromatography (Agilent 6890) and mass spectrometry (Agilent 5973). 1µl injections were made in pulsed splitless mode at 280˚C on a DB-XLB 30m x 0.18µm x 180µm column. An initial oven temperature of 60˚C was held for 2 minutes then increased at a rate of 6˚C per minute to 300˚C and held for 10 minutes. A constant flow rate of 39cm per second of Helium was used for a total run time of 52 minutes. The mass spectrometer was set to have a transfer line temperature of 280˚C, with a source temperature of 230˚C and quadrapole temperature of 150˚C. Average recovery rates for sediment cores from the NWT lakes ranged from 27% to 94%, not including the most volatile two- and three-ringed-PAHs, which were, for the most part, in the 10% to 50% range. Details on methods and recoveries for sediment cores from lakes in Alberta can be found in Korosi et al. (2013) and Kurek et al. (2013). All PAH concentrations were calculated using the isotope dilution method, which was used to correct for variability in extraction and analytical methods and low recovery rates.

2.3.3. Data Analysis

Per cent change in concentrations were calculated using the following calculation:

$$\% \text{ Change PAH} = (\text{Avg. } \Sigma PAH_{POST} - \text{Avg. } \Sigma PAH_{PRE})/\text{Avg. } \Sigma PAH_{PRE} \text{ X } 100 \quad \text{(eq.1)}$$

Relative abundances were calculated using the following calculation:

$$\% \text{ Individual } PAH_{\text{Rel. Abun.}} = (\text{Individual PAH}/\Sigma PAH) \times 100 \quad \text{(eq. 2)}$$

ΣParent included the following compounds: acenaphthylene (ACY), acenapthene (ACE), fluorene (FLU), anthracene (ANT), phenanthrene (PHE), fluoranthene (FLA), pyrene (PYR), banz[a]anthracene (BaANT), chrysene (CHR), benzo(b)fluranthene (BbFLA),

benzo(k)fluoranthene (BkFLA), benzo[a]pyrene (BaPYR), indeno(1,2,3-cd)pyrene (IcdP), dibenz(a,h)anthracene (DahA) and benzo(g,h,i)perylene (BghiP). The ΣAlkyl used included all C1-, C2- and C3-fluorenes, C1-, C2-, C3and C4-Phenanthrene/Anthracenes, C1-, C2-, C3and C4-Fluoranthene/Pyrenes, C1-, C2-, C3 and C4-Benz[a]anthracene/Chrysenes and C1- and C2-Benzofluoranthene/Benzopyrenes. ΣPAH includes all compound mentioned above (i.e. both sum of parent and sum of alkyl PAHs). Naphthalene (NAPH) and its alkylated counterparts were left out of all statistical analyses and calculations of ΣParent and ΣAlkyl due to highly variable recovery rates.

The following PAH ratios were calculated and used in statistical analyses or figures:

FLA/PYR ratio = FLA/(FLA+PYR) (eq.3)

IcdP/BghiP ratio =IcdP/(IcdP+BghiP) (eq.4)

These particular ratios were chosen because they are appropriate for distinguishing the general sources of pyrogenic versus petrogenic PAHs that we expected to find (Tobiszewski and Namiesnik 2012, Lima et al. 2005), and have been used effectively in previous studies to identify sources in the AOS (Jautzy et al. 2013). Mann-Kendall trend tests were conducted on PAH concentrations and ratios in order to elucidate any trends in PAHs over time. In order to assess how modern PAH composition might differ in key compounds amongst the various regions, principal components analyses (PCA) were conducted on relative abundance of individual parent and alkyl PAHs in sediments from each of the sites averaged over their respective post-development time periods. In reference lakes, the timing of the nearest development was used to determine which intervals were theoretically post-development. As we had few sampling sites (n=14 lakes) compared to the number of compounds included in the statistical analysis (35 parent and alkyl PAHs), PAHs were grouped in each PCA based on their inclusion in diagnostic ratio pairs, and included their alkylated counterparts. PCAs were run on square-root transformed relative abundances and were tested for normality using the Shapiro-Wilk Normality Test to ensure the analyses were not driven by comparatively high relative abundances. All data were analysed using R statistical software, while specific analyses were done with the following add-on packages: Mann-Kendall trend tests were run using the Kendall package, and PCAs were conducted using the vegan package.

2.4. Results

2.4.1. Sedimentary PAH concentration trends

In the Athabasca oil sands region (AOS)(open-pit mining of oil sands, n=3), Mann-Kendall trend tests show that both the ΣParent and the ΣAlkyl PAHs have undergone statistically significant increases over time in all sites (ΣParent: NE20 τ=0.779 $p<0.00001$, SE22 τ=0.743 $p<0.00001$; ΣAlkyl: NE20 τ=0.874 $p<0.0001$; SE22 τ=0.673 $p<0.0001$; NW35 τ=0.421 $p=0.01$), with the exception of ΣParent in lake NW35, where the increase wasn't statistically significant (τ=0.147, p= 0.38). The two cores from the Cold Lake region (CL)(*in-situ* oil sands extraction, n=2) showed increases in PAH concentrations over time in all cores for both parent and alkyl PAHs, although none were statistically significant. The exception to this is a decrease in ΣParent in Ethel Lake, which was found to be statistically significant (τ=-0.467, p=0.013). It should be noted that the ^{210}Pb dating shows the start of variations in Hilda Lake is consistent with the onset of *in-situ* oil sands extractions in the area (mid-1980's). Three lakes in the Cameron Hills (CAM)(oil and gas (OG) pump, n=5) region showed a statistically significant increase in parent PAHs (CamH1 τ= 0.556, p=0.032; CamH5 τ= 0.556, p=0.048; CamH7 τ=0.745, $p<0.01$) over the history of the cores representing approximately 200 years (estimated from ^{210}Pb dating); all other sites in this region showed no significant trends over time for either ΣParent or ΣAlkyl (Fig. 3.3-3.6, Tables A1-A14). Two cores in the Mackenzie Delta (MDU)(natural gas (NG) sump, n=4) were found to decrease significantly over time in ΣParent concentrations (I17 τ=-0.566, p=0.032; C23 τ=-0.611, p=0.028), but not in ΣAlkyl (Fig. 3.1-3.2, Tables A1-A14). No major differences in concentration were found between reference and impacted sites in the MDU and CAM regions, with the exception of C1A (reference lake in MDU) and CamH5 (impacted lake in CAM), which both showed generally higher concentrations throughout their respective core's histories, than the other conventional lakes.

The average concentrations of ΣParent were highest in the Cold Lake region (*in-situ* oil sands mining) (202 ng/g dry wt.) compared to the other three regions (173 ng/g dry wt., 157 ng/g dry wt. and 131ng/g dry wt. for the Athabasca oil sands (AOS),

Cameron Hills (CAM) and Mackenzie Delta Uplands (MDU) respectively)(Tables A1-A14). ΣAlkyl PAHs were far greater in the lakes from the AOS (near open-pit mining) (1038 ng/g dry wt.), compared to the other three regions investigated (628 ng/g dry wt., 562 ng/g dry wt. and 66 ng/g dry wt. in CAM, MDU and CL respectively) (Tables A1-A14).

In order to determine what the concentration profiles meant in terms of regulatory guidelines for sediment quality, we looked at the proportion of sediment intervals for each region to exceed the federal Interim Sediment Quality Guidelines (ISQG) developed by the Canadian Council for Ministers of the Environment (CCME) both pre-development and post-development. Combined, sediments from both oil sands regions (Cold Lake and Athabasca) exceed the ISQG (CCME 1999) twice as often (14 out of a possible 24 times) as the conventional sites combined (Mackenzie Delta Uplands and Cameron Hills) (7 out a possible 24 times)(Fig. 2.3) in modern sediments (i.e. post-development). While pre-development sediments exceeded guidelines to a similar extent in both regions (8 out of 24 times for the conventional sites and 10 out of 24 times for the oil sands sites) (Fig. 2.3). Additionally, when the Cold Lake region exceeds ISQG (CCME 1999) it tends to be in the lighter weight PAHs in pre- and post-development sediments, while the Athabasca region exceeds for LMW PAHs in pre-development sediments and the HMW PAHs in post-development sediments. Both conventional regions tend to exceed in the LMW PAHs in pre- and post-development sediments.

2.4.2. Comparing PAH composition across regions

Clear differences in PCA axis scores were observed between regions for PHE, ANT and their alkylated counterparts. As seen in the PCA biplot ($\lambda1=0.44$, $\lambda2=0.42$)(Fig. 2.4), lakes in the Cameron Hills (OG pump) tended to exhibit greater abundance in PHE, whilst the largest component of the alkyl PAH profiles throughout the open-pit region were the C4-PHE/ANTs, which comprises roughly a third of all alkyl PAHs across the three lakes in the region throughout the sediment deposits. Lakes near open-pit mining were also comparatively abundant in BaANT and CHR, as indicated by the PCA bi-plot in Figure 2.5 ($\lambda1=0.85$, $\lambda2=0.09$), with average %relative abundance of 7% and 12% respectively, compared to all other regions (average <2% for BaANT and <6% for CHR).

Furthermore the average %relative abundance for both compounds (BaANT and CHR) roughly doubles from pre-development sediments to post-development sediments in all three lakes in the AOS, a trend we don't see in any other region.

ACY comprises on average 19% of the parent PAHs seen in the Cold Lake cores, however the timing of the deposition of this compound differs amongst the cores. The average relative abundance in Hilda Lake increases dramatically from 4% in pre-development sediments to 33% in modern sediments, with large peaks around 1999 and 2009. Meanwhile, Ethel Lake has seen a variable ACY profile that decreases to virtually zero during post-development from an average of 30% across pre-development sediment. Interestingly, both conventional regions as well as the AOS sites see a consistent average %relative abundance of ACY of less than 2% throughout their respective core histories.

PHE was the only compound to be found in average regional abundances of greater than 10% across all regions. In addition, the relative abundance of PHE showed increases over time greater than 5% at all sites in the Cameron Hills (OG pump), with the exception of CamH5. The Cameron Hills lakes were greatest in both relative abundance and concentration in C4-BaANT/CHR compared to the other regions, while the MDU lakes were greatest in IcdP and BghiP. No major differences were found in relative abundances between the reference and impacted lakes in the MDU and CAM regions. Both oil sands regions (*in-situ* and open-pit) show markedly lower proportions of BbFLA (average 5% across all oil sands lakes) than either of the conventional regions (NG sump and OG pump) (average 13% across all conventional lakes). In addition, C3-FLU was relatively high in all of the conventional lakes (>11%) and low in all of the oil sands lakes (<5%).

2.4.3. Spatial and temporal trends in PAH sources

Mann-Kendall trend tests show that the declines in FLA/PYR and IcdP/BghiP ratios over time is statistically significant in the Athabasca oil sands region (FLA/PYR: NE20 τ=-0.926 $p<0.00001$, NW35 τ=-0.792 $p<0.00001$, SE22 τ=-0.684 $p<0.0001$; IcdP/BghiP: NE20 τ=-0.904, $p<0.00001$; NW35 τ=-0.581 $p<0.001$), with the exception of the IcdP/BghiP ratio in Lake SE22 where the slight decline is not statistically significant (SE22 τ=-0.030 $p=0.89$). A cross-plot of these two key ratios down-core for

representative cores from each region (I20, CamH3, Ethel and NE20 for the NG sump, OG pump, *in-situ* and open-pit sites respectively) (Fig. 2.6) shows that FLA/PYR ratios have, for the most part, remained above the pyrogenic threshold of 0.4 for the Mackenzie Delta (NG sump), Cameron Hills (OG pump) and Cold Lake (*in-situ*) sites, which is confirmed by the statistically non-significant results of the Mann-Kendall trend tests for the FLA/PYR ratio over time in these regions. The Athabasca region, however, has undergone a shift in this ratio from an average of 0.49 in pre-development sediments to 0.26 in post-development sediments, crossing the threshold from pyrogenic to petrogenic sources. Although the shift is not as dramatic in the IcdP/BghiP ratio, a steady decrease in the ratio from 0.5 at the bottom of the core to 0.24 at the top (Fig. 2.6) corroborates a shift in sources in the Athabasca region, while the conventional regions exhibit an IcdP/BghiP ratio that has remained relatively constant throughout their respective cores (I20, CamH3). Interestingly, the IcdP/BghiP ratio in Ethel Lake increased towards the surface, from 0.06 at the bottom of the core to 0.48 at the top (Fig. 2.6), indicating that some PAH sources are becoming more pyrogenic in recent times. These results are supported by the Mann-Kendall trend test for these three regions (I20: τ=0.092 p=0.730; CamH3: τ=0.089 p=0.788; Ethel: τ=0.4 p=0.007)

2.5. Discussion

2.5.1. Impacts on sedimentary PAH concentrations

Using a comparative analysis across all four regions has allowed us to put the PAH concentration profiles of each individual region into the greater context of sedimentary PAH concentration across northwestern Canada. Our analyses clearly showed that the trend of enrichment in sedimentary PAH concentrations in the AOS lakes was far greater than any other regions examined. In addition, the timing of this enrichment is consistent with the documented onset of open-pit mining in the area (late 1960's), as demonstrated by per cent change in average concentrations from pre-development (ie: background) concentrations compared to average concentrations in post-development sediments (Fig. 2.7). Along with previous studies, that have shown PAH uptake and detrimental effects to biological function at different trophic levels from

exposure to PAHs (Camus et al. 2003, Wayland et al. 2008, Carls et al. 2010, De Laender 2014), this provides further evidence that bitumen mining in the Athabasca region constitutes an important stressor on aquatic ecosystems through the amplification of PAH concentrations that is not evident in lakes near the other types of petrochemical development included in this study.

Some increases in PAH concentrations are seen towards surface sediments in the lakes around *in-situ* oil sands mining at Cold Lake (CL), and it would appear that in Hilda Lake, the change is consistent with the timing of the oil sands development, according to ^{210}Pb dating (Korosi et al. 2013). Although we do see some spikes in individual PAH concentrations into the high 100's of ng/g dry wt., these occur in only a few sediment intervals (eg: ACY, ACE and C2-PHE/ANT in the late 1990's) and may be due to specific release events, as sedimentary PAH concentrations of these compounds generally have a low background in the Cold Lake region. Korosi et al. (2013) found that soil samples taken in and around the most intensive *in-situ* development area near Cold Lake were generally low in PAHs with the exception of soil samples taken to the southwest of the most intensive development, where high levels of alkylated PAHs were observed. The authors go on to point out, however, that although no quality guidelines exist for alkylated PAHs, none of their samples exceed the parent PAH concentrations included in the Canadian Soil Quality guideline. By contrast, the concentrations of some compounds (eg: DahA, genotoxic and a probable carcinogen to humans) (CCME 1999) in lake NE20 near intensive open-pit mining in the AOS, are steadily increasing and have exceeded the CCME sediment quality guidelines (CCME 1999) since the mid 1980's, according to ^{210}Pb dating models. This would suggest that the impact of petrochemical development on PAH concentrations is less near *in-situ* petrochemical operations compared to lakes near open-pit mining. This difference is likely due to the larger scale of land-use change in open-pit mining sites compared to *in-situ* mining sites, as well as the lack of bitumen upgraders in Cold Lake compared to the AOS region which houses two upgraders (Suncor and Syncrude) in the immediate vicinity of the study lakes, with a third upgrader further north of the study lakes (CNRL Horizon). Furthermore, a recent study has suggested that the 182 km^2 of open-pit mining waste-water tailings ponds in the AOS region export up to 1069 kg of PAHs yr^{-1} into the local atmosphere (Galarneau et al.

2014) which could also contribute to the PAH loads in surrounding lakes through precipitation or dry deposition, while this potential source is absent in the Cold Lake region.

In the conventional resource regions concentrations of PAHs, especially the alkylated compounds, were generally higher or comparable to what a previous study of PAHs in a similarly remote region found (Headley et al. 2002) although much lower than urban areas or industrial centres (Yuan et al. 2014). This was particularly true for some lakes in the Mackenzie Delta uplands (MDU) and especially when corrected for organic carbon, as sedimentary organic carbon is low in this region (4%-25%) compared to the other study regions (14%-42%) (Tables A1-A14). However, since concentrations in sediment cores from the conventional resource areas remain constant over time, or decrease slightly, the higher than expected values are probably due to global background sources and the naturally rich hydrocarbon geology in the area. A massive Cretaceous marine shale formation, known to be rich in unconventional shale oil and gas, underlies both of the conventional development regions (MDU and CAM) (AANDC 2011) and may be causing the consistently elevated baseline concentrations in some of these lakes, especially in the alkylated PAHs, in both pre- and post-development sediments. The steady concentrations of most PAHs in the conventional lake sediment cores contrasts sharply with the increasing concentrations in the sediment cores from lakes near the Athabasca oil sands, where the average concentration of 31 out of 32 PAHs included in the statistical analyses increased from pre-development sediments to post-development sediments, compared to increases in only 3 compounds in the MDU lakes and 6 compounds in the CAM lakes.

2.5.2. Identification of compounds as regional markers

Over 90% of future growth in the development and production of petrochemical resources in Canada is expected to be in northwestern Canada (CAPP 2014). As such it would be helpful to monitoring efforts to characterize the key components in background and more modern PAH profiles in ecosystems near the different extraction methods used in the region. By comparing PAH compositions in the four study regions we were able to isolate specific compounds that may be used as regional identification markers in

sediments. For example, C2-FLU seems to be a fairly strong indicator of conventional sites and is found, on average, at four times greater abundance in the conventional resource regions of the Northwest Territories (MDU and CAM) than in the oil sands regions of Alberta (AOS and CL). Important contributors to the environmental PAH signatures in the MDU are C3-FLU, C3-PHE/ANT and C3-FLA/PYR, which show both the greatest average relative abundance and average concentrations for these compounds, while C4-BaANT/CHR is an important contributor in the Cameron Hills region, although PHE is also an important contributor to the PAH load in this area, making up nearly a quarter of the parent PAHs across the region. This is corroborated by the PCA in Figure 2.4, which also shows enrichment in PHE in this region. Figure 2.4 also corroborates the relative abundance results that show C4-PHE/ANT as an important marker for the Athabasca oil sands. Relative abundances and a PCA of the BaANT/CHR ratio partners and their alkyl counterparts (Fig. 2.5) also suggest that known carcinogens (CCME 1999), benz(a)anthracene and chrysene, are also important markers in the Athabasca region. In the Cold Lake region, there is a predominance of the three lowest molecular weight (LMW) PAHs, which is not apparent in the other study regions. This is particularly true for ACY which exceeds the Interim Sediment Quality Guidelines developed by the Canadian Council for Ministers of the Environment (CCME) throughout most of both sediment cores (Ethel and Hilda), from this region. Additionally, three intervals in Ethel (at 32cm, 26cm and 8.5cm) and two intervals in Hilda (at 0.5cm and 2.5cm) exceed Probable Effects Levels developed by the CCME (CCME 1999), indicating that ACY is a compound of particular concern in the Cold Lake region.

2.5.3. Shifts in PAH sources

Directly comparing the PAH deposition history in the different regions has allowed us to see that a considerable shift in sources of PAHs has occurred in recent years in the AOS lakes (open-pit mining of oil sands), and to a lesser degree near *in-situ* extraction of oil sands in Cold Lake, that is not apparent in either of the conventional development regions. The crossplot of the FLA/PYR and IcdP/BghiP ratios shows that modern sediments in the conventional (MDU and CAM) and CL regions all have sizeable contributions from pyrogenic PAHs to their inventory (Fig. 2.6). The AOS core,

on the other hand, seems to have undergone a distinct shift from pyrogenically sourced PAHs, in sediments that were deposited around the early 1950's, when coal and wood were the chief fuel sources in the area, to more petrogenically sourced PAHs in modern sediments.

As seen in the FLA/PYR and IcdP/BghiP crossplot (Fig. 2.6), the study regions were all dominated historically by PAHs from pyrogenic sources, with the exception of Cold Lake (*in-situ* oil sands extraction) which appears to have petrogenic origins to its IcdP in older sediments, shifting to sources that are more pyrogenic in origin towards modern sediments. The sources of FLA, on the other hand, remain consistently pyrogenic throughout the history of the Cold Lake core. The major shift in the IcdP/BghiP axis of the ratio crossplot is mainly due to a marked decrease in BghiP towards the surface of the core. IcdP and BghiP (molar mass=276.33 g/mol) have fairly similar physical properties that would dictate their behaviour during transportation and degradation. This might suggest that the cause of the shift in composition would be a change in source rather than transportation or degradation, as we would expect these two compounds to behave very similarly during transportation and degradation. We do see a slight decrease in IcdP but the magnitude of decrease is much greater for BghiP, indicating that the transportation and/or degradation processes of these compounds may have changed slightly, but the major cause for the shift in BghiP, and thus the IcdP/(IcdP+BghiP) ratio, is in source. As part of the *in-situ* extraction method, natural gas is burned to create the steam necessary to liquefy the bitumen at depth for *in-situ* separation from the sand before being brought to the surface (ie: steam-assisted gravity drainage (SAGD)). Thus the shift in source of IcdP and BghiP we are seeing may indicate that this method of oil sands extraction is altering the relative abundance of these PAHs. It should be noted that concentrations of IcdP and its ratio partner, BghiP, are fairly low, and the calculated ratio values become more difficult to interpret at low concentrations of both compounds, as even small changes in concentrations can cause larger relative shifts in the ratios. However, the different shifts in sources seen in the two oil sands regions, petrogenic to pyrogenic in CL and pyrogenic to petrogenic in the AOS may be due to differences in their respective extraction techniques; as mentioned above the *in-situ* extraction method involves the

burning of natural gas, while open-pit mining causes the scattering of oil sands particles through wind action.

Petroleum extracts from both conventional sites are piped off-site for processing and upgrading, thus there are no up-graders or refineries near the sampling sites, nor does any major off-gassing take place in the MDU or CAM regions. As such, if conventional developments were affecting PAH sources to lakes in these regions we would expect to find a significant increase in the dominance of petrogenically sourced PAHs, from disturbance to the surrounding hydrocarbon-rich catchments during petrochemical extraction, as was observed by Sojinu et al. (2010) in the Niger River Delta. The FLA/PYR and IcdP/BghiP ratio cross-plot (Fig. 2.6) clearly shows that this is not the case in either of the conventional study regions. Indeed, the representative cores from both the MDU and CAM show sources of PAHs in the regions are mixed, with pyrogenically sourced PAHs dominating throughout their respective core's histories. We do see some changes in concentration of parent PAHs over time, though they are small and likely due to shifts in natural background sources. Furthermore, while somewhat elevated the concentrations of alkylated PAHs, whose enrichment is typically attributed to petrogenic sources, shows some variability but no clear trend that is coincident with the timing of petrochemical development in these areas. PAHs in these regions are likely coming from multiple background sources, which could include some input from local forest fires, as well as long-range transport from distant industrial sources (Hung et al. 2005, Genualdi et al. 2009, Headley et al. 2002, Ruiz-Fernández et al. 2014). Furthermore, in light of the elevated average ΣAlkyl PAH concentrations found at some sites (CamH5 801 ng/g dry wt., CamH7 1093 ng/g dry wt. and C1A 1053 ng/g dry wt.), sources likely also include a considerable input from the naturally abundant hydrocarbon geology in these regions, which is why these areas are developed for petrochemical extraction in the first place.

The unique comparative approach of this study has allowed us to place the impacts previously observed from the unconventional, or oil sands, developments to PAH deposition into the greater framework of spatial and temporal trends across northwestern Canada. The comparative approach we used, including statistics and PAH crossplots, in contrasting the historical trends in PAH concentrations has proven useful in establishing the baseline concentrations of these contaminants across different geological areas (ie: oil

sands and conventional deposits), and is essential if we are to understand the impacts petrochemical developments may be having on PAH deposition to aquatic ecosystems. This is especially true for these more remote regions where little to no data exist on historical PAH deposition to the environment, and where exploration and development of petrochemical resources is expected to increase (CAPP 2014). Ultimately our comparative analyses suggest that the scale of change in the concentrations of PAHs in the Athabasca region is much larger than in the other three regions (Figs. 2.3, 2.6, 2.7), and that the increase seems to be related to the development of open-pit mining of the oil sands in the area, while all other regions have seen relatively little (Cold Lake) or no impacts (MDU, CAM) to the concentrations of PAHs in lake sediments from petrochemical development in their proximity (Figs. 2.3-2.7). Additionally, we were able to see that changes in PAH sources in the Athabasca and Cold Lake regions are largely reflective of the onset of petrochemical developments in these areas, while sources in the Cameron Hills and Mackenzie Delta are a mix of background sources that have remained virtually unchanged for the history of the sediment cores (Fig. 2.6). This interpretation agrees with previous studies (Kelly et al. 2009, Kurek et al. 2013, Timoney and Lee 2011) that have suggested that the scale of land-use change in the Athabasca area has amplified naturally elevated levels of PAHs in nearby ecosystems. We have found that some regions have high background levels of PAHs while other regions have lower background levels, and have taken the first steps to understanding how different types of petrochemical developments may affect these. However, more detailed analysis will be needed to understand the exact sources of variation in the background levels of PAHs in these regions. Finally, this study has identified specific PAH compounds as potential chemical markers for the different regions that would benefit from further research into their toxicological potential and risk to vital ecosystem services.

Table 2.1: Details of petrochemical developments near all sampled lakes

Lake ID	Region	Impact	Associated Development – Industry ID	Resource Type	Development Status
C1A	**Mackenzie Delta, NWT**	**Reference**	**N/A**	**N/A**	**N/A**
C23	**Mackenzie Delta, NWT**	**Reference**	**N/A**	**N/A**	**N/A**
I17	**Mackenzie Delta, NWT**	**Natural Gas Sump**	**Parsons-F09**	**Gas**	**Abandoned**
I20	**Mackenzie Delta, NWT**	**Natural Gas Sump**	**Parsons-F09**	**Gas**	**Abandoned**
CamH1	**Cameron Hills, NWT**	**Oil/Gas Jack pump**	**2M-73**	**Oil**	**Producing**
			M-73	**Oil**	**Producing**
			L-73	**Oil**	**Producing**
CamH2	**Cameron Hills, NWT**	**Reference**	**N/A**	**N/A**	**N/A**
CamH3	**Cameron Hills, NWT**	**Oil/Gas Jack pump**	**A-19**	**Oil**	**Planned**
			C-19	**Oil**	**Shut-In**
			F-19	**Oil**	**Shut-In**
			K-19	**Oil**	**Shut-In**
			O-19	**Oil**	**Shut-In**
CamH5	**Cameron Hills, NWT**	**Oil/Gas Jack pump**	**J-62**	**Gas**	**Shut-In**
			E-52	**Oil**	**Planned**
CamH7	**Cameron Hills, NWT**	**Reference**	**N/A**	**N/A**	**N/A**
Ethel	**Cold Lake, AB**	***In-situ* Extraction**	**SAG-D**	**Oil Sands**	**Producing**
Hilda	**Cold Lake, AB**	***In-Situ* Extraction**	**SAG-D**	**Oil Sands**	**Producing**
NE20	**Fort McMurray, AB**	**Open-pit Extraction**	**AR6 Mine**	**Oil Sands**	**Producing**
NW35	**Fort McMurray, AB**	**Open-pit Extraction**	**AR6 Mine**	**Oil Sands**	**Producing**
SE22	**Fort McMurray, AB**	**Open-pit Extraction**	**AR6 Mine**	**Oil Sands**	**Producing**

Figure 2.1: Map of Canada indicating approximate locations of the four study regions. (map source: Canadian Geographic)

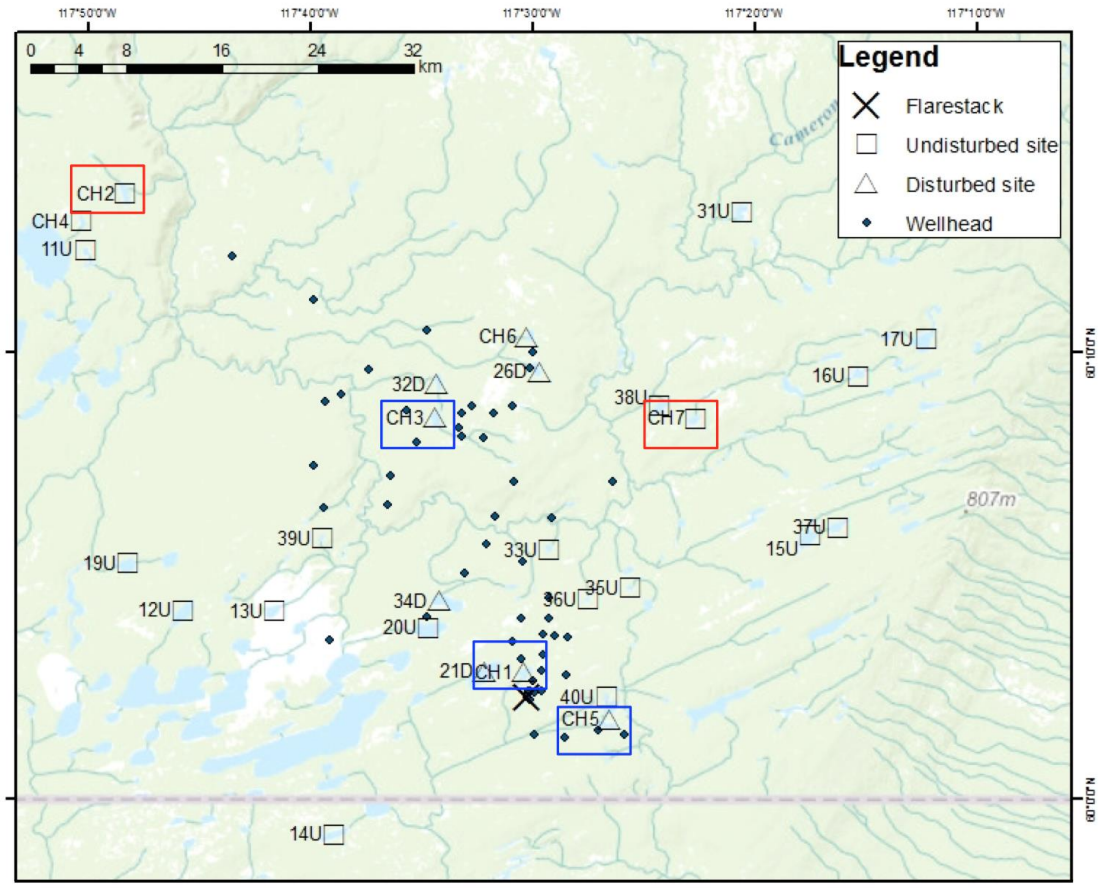

Figure 2.2: Map of the Cameron Hills showing the impacted sites in blue boxes and reference sites in red boxes. (map courtesy of Michael Palmer, Cumulative Impact Monitoring Program, Government of the Northwest Territories)

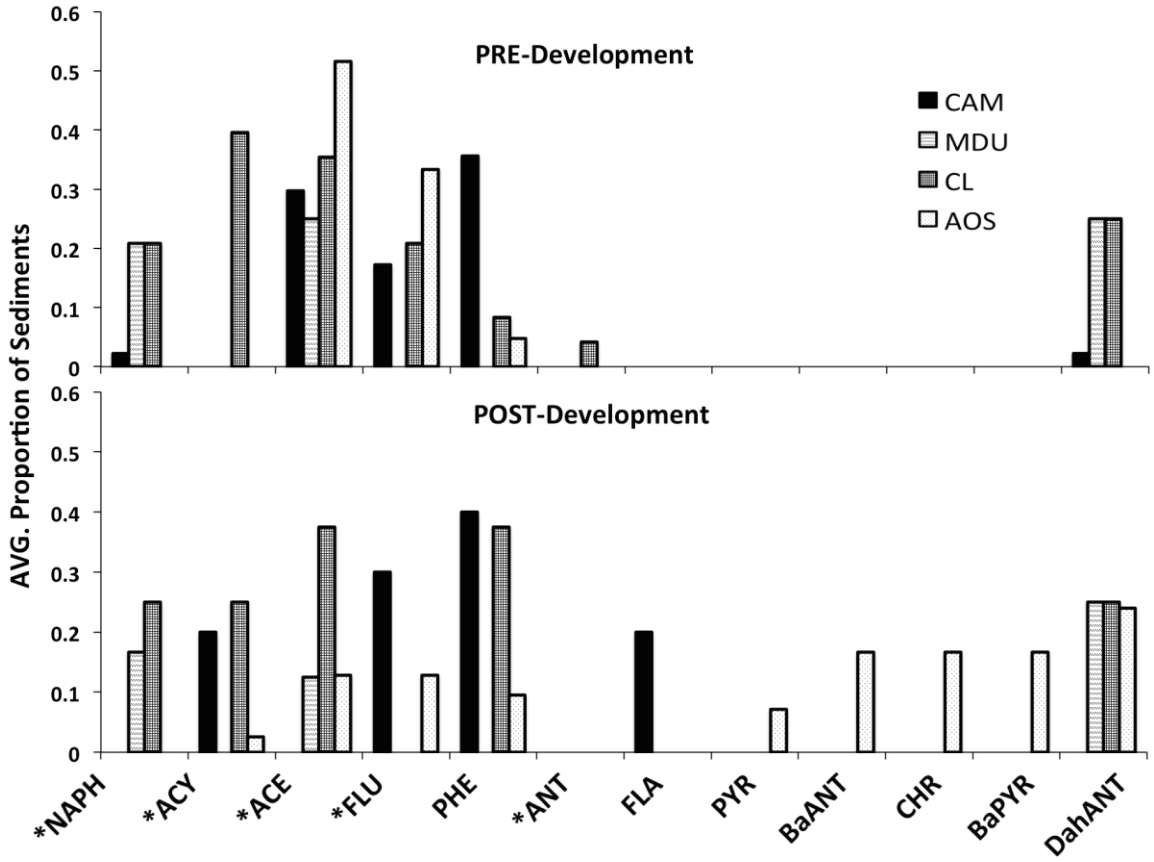

Figure 2.3: The average proportion of sediment intervals per region that exceed Interim Sediment Quality Guidelines (ISQG) developed by Environment Canada for freshwater sediments; showing oil sands regions (Cold Lake (CL) (n=2) and Athabasca Oil Sands (AOS) (n=3)) exceeding guidelines more than twice as often as conventional regions (Mackenzie Delta Uplands (MDU) (n=4) and Cameron Hills (CAM) (n=5)) in post-development sediments. From L to R PAHs are in order of increasing molecular weight. (* indicates compounds whose guidelines were adapted from marine sediment guidelines)

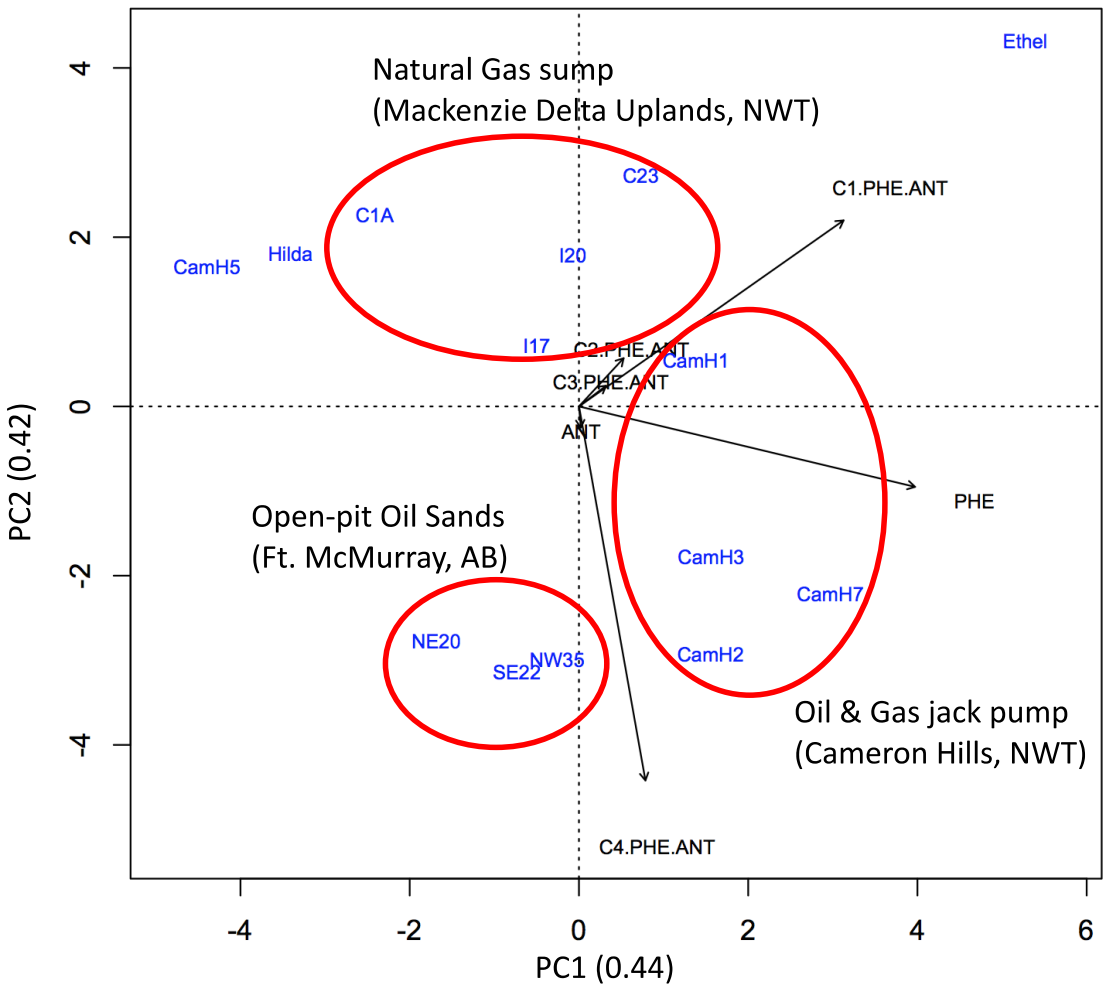

Figure 2.4: Principle components analysis of post-development sediments from all sites for ANT, PHE and their alkylated counterparts, showing enrichment in PHE in the Cameron Hills lakes and enrichment on C4-PHE/ANT in the Athabasca oil sands lakes near Fort McMurray.

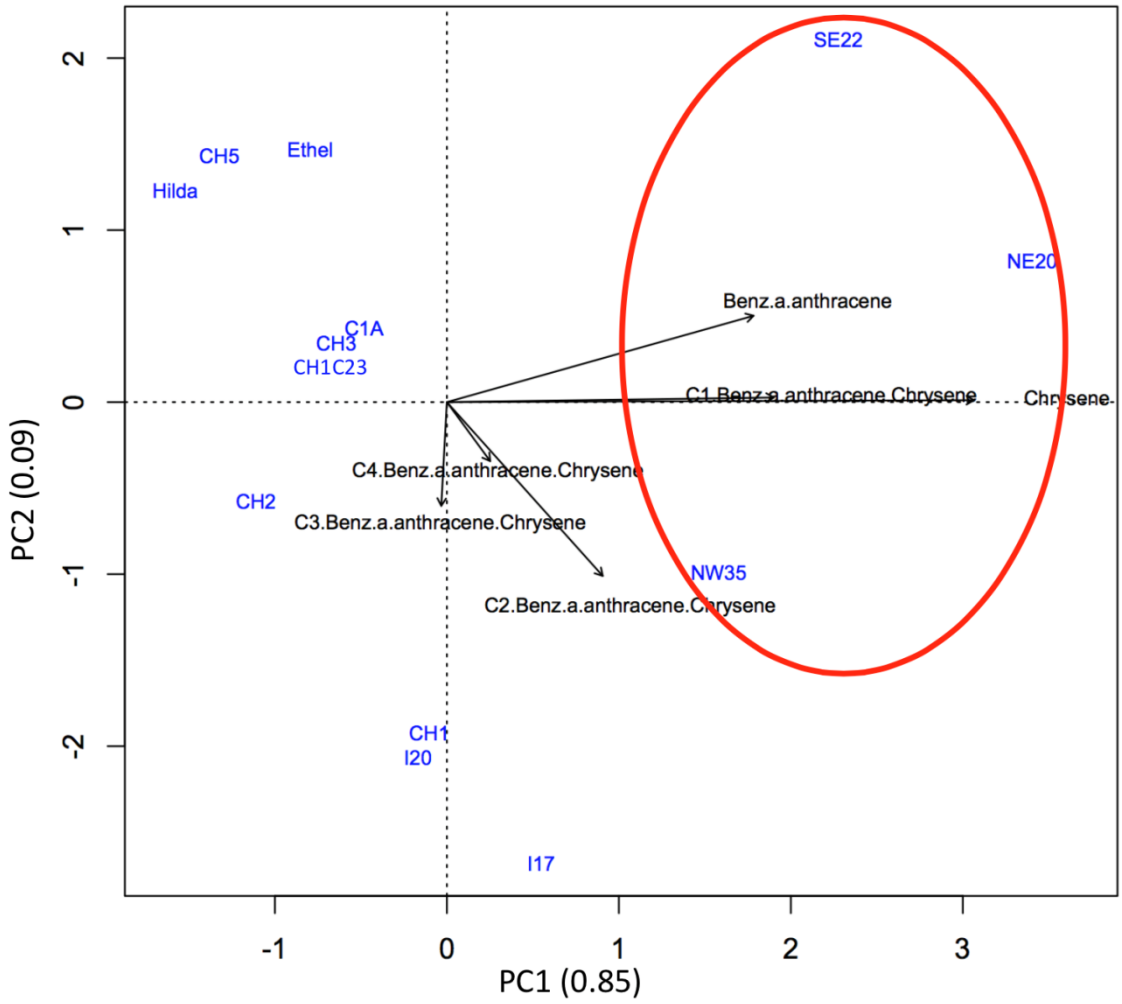

Figure 2.5: Principle components analysis of relative abundance of select PAHs showing enrichment of Athabasca oil sands (circled in red) around known carcinogens BaANT, CHR and C1-BaANT/CHR.

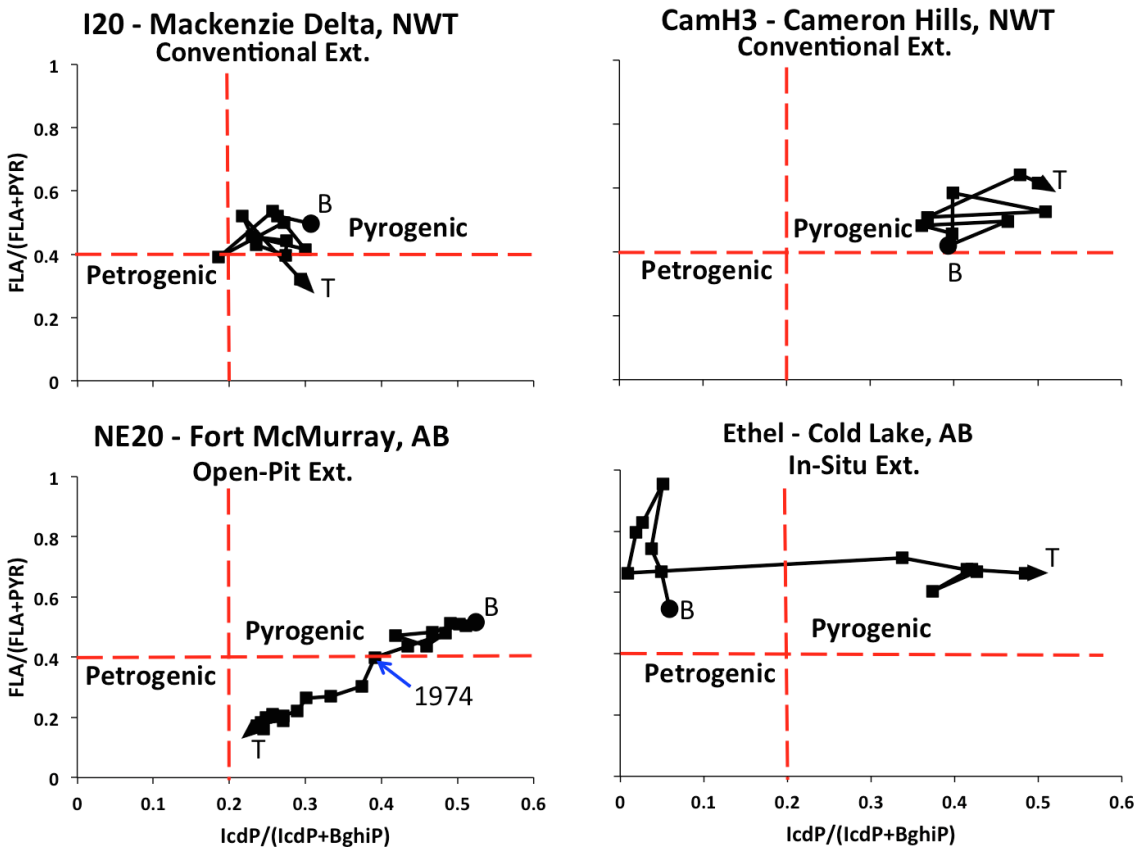

Figure 2.6: Cross-plot of ratios down-core from representative cores in each region showing a large shift in PAH sources in the Athabasca region and a moderate shift in sources of the heavier PAHs in the Cold Lake region. Dashed red lines represent ratio thresholds; while B marks the bottom of the cores (i.e. older sediments) and T marks the most recently deposited sediments at the top of the core.

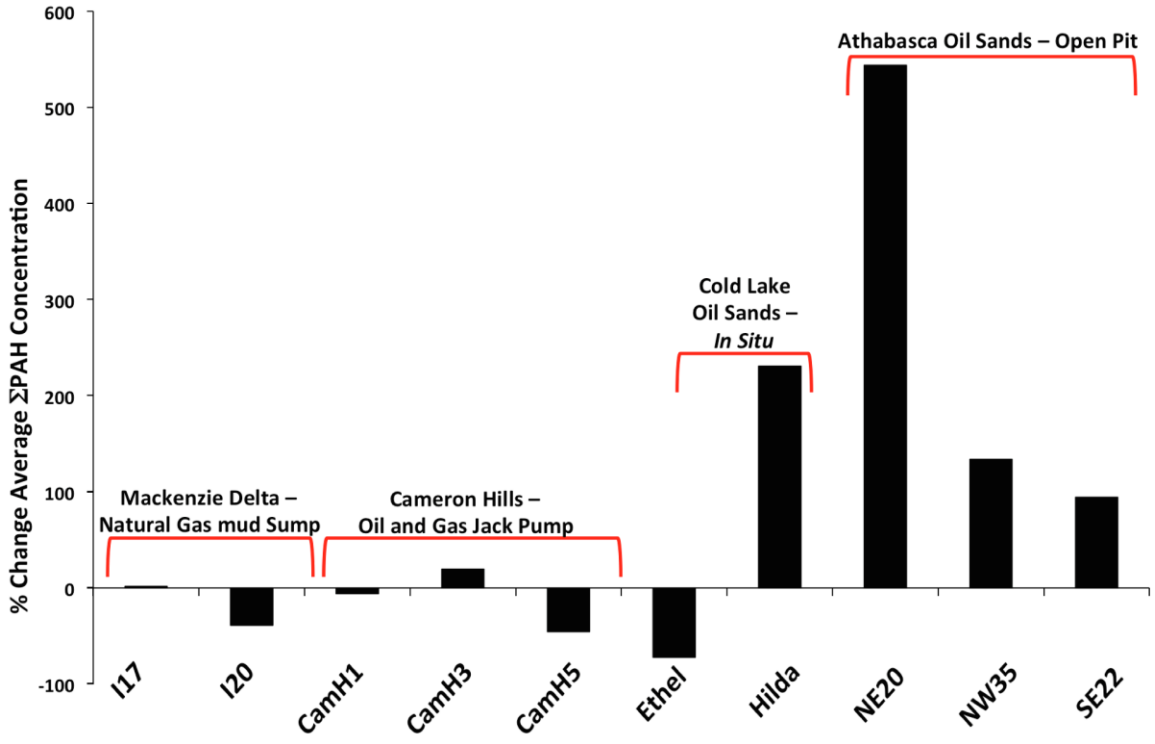

Figure 2.7: Per cent change (% Change, see eq. 1) of the average concentrations of total PAHs from post-development sediments compared to the average concentrations of total PAHs in pre-development sediments, showing large increases in concentration across all lakes in the Athabasca oil sands compared to small variations across impacted conventional resource sites.

CHAPTER 3. Investigating linkages between climate warming and PAH deposition histories in the NWT

3.1. Abstract

Among the documented effects of climate change, increases in forest fire disturbance and increased primary production have the capacity to alter the concentrations and sources of PAHs in the environment. Through the analysis of radiometrically-dated lake sediment cores from the Mackenzie Delta Uplands, NWT, this study examines how recent changes in climatic conditions have affected the delivery of polycyclic aromatic hydrocarbons (PAHs) to aquatic ecosystems. PAH parameters (concentration and composition) were examined against proxies for climate change: diatom community assemblages, inferred chlorophyll *a* and its diagenetic products, per cent organic carbon (%OC) and the different carbon fraction of Rock Eval pyrolysis. In addition, retene, an alkylated PAH in the phenanthrene/anthracene homologous family, known to be indicative of forest fires, was used to assess the general fire history in the area, as well as the relative contribution of forest fires to the PAH profile in lake sediments. The relative abundance of retene was generally below 5% of the alkyl PAHs found in the study lakes. The exception to this was a spike of 35% in retene in CamH5, which was accompanied by a small increase in %OC at the same time, suggesting this marks a fire event. Spearman's correlation analysis of all PAH parameters with all climate proxies were not statistically significant, except for the negative correlations between %OC and the sum of parent PAHs in CamH7 (ρ= -0.89, $p<0.005$) and C23 (ρ= -0.9, $p< 0.005$), and the positive correlation between the S3 fraction of the Rock Eval pyrolysis in CamH5 (ρ= 0.87, $p< 0.005$), which were found to be statistically significant. Previous spatial and temporal analyses show that petrochemical developments near the study lakes have not impacted PAH deposition to lake sediments studied. The current analysis has shown that forest fires and increased primary production do not seem to be affecting the concentration or source of PAHs in the study lakes, thus are not likely to be

affected by changing climatic conditions. Furthermore, this study shows that impacts from climate change are not confounding our interpretations of the lack of impacts from petrochemical development.

3.2. Introduction

3.2.1. Climate change at northern latitudes

Although climate change has been well documented across the globe, it is the northern regions that are expected to experience the largest scale of change (Macdonald et al. 2005). From 1948 to 2010 the mean annual air temperature in Canada increased 1.6˚C (Environment Canada), while mean annual temperatures in the Mackenzie Delta uplands, NWT have risen by more than 2.5˚C since the 1970s (Burns and Kokelj 2009). The northern regions of Canada have also undergone major development of petrochemical resources, which has been facilitated by warming temperatures and shorter winters. Concerns have been raised over the impacts from the combined stress of petrochemical development and climate change to aquatic ecosystems and thus freshwater resources in these regions. Petrochemical developments are associated with emissions of polycyclic aromatic hydrocarbons (PAHs), a group of organic contaminants that are generally produced during the incomplete combustion of organic material, and so can be generated from natural and anthropogenic activities. In addition to potential emission of PAHs from petrochemical developments, climate change has increased disturbance from forest fire events (Vergnoux et al. 2011, Golder Associates 2009), which may be an increasingly important source of PAH emissions in certain regions. Warming-mediated increases in chlorophyll *a* through rising primary production have also been documented (Thienpont et al. 2013b, Rühland et al. 2013), and could in turn alter how PAHs cycle through aquatic ecosystems, through increased fluxes of organic carbon that PAHs preferentially bind to, or by altering degradation conditions through longer ice-free periods. Understanding how impacts from climate change might affect PAHs in aquatic ecosystems is important to determining whether these impacts may confound interpretations of historical reconstructions of PAH deposition. This is particularly true in remote northern areas where impacts from climate change are

expected to increase whilst we are trying to understand the impacts of increasing petrochemical development on PAH delivery to the environment in these northern regions.

3.2.2. Study Objectives

Our goal in this study was to explore the relationship between climate and PAHs, and to determine whether changing climatic conditions as tracked through climate proxies over time, can explain variation in PAH deposition. Specifically we addressed two research objectives: 1) to determine whether PAH concentrations are tracking changing climatic conditions; and 2) to determine whether changes in PAH composition have tracked the timing of climate change. These objectives were accomplished by comparing the concentration and composition of PAHs in dated lake sediment cores from two different regions in the Northwest Territories that experience different climatic conditions to the timing of shifts in climate proxies over their respective cores' histories. In addition, we quantify the relationship between PAH concentrations and composition with climate proxies using correlation matrices.

3.2.3. PAHs in Aquatic Ecosystems and potential impacts from climate change

In remote lakes, such as those in the Northwest Territories, PAH input to aquatic ecosystems likely stem from general global atmospheric background, forest fires and the surrounding geology. These PAHs enter the aquatic environment through runoff, precipitation, dry deposition or diffusion. Their behaviour once in the water column depends largely on whether the PAHs are attached to particulate matter when they enter the water column, as well as the make up of that particulate matter. If PAHs diffuse across the air-water interface and become dissolved in the water column, that is they enter the water column not attached to particulate matter, they can then re-volatize to the atmosphere, are available for photodegradation, bind to particulate matter in the water column or remain dissolved in the water column. Due to their hydrophobicity, PAHs have a strong affinity for particulate matter, especially particulates that are organic in nature, and so the bulk of PAHs in water are typically in the particulate phase, with the remaining PAHs found dissolved in the water column (Zheng et al. 2013). For example, a

recent experimental study suggests that PAHs entering the water column through runoff from the surrounding land become more enriched in sediments fed by the runoff from carbon-rich soils than soils with half as much carbon (Zheng et al. 2013). Thus any alteration to the carbon in the catchment of lakes due to climate change (i.e. shifting vegetation communities or changes in vegetation decomposition conditions) may alter the delivery of PAHs to lake sediments. In addition, models by Mackay and Paterson (1991) indicate that general increases in carbon from warming-induced primary production in lakes will lead to an increase in PAH burial. This would suggest that any increase in primary production in the lake or within the catchment of the lake would affect the amount of PAHs delivered to the sediment. Interestingly however, Jeremiason et al. (1999) found that despite a 3-fold increase in the flux of organic carbon to the sediments throughout the history of the sediment core from a temperate lake in northwestern Ontario, PAH burial actually decreased over time. Thus exploring the relationship between PAH deposition and organic carbon under different environmental conditions would allow us to further our understanding of how changes to the organic carbon pool through warming-mediated primary production may impact PAH deposition.

As PAHs are produced during the combustion of organic material, forest fires are often considered to be sources of PAHs to the regional atmospheric pool of PAHs as well as to the pool of PAHs in soils and sediments. They can also be responsible for increases to organic carbon, delivered to aquatic ecosystems as pulses during rainfall events (Gabos et al. 2001, Vergnoux et al. 2011). Retene, an alkylated PAH in the C2-PHE/ANT homologous family, is produced from the burning of abietic acid in resin commonly found in pine trees, and is therefore often used as a chemical marker in lake sediments to indicate the occurrence of forest fires within the lake's catchment. For example, Gabos et al. (2001) found elevated levels of retene in surface sediments in northern Alberta after a series of forest fires. As a result, PAH delivery to lake sediments may increase with the expected increases in disturbance from forest fires due to climate change. Therefore, investigating how forest fires impact PAH deposition in remote locations will help to inform interpretations of PAH reconstructions and further our understanding of the natural variations in PAH deposition to remote lake sediments.

To the best of our knowledge this study represents the first attempt to explore the relationship between proxies of climate change and PAH deposition in lake sediments from across regions experiencing different climatic conditions in northwestern Canada.

3.3. Materials & Methods

3.3.1. Study site description

The six study lakes are located in the Taiga Plains ecoregion in the Northwest Territories, which runs from the Beaufort Sea in the north to the British Columbia and Alberta borders in the south. The study lakes are divided between two sub-regions, the Mackenzie Delta Uplands and the Cameron Hills, and have experienced different climate warming scenarios over the past century.

Occupying 9604 km^2 in the High Subarctic sub region, the Mackenzie Delta is the largest Delta in Canada and is characterized by thousands of ponds (most are smaller than 100 hectares). Stream channels are also abundant in the alluvial deposits that make up the active layer of much of the delta. These stream channels are part of the broad, flat floodplain network that makes up the delta. Soils in the region are underlain by hydrocarbon rich Cretaceous marine shale. The climate tends to be colder than the lower regions of the Taiga plains, with mean annual temperatures ranging from -5 to -11˚C (Government of the Northwest Territories 2008). Mean precipitation for the year ranges from 170 to 340mm, with most falling between June and October. December can see 0 to 0.6 mj m^{-2} day^{-1} of solar energy while June can see 22 to 22.4 mJ m^{-2} day^{-1} (Government of the Northwest Territories 2008). Previous studies suggest that warming in this region began in the 1950's (Thienpont et al. 2013a, 2013b).

The Cameron Hills cover an area of 1833km^2 and lie on a large plateau in the Lower Subarctic Taiga Plains with a range of elevation of 475 to 875 meters above sea level (ASL). The active layer of soil is made up of till deposits, with occasional deposits of glacial-alluvial and fluvial materials. Small ponds occur much less frequently in this region covering only about 3% of the region, compared to the Mackenzie Delta region where lakes cover about 36% of the region. Mean annual temperatures in the Cameron Hills area range from -3.5 to -9˚C, with the coldest temperatures occurring in January and

the warmest in July (Government of the Northwest Territories 2008). The Cameron Hills experience a shorter wet season, which runs from June to August, compared to the Mackenzie Delta (June to October), however mean annual precipitation ranges are similar with the Cameron Hills experiencing precipitation ranging from 230 to 350 mm per year. In addition, being further south the Cameron Hills experience more sunlight in the winter (0.2 to 1.5 mj m^{-2} day^{-1}) but similar sunlight in the summer months (21.5 to 22.5 mJ m^{-2} day^{-1}) compared to the Mackenzie Delta (Government of the Northwest Territories 2008). Changes related to climate warming began in this region around the 1910s to early 1940s, in some cases several decades before the Mackenzie Delta region (Thienpont et al. 2013a, 2013b).

3.3.2. Field Methods

In the Mackenzie Delta Uplands, Lake I20 (68.97˚N, 133.52˚W) (unofficial names) is downslope from abandoned natural gas drilling mud sumps (storage and containment pits for drilling waste, including borehole cuttings and drilling fluid), while the regional reference lake (C23 (68.00˚N, 133.51˚W)) is upslope from any petrochemical activity. It should be noted that petrochemical development has been shown to have no impact on PAH deposition in this region (see Chapter 2 of this thesis).

In the Cameron Hills (CAM) the two regional reference lakes (CamH2 (60.22˚N, 117.80˚W) and CamH7 (60.14˚N, 117.38˚W)) (unofficial names) are upstream of any petrochemical activity in the area, while lakes CamH3 (60.14˚N, 117.57˚W) and CamH5 (60.03˚N, 117.48˚W) (unofficial names) have at least one working or capped conventional gas or oil pump within their catchments. Previous analyses have shown that petrochemical developments have little or no impact on the deposition of PAHs in this region (see Chapter 2 of this thesis).

Sediment cores were taken from MDU lakes in August 2009 and July 2010, and from CAM lakes in September 2012, with a Glew gravity corer (1989) and extruded onsite at a resolution of 0.5-1.0 cm with a Glew vertical extruder (1989). Sediment intervals were placed immediately into Whirl-Pak© sample bags and frozen until analysis.

3.3.3. Laboratory Methods

Sediment radiochronologies for all cores were estimated following standard ^{210}Pb radioisotopic methods, and the constant rate of supply model (Appleby 2002) was used to calculate a dating profile for all cores except CamH7, which was calculated using the constant flux/constant sedimentation model, (Robins et al. 1978), as it was found to have a better fit. All samples were spiked with recovery standards (either the appropriate deuterated or ^{13}C-labeled USEPA priority 16 PAHs) as part of the stringent quality assurance and quality control methods that were applied to all samples in all regions to ensure data were comparable, though extraction and clean-up methods were slightly different among regions. Samples were manually homogenized and water removed with the addition of either Na_2SO_4 or Agilent brand Hydromatrix®. PAHs were then extracted from the lake sediments using either Soxhlet or accelerated solvent extraction (ASE, Dionex) and further cleaned-up using alumina or silica columns. All PAH concentrations were calculated using the isotope dilution method, which was used to correct for variability in extraction and analytical techniques. (For greater details on laboratory methods used, see Chapter 2 Materials and Methods)

Diatom slides were prepared according to standard methods outlined in Battarbee et al. (2001). Briefly, sediments were digested in a 1:1 (by weight) mixture of nitric and sulphuric acids and placed in a water bath (80˚C) for 3-4 hours to isolate diatoms and digest organic material, then rinsed over several days until a neutral pH (~7) was achieved. Diatoms were then mounted using Naphrax™ and analyzed using a Leica DMRB light microscope equipped with differential inference contrast filters under an oil immersion lens at 1000x magnification. Using standard diatom reference guides, such as Kramer and Lange-Bertalot (1997, 1999, 2000) diatoms were identified to the species level, and valves were counted per sediment interval in four out of six study lakes (I20, C23, CamH3, CamH7) to a minimum of 300 valves, with most counts ranging from 400 to 500 valves per interval.

The chlorophyll *a* (chl-*a*) content per sediment interval was inferred using visible reflectance spectroscopy according to methods described in Michelutti et al. (2005, 2010). This method has been shown to be an accurate reconstruction of trends in both chlorophyll *a* concentration and it's diagenetic products in lakes over time (Michelutti et

al. 2010). Freeze-dried sediments were sieved through 125µm mesh and analysed using FOSS NIRSystem (Model 6500) rapid content analyser.

Rock Eval pyrolysis was conducted on cores from 4 lakes: I20 and C23 in the Mackenzie Delta Uplands, and CamH2 and CamH5 in the Cameron Hills, to determine the contribution of the main three fractions of organic carbon to the total organic carbon pool. Samples underwent pyrolysis at 300˚C to release small volatile compounds in the S1 fraction, measured by flame ionisation detection (FID) and reported in mg hydrocarbons (HC)/g sample. The temperature was then increased at a rate of 25˚C/min until 650˚C, releasing the S2 fraction of organic carbon, and is also measured by FID and reported in mg HC/g sample. The S2 fraction is typically made up primarily of lipid cell wall material from photosynthetic algae (Sanei et al. 2005), and is thus usually considered the algal derived fraction of organic material. Throughout the pyrolysis step oxygen containing carbon molecules, namely CO and CO_2, were continuously measured by infrared spectroscopy and make up the S3 (individually the S3CO (mg CO/g) and $S3CO_2$ (mg CO_2/g) fractions). Finally, residual carbon (RC) was released by heating the samples to 850˚C in oxic conditions. RC was measured by infrared spectroscopy and reported in per cent weight of RC in the sample.

3.3.4. Data analysis

Relative abundances were calculated using the following calculation:

$$\%\ \text{Individual PAH}_{\text{Rel. Abun.}} = (\text{Individual PAH}/\Sigma\text{PAH}) \times 100 \quad \text{(eq. 1)}$$

The sum of Parent PAHs (ΣParent) included the following compounds: acenaphthylene (ACY), acenapthene (ACE), fluorene (FLU), anthracene (ANT), phenanthrene (PHE), fluoranthene (FLA), pyrene (PYR), banz[a]anthracene (BaANT), chrysene (CHR), benzo(b)fluranthene (BbFLA), benzo(k)fluoranthene (BkFLA), benzo[a]pyrene (BaPYR), indeno(1,2,3-cd)pyrene (IcdP), dibenz(a,h)anthracene (DahA) and benzo(g,h,i)perylene (BghiP). The sum of Alkyl PAHs (ΣAlkyl) used included all C1-, C2- and C3-fluorenes, C1-, C2-, C3and C4-Phenanthrene/Anthracenes, C1-, C2-, C3and C4-Fluoranthene/Pyrenes, C1-, C2-, C3and C4-Benz[a]anthracene/Chrysenes and C1- and C2-Benzofluoranthene/Benzopyrenes. ΣPAH includes all of the above-mentioned

PAHs (i.e. includes both parent and alkyl PAHs). Naphthalene (NAPH) and its alkylated counterparts were left out of all statistical analyses and calculations of ΣParent and ΣAlkyl due to highly variable recovery rates.

Principle components analyses (PCA) were conducted on the relative abundance of diatom species (Coleman, unpublished data) and of PAH compounds in sediments intervals throughout the history of the cores. In order to mitigate skewed results from large discrepancies in relative abundances, the following criteria were followed for inclusion of diatoms and PAHs into their respective analyses: the diatom species or PAH compound had to be found at 2% or greater abundance in two or more intervals within each core to be kept in the PCA analysis. The relative abundances of diatom species were then square-root transformed while relative abundance of PAH compounds were log transformed before conducting the PCA.

In order to determine if PAH parameters (sum of Parent PAHs, sum of Alkyl PAH, the P:A ratio (which was calculated as the sum of Parent PAHs over the sum of Alkyl PAHs) and the axis 1 and axis 2 scores from the PCA on PAH relative abundances described above) are tracking changes in climatic conditions, correlations were conducted on all PAH parameters and all climate proxies (VRS chl-*a*, Rock Eval fractions, percent organic carbon (%OC), and axis 1 and 2 scores from the PCA on diatom species relative abundances described above). Not all PAH analyses were done on the same sediment intervals as climate proxy analyses, with PAHs being analyzed at the coarsest temporal resolution (due to the amount of material needed for PAH analysis). Therefore intervals used for the correlation analysis were binned by which intervals were used for PAH analysis. Wherever sediments used for climate proxy analyses did not overlap with bins (i.e. sediments used for PAH analysis), climate proxies were interpolated and averaged over the sediment intervals included in the bins where multiple intervals were used for individual bins. Assumptions of normality were not met (according to the Shapiro-Wilk test), thus Spearman's non-parametric correlation was used to generate correlation matrices for each lake (Tables 3.1-3.6). In addition, due to the number of variables in the correlation analyses, the alpha values used to determine statistical significance were corrected following the Bonferonni procedure, by dividing the typical p-value used (0.05) by the number of variables included in the individual correlation matrices.

All data were analysed using R statistical software, while specific analyses were done with the following add-on packages: PCAs were conducted using the vegan package and Spearman correlations using the Hmisc package.

3.4. Results

3.4.1. Mackenzie Delta Uplands

In Lake I20 the sum of Parent PAHs (ΣParent) ranges from 25-92 ng/g dw, which is less than the sum of Alkyl PAHs (ΣAlkyl), ranging from 161-583 ng/g dw (Fig. 3.1). Organic carbon content in I20 is variable over time ranging from 23.7-28.7%, and does not follow any clear trend throughout the core (Fig 3.8). The PCA axis scores from the analysis of PAH relative abundance indicate that the composition of PAHs in I20 is fairly constant until about 8cm down, and becomes more variable towards the surface (Fig. 3.1). Shifts in diatom assemblage and inferred chlorophyll *a* content were used in a previous study by Thienpont et al. (2013a) to establish that the onset of changing climatic conditions occurred in this lake around the late 1950s. All correlations in I20 are not statistically significant (Table 3.1). Axis scores from the diatom assemblage principle components analysis are not significantly correlated with any of the PAH concentration parameters, nor do the axis score from the PCA of PAH relative abundance show any statistically significant correlations to any of the climate proxies (diatom assemblage PCA axis scores, VRS Chl-*a*, %OC, Rock Eval fractions) (Table 3.1).

In Lake C23, ΣAlkyl (ranging from 177-992 ng/g DW), was greater than ΣParent (ranging from 50-121 ng/g DW) (Fig. 3.2). Organic carbon content was fairly consistent throughout the core, with an average % organic carbon (%OC) of 12.4% for the history of the core (Fig. 3.8). The composition of PAHs in C23 sees a definite shift, indicated by the shift in PCA axis 2 at 6.75cm (Fig. 3.2), which according to the ^{210}Pb dating corresponds roughly to the turn of the 20th century. The onset of changing climatic conditions in this region, according to diatom and cladocera proxies, occurred in the early 1950s, as indicated in Thienpont et al. (2013a). The Spearman correlation analysis indicates none of the correlations between climate and PAH parameters are statistically significant with the exception of the sum of parent PAHs and organic carbon. ΣParent

shows a statistically significant negative correlation with %OC (ρ=-0.9 p=0.002) (Table 3.2).

3.4.2. Cameron Hills

In CamH2 ΣParent range from 44-164 ng/g dw, while ΣAlkyl ranged from ND (non-detection) to 737 ng/g dw (Fig. 3.3). Organic carbon was constant near the bottom of the core, until about 17cm after which it became more variable, showing a general decline of about 3% towards the top of the core (Fig. 3.8). PCA axis 1 and 2 scores indicate that PAH composition is stable near the bottom of the core until 10 cm (~1920), when the composition becomes a lot more variable towards modern sediments (Fig. 3.3). Increasing inferred chlorophyll *a* content (Coleman, unpublished data) suggests that warming began around the 1920s in this lake. According to the Spearman correlation analysis, none of the climate parameters correlate significantly with any of the PAH parameters (Table 3.3).

Concentration profiles for CamH5 indicate that ΣParent ranges from 11-534 ng/g dw, while ΣAlkyl ranges from 22-2268 ng/g dw (Fig.3.5). Unlike other lakes in the Cameron Hills, the contribution of parent PAHs to the overall inventory of PAHs varies widely from about 1% to about 96%, while all other lakes are in the 15-35% range with only a few intervals straying outside that range. Organic carbon content is stable throughout the core, with a mean of 33.2% (Fig. 3.8). According to inferred chlorophyll *a* content (Coleman, unpublished data), S2 (algal derived carbon) and extrapolated ^{210}Pb dating the onset of shifts in biological communities due to climate change in this lake began around the early 1900's. ΣParent shows a statistically significant positive correlation with the S3 fraction of Rock Eval pyrolysis (ρ=0.87 p=0.0025), while all other correlations are not statistically significant or show any clear trend (Table 3.4).

CamH3 also shows greater alkyl concentrations (65-558 ng/g dw) than parent (19-164 ng/g dw) (Fig.3.4). The proportion of parent PAHs peaks at 8cm and declines thereafter. Two thresholds for changes in biological parameters (shift in diatom communities and increases in inferred chl-*a*) (Coleman, unpublished data) were found in this lake: one in the early 1940s (11cm) and again in the early 1970's (5cm) (Fig. 3.4).

Organic carbon slowly increases at the bottom of the core until 15cm at which point %OC declines by about 3.5% and continues to decline slowly towards the surface of the core (Fig. 3.8). PAH composition is highly variable throughout the whole core, exhibiting no clear trend over time, which is reflected in the variability of the axis 1 and 2 scores from the PCA on the relative contribution to the environmental inventory of the most abundant PAHs (Fig. 3.4). Our Spearman correlation analysis indicates that none of the correlations between the PAH and climate parameters are statistically significant (Table 3.5).

CamH7 has the greatest average concentration of parent PAHs in the Cameron Hills region (120 ng/g dw), which generally increase in concentration towards the surface of the core (Fig. 3.6). Alkyl concentrations are also greatest in the region ranging from 327-2850 ng/g dw, with an average of 1090 ng/g dw (Fig. 3.6). %OC decreases steadily towards the surface over the length of the core, from about 39% at the bottom of the core to barely 30% in surface sediments (Fig. 3.8). Shifts in the diatom community assemblage and increases in the inferred chl-a content (Coleman, unpublished data) suggest that climatic conditions began changing in this region around 1911. Correlations between PAH and climate parameters in this lake are not statistically significant, with the exception of ΣParent and %OC, which is statistically significant (ρ=-0.89 p=0.002) (Table 3.6).

3.5. Discussion

Comparing PAH deposition histories with climate change proxies across regions that have experienced different warming scenarios has allowed us to explore the broader relationship between the impacts from the changing climate. More specifically we investigated the relationship between the impacts from potential increased forest fire disturbance and primary production as well as changes to the different fractions of organic carbon, with the concentration and composition of PAHs in lake sediments in the NWT, which has to date been largely uncharacterized.

As seen in Tables 3.1-4, inferred chlorophyll *a* content and algal-derived carbon (S2 fraction of the Rock Eval pyrolysis) do not significantly correlate with organic carbon in our study lakes from the Northwest Territories. This finding agrees with lakes in the Athabasca oil sands where an increase in primary production was found beginning

around the late 1970s across lakes near open pit mining activity (Kurek et al. 2013), while organic carbon remained generally stable throughout their core's histories (figure 3.8, NE20, SW35, SE22). Thus, the increases we are seeing in inferred chlorophyll *a*, and therefore primary production, due to climate warming, are not likely to affect total sedimentary organic carbon deposition.

Our analysis clearly shows that the changes in the concentration of PAHs across all lakes have not followed the changes in either the diatom assemblage or the increase in inferred chlorophyll *a*. In addition, %OC only correlates significantly with ΣParent in 2 out of our 6 study lakes, while ΣAlkyl does not significantly correlate with %OC in any of our study lakes. This agrees with the findings of Jeremiason et al. (1999), who examined the relationship of organic carbon to PAHs in lake sediments from Lake 227 in the Experimental Lakes area in northwestern Ontario, Canada. Lake 227 has no known point source of PAHs, i.e. it is similar to the current study lakes in that they are only subject to general atmospheric background sources. The authors found that despite an almost 2-fold increase in sediment mass accumulation and a 3-fold increase in organic carbon due to experimental additions of phosphorus, PAHs generally decreased over time after an initial spike immediately following the start of phosphorus additions. This may suggest that after the initial response seen in PAH concentration to the onset of increasing organic carbon (and sediment mass accumulation), the accumulated pool of dissolved PAHs is scavenged from the water column and the flux of PAHs to the sediment subsequently declines. Although, this would also depend on the residency time of water and PAHs in these lakes, which would warrant further study. Additionally, Jeremiason et al. (1999) suggested that shifts over time in biota (i.e. phytoplankton assemblage) to dominant species that have lower bioconcentration factors of PAHs may also account for fewer PAHs being scavenged from the water column over time and making their way to the sediments. Although our study does not look at phytoplankton assemblages, we do see major shifts in benthic diatom assemblages from one characterized by cold-tolerant benthic species, to more planktonic species that prefer more ice-free conditions, and may likewise have an effect on the movement of PAHs to the sediments. It is possible that planktonic diatom species with lower bioconcentration factors are likely to scavenge fewer PAHs from the water column and consequently deposit fewer PAHs to the

sediment as they die and sink to the bottom of the lake. This in turn frees up the PAHs to leave the lake through connected lakes and rivers. Alternatively, in closed basins such as our study lakes, PAHs that remain in the water column have a greater chance of being degraded by photooxidation or leaving the system through volatilization. The concentration of PAHs in the study lakes generally decline over the history of the sediment cores and thus may be subject to similar mechanisms of removal. It should be noted, however, that although global trends in PAHs have begun to rise again in the past 10 years after a prolonged period of decline since the late 1970s, and that these types of changes are often delayed in northern remote regions, such as those where our lakes are located (Macdonald et al 2005).

Previous analysis (see chapter 2 of this thesis) showed that despite being close to petrochemical developments, the PAH deposition in the study lakes have not been affected by this development. Forest fires are often cited as sources of local background PAHs in remote areas that are not affected by anthropogenic point sources such as heavy traffic or industrial activity (Gabos et al. 2001, Denis et al. 2012). Studies suggest that the changing climate has increased fire disturbance, particularly in northern ecosystems (Golder Associates 2009), and is likely to increase the emission of pyrogenic PAHs to the environment. However, this trend was not observed in our study lakes. The contribution of retene to the alkyl profile throughout the history of the sediments cores in our study lakes is relatively low in both regions. The exception to this is in CamH5, which spikes to about 35% at 5cm, and is also reflected in a spike in the concentration of retene at this time. Gabos et al. (2001) found a similar contribution of retene (roughly 40%) in surface sediments at a site burned by forest fires in northern Alberta. It should be noted however, that the concentration of retene they found was two orders of magnitude greater than what was found in CamH5, although background level at their reference site was also far greater than any concentrations we found at either the CAM of the MDU sites. Interestingly we see a slight increase in organic carbon at the same depth that we find the spike in retene in CamH5, compared to the average %OC prior to this. Although this may indicate a local fire, %retene declines immediately following this to below 5% abundance, which is similar to the average abundance of retene in all the other study lakes. This would suggest that fires are not likely large contributors to the PAH

deposition history in these lakes, and any warming-mediated increases in forest fire disturbance is not likely to have an immediate appreciable effect on the PAHs deposited to lake sediments in these regions.

Our analysis represents the first attempt to characterize the relationship between impacts from climate change and PAH deposition to lake sediments in northwestern Canada. We clearly show that while organic carbon content was expected to increase in lake sediments due to increasing primary production from climate warming, this is not the case in the current study lakes. In addition, we show that so far climate change has not increased PAH burial, nor has it changed the composition of PAHs appreciably due to forest fire disturbance. Although more detailed studies are required to determine the source of variations in PAH profiles from individual lakes, as well as to further investigate linkages between climate and PAH deposition, our study has expanded our understanding of PAH deposition patterns in these regions, and has shown that, to date, climate change has not confounded our interpretation of PAH sources in these areas.

Table 3.1: Spearman Correlation results for lake I20.

		VRS Chl-*a*	Diatom PC1	Diatom PC2	S1	S2	S3	TOC	%RC	% OC
%OC	*Rho*	0.69	-0.67	0.43	0.13	0.28	0.33	0.29	0.31	1
	p-value	0.26	0.47	0.70	0.66	0.68	0.53	0.88	0.96	1
ΣParent	*p-value*	-0.22	0.19	-0.24	0.12	0.43	0.28	0.40	0.40	0.20
	p-value	0.48	0.56	0.46	0.71	0.16	0.38	0.20	0.20	0.54
ΣAlkyl	*Rho*	-0.53	0.39	-0.49	0.22	0.58	0.32	0.48	0.61	0.22
	p-value	0.08	0.21	0.11	0.48	0.05	0.31	0.11	0.04	0.48
P:A	*Rho*	0.17	-0.07	-0.03	-0.49	-0.31	-0.58	-0.37	-0.44	0.06
	p-value	0.60	0.83	0.91	0.11	0.33	0.05	0.24	0.15	0.84
PAH PC1	*Rho*	-0.01	-0.21	-0.42	0.51	0.03	0.29	0.20	0.15	0.39
	p-value	0.98	0.51	0.17	0.09	0.91	0.37	0.53	0.63	0.21
PAH PC2	*Rho*	-0.20	0.23	-0.17	0.22	-0.02	-0.01	-0.01	0.14	-0.10
	p-value	0.54	0.47	0.60	0.50	0.95	0.96	0.96	0.66	0.76

Table 3.2: Spearman Correlation results for lake C23.

		VRS Chl-*a*	Diatom PC1	Diatom PC2	S1	S2	S3	TOC	%RC	% OC
%OC	*rho*	0.69	-0.67	0.43	0.13	0.28	0.33	0.29	0.31	1
	p-value	0.06	0.07	0.29	0.76	0.51	0.42	0.49	0.45	1
ΣParent	*rho*	-0.69	0.71	-0.29	-0.20	-0.46	-0.48	-0.43	-0.38	-0.90
	p-value	0.06	0.05	0.49	0.63	0.26	0.23	0.29	0.35	**0.002***
ΣAlkyl	*rho*	-0.64	0.67	-0.10	-0.08	-0.32	-0.24	-0.29	-0.12	-0.64
	p-value	0.08	0.07	0.82	0.84	0.43	0.57	0.49	0.78	0.08
P:A	*rho*	0.07	-0.10	-0.07	-0.22	-0.11	-0.31	-0.12	-0.25	0.02
	p-value	0.87	0.82	0.87	0.61	0.80	0.46	0.78	0.55	0.96
PAH PC1	*rho*	0.14	-0.17	-0.05	-0.35	-0.11	-0.19	-0.14	-0.17	0.36
	p-value	0.74	0.69	0.91	0.40	0.80	0.65	0.74	0.69	0.38
PAH PC2	*rho*	0.50	-0.48	-0.29	0.06	-0.30	-0.24	-0.29	-0.60	0.12
	p-value	0.21	0.23	0.49	0.89	0.47	0.57	0.49	0.12	0.78

* indicates statistical significance

Table 3.3: Spearman Correlation results for lake CamH2.

		VRS Chl-*a*	S1	S2	S3	TOC	%RC	% OC
% OC	*rho*	-0.35	0.51	0.70	0.081	0.75	0.71	1
	p-value	0.30	0.11	0.02	0.003	0.007	0.01	1
ΣParent	*rho*	-0.29	0.08	0.21	-0.08	0.22	0.32	-0.04
	p-value	0.38	0.81	0.54	0.81	0.52	0.34	0.92
ΣAlkyl	*rho*	0.45	-0.16	-0.15	-0.30	-0.16	-0.19	-0.40
	p-value	0.17	0.63	0.65	0.37	0.63	0.57	0.22
P:A	*rho*	-0.74	0.17	0.48	0.13	0.54	0.66	0.37
	p-value	0.01	0.61	0.13	0.71	0.09	0.03	0.26
PAH PC1	*rho*	-0.21	-0.44	-0.57	-0.51	-0.56	-0.43	-0.46
	p-value	0.54	0.18	0.06	0.11	0.07	0.19	0.15
PAH PC2	*rho*	0.61	-0.25	-0.34	-0.05	-0.35	-0.49	-0.03
	p-value	0.05	0.47	0.31	0.89	0.28	0.12	0.94

Table 3.4: Spearman Correlation results for lake CamH5.

		VRS Chl-*a*	S1	S2	S3	TOC	%RC	% OC
%OC	*rho*	0.50	0.48	-0.27	0.73	0.38	-0.18	1
	p-value	0.17	0.19	0.49	0.02	0.31	0.64	1
ΣParent	*rho*	0.80	0.63	-0.50	0.87	0.07	-0.43	0.82
	p-value	0.01	0.07	0.17	<**0.003***	0.86	0.24	0.01
ΣAlkyl	*rho*	-0.33	-0.42	-0.05	-0.45	0.03	0.60	-0.38
	p-value	0.38	0.26	0.90	0.22	0.93	0.09	0.31
P:A	*rho*	0.63	0.62	-0.20	0.65	-0.10	-0.63	0.50
	p-value	0.07	0.08	0.61	0.06	0.80	0.07	0.17
PAH PC1	*rho*	0.32	0.43	-0.05	0.32	-0.20	-0.68	0.28
	p-value	0.40	0.24	0.90	0.41	0.60	0.04	0.46
PAH PC2	*rho*	-0.17	0.00	0.12	-0.23	0.05	0.15	0.12
	p-value	0.67	1.00	0.76	0.54	0.90	0.70	0.76

* indicates statistical significance

Table 3.5: Spearman Correlation results for lake CamH3.

		VRS Chl-*a*	Diatom PC1	Diatom PC2	% OC
%OC	*rho*	-0.36	0.95	0.88	1
	p-value	0.38	<**0.005***	<**0.005***	1
ΣParent	*rho*	0.72	-0.55	-0.64	-0.89
	p-value	0.23	0.06	0.29	0.06
ΣAlkyl	*rho*	0.01	0.26	-0.08	-0.15
	p-value	0.26	0.16	0.57	0.16
P:A	*rho*	0.42	-0.55	-0.21	-0.26
	p-value	0.01	0.87	0.91	0.82
PAH PC1	*rho*	-0.31	0.10	-0.02	-0.04
	p-value	0.10	0.65	0.46	0.42
PAH PC2	*rho*	-0.41	0.60	0.30	0.36
	p-value	0.05	0.57	0.69	0.38

*indicates statistical significance

Table 3.6: Spearman Correlation results for lake CamH7.

		VRS Chl-*a*	Diatom PC1	Diatom PC2	%OC
%OC	*rho*	-0.81	0.81	0.82	1
	p-value	<**0.005***	<**0.005***	<**0.005***	1
ΣParent	*rho*	0.72	-0.55	-0.64	-0.89
	p-value	0.02	0.10	0.05	<**0.005***
ΣAlkyl	*rho*	0.01	0.26	-0.08	-0.15
	p-value	0.99	0.47	0.83	0.68
P:A	*rho*	0.42	-0.55	-0.21	-0.26
	p-value	0.23	0.10	0.56	0.47
PAH PC1	*rho*	-0.31	0.10	-0.02	-0.04
	p-value	0.38	0.78	0.96	0.91
PAH PC2	*rho*	-0.41	0.64	0.30	0.36
	p-value	0.24	0.05	0.40	0.31

* indicates statistical significance

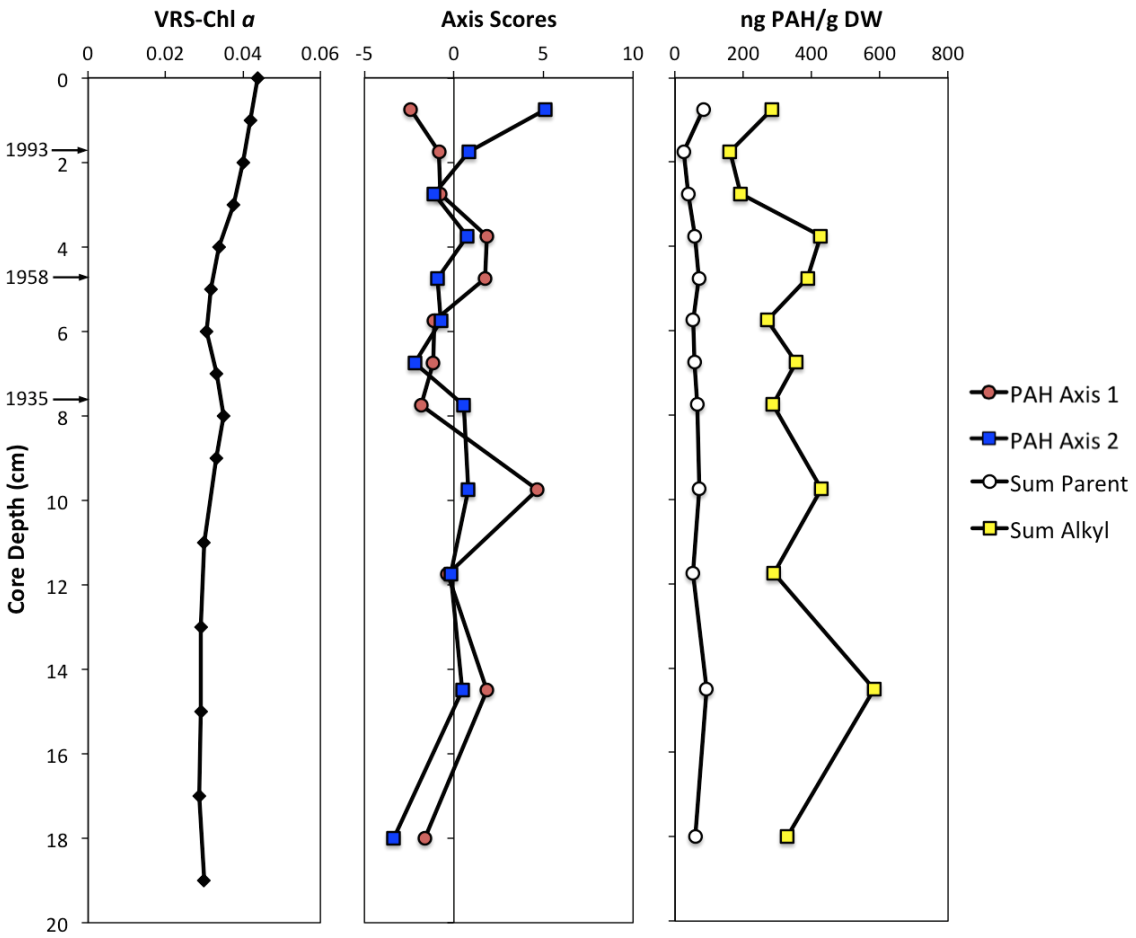

Figure 3.1: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake I20 (Mackenzie Delta Uplands, NWT).

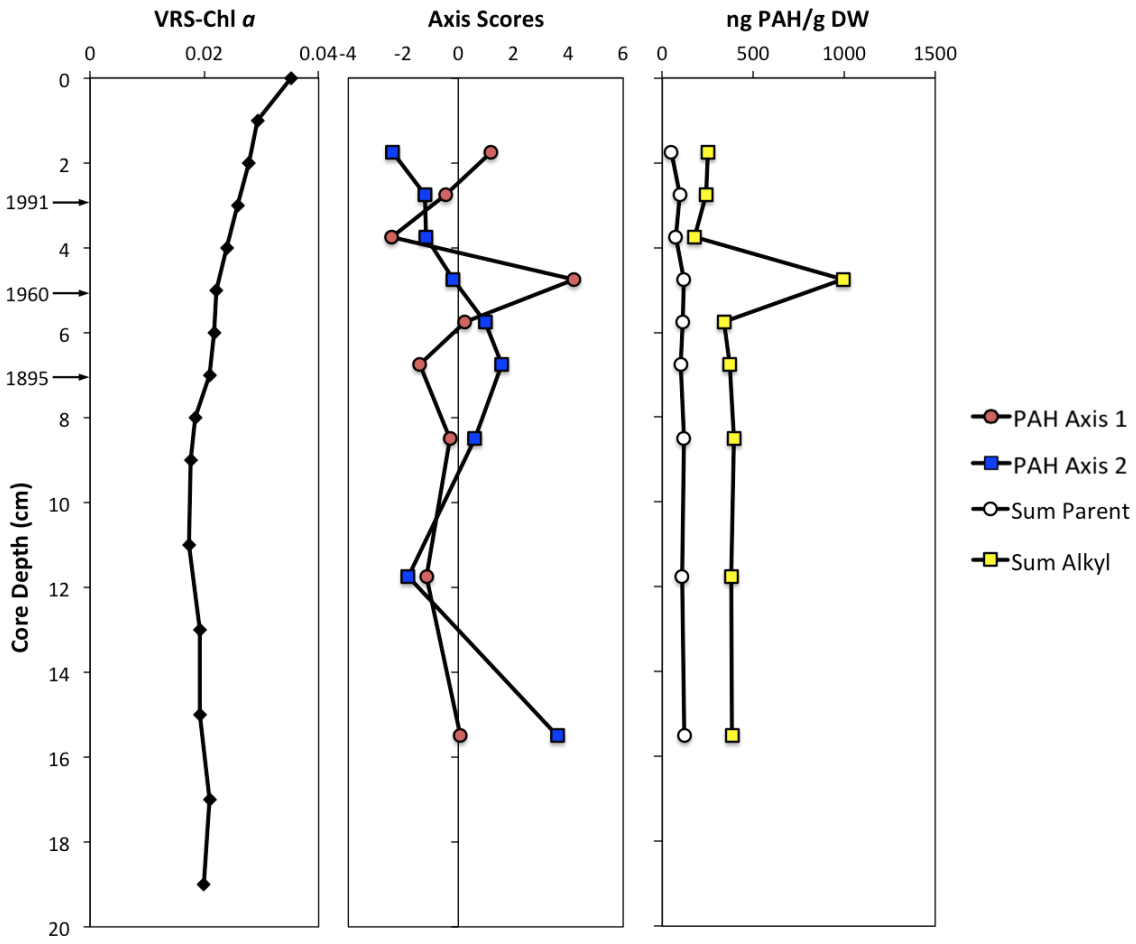

Figure 3.2: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake C23 (Mackenzie Delta Uplands, NWT).

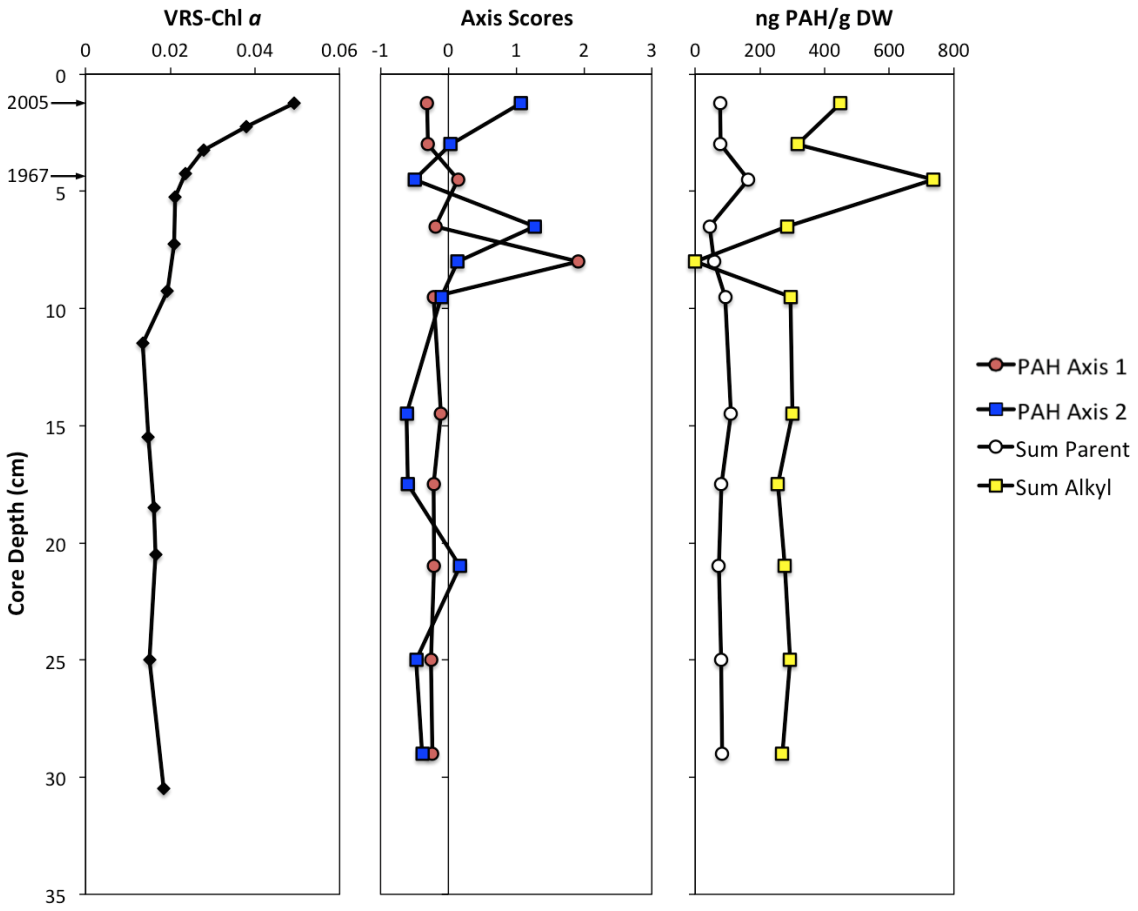

Figure 3.3: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake CamH2 (Cameron Hills, NWT).

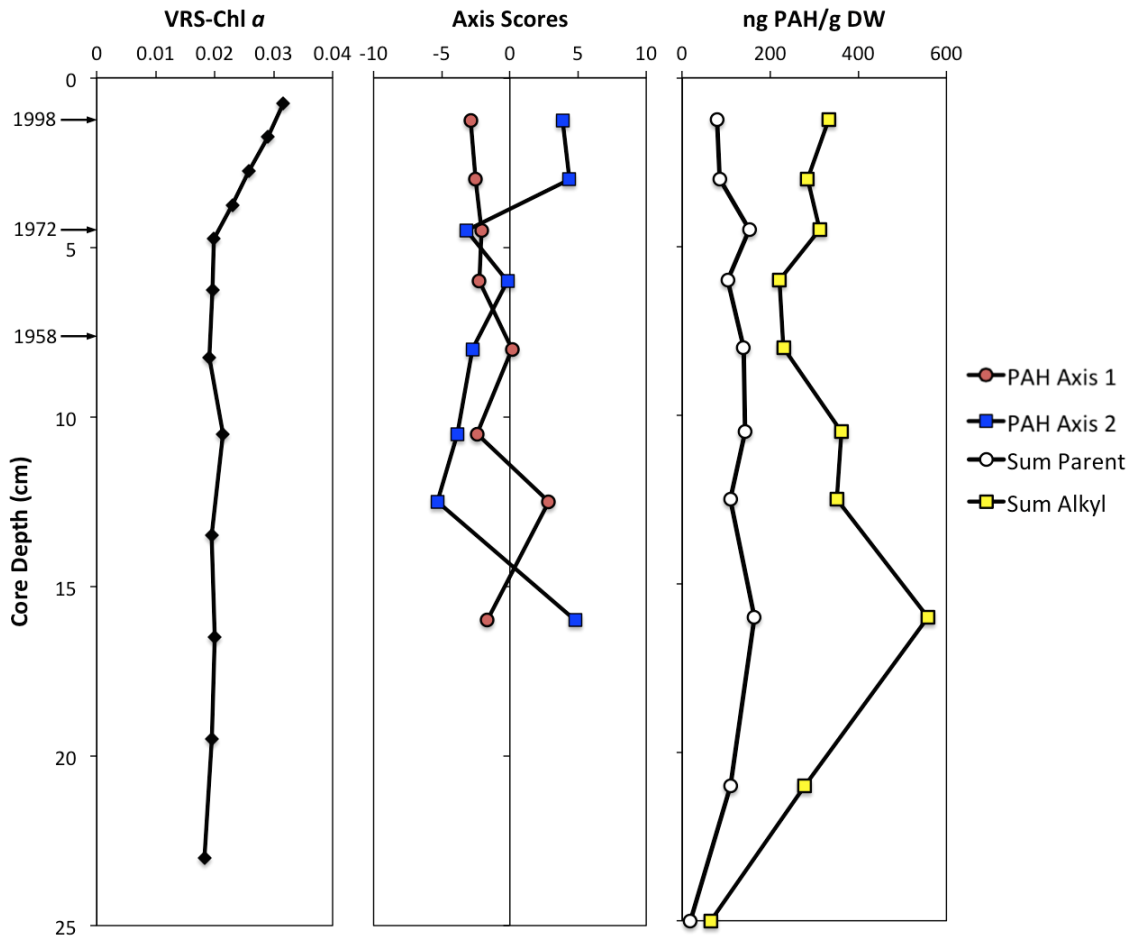

Figure 3.4: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake CamH3 (Cameron Hills, NWT).

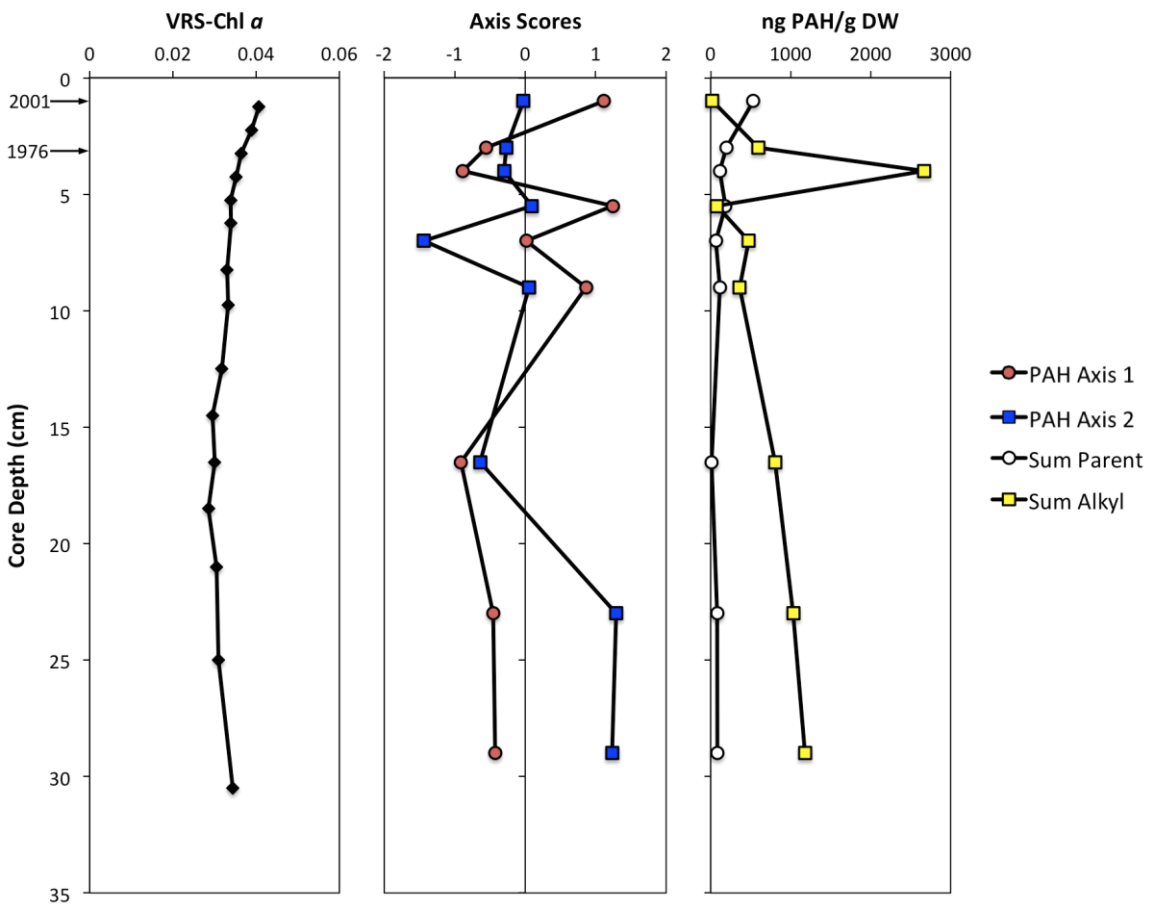

Figure 3.5: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake CamH5 (Cameron Hills, NWT).

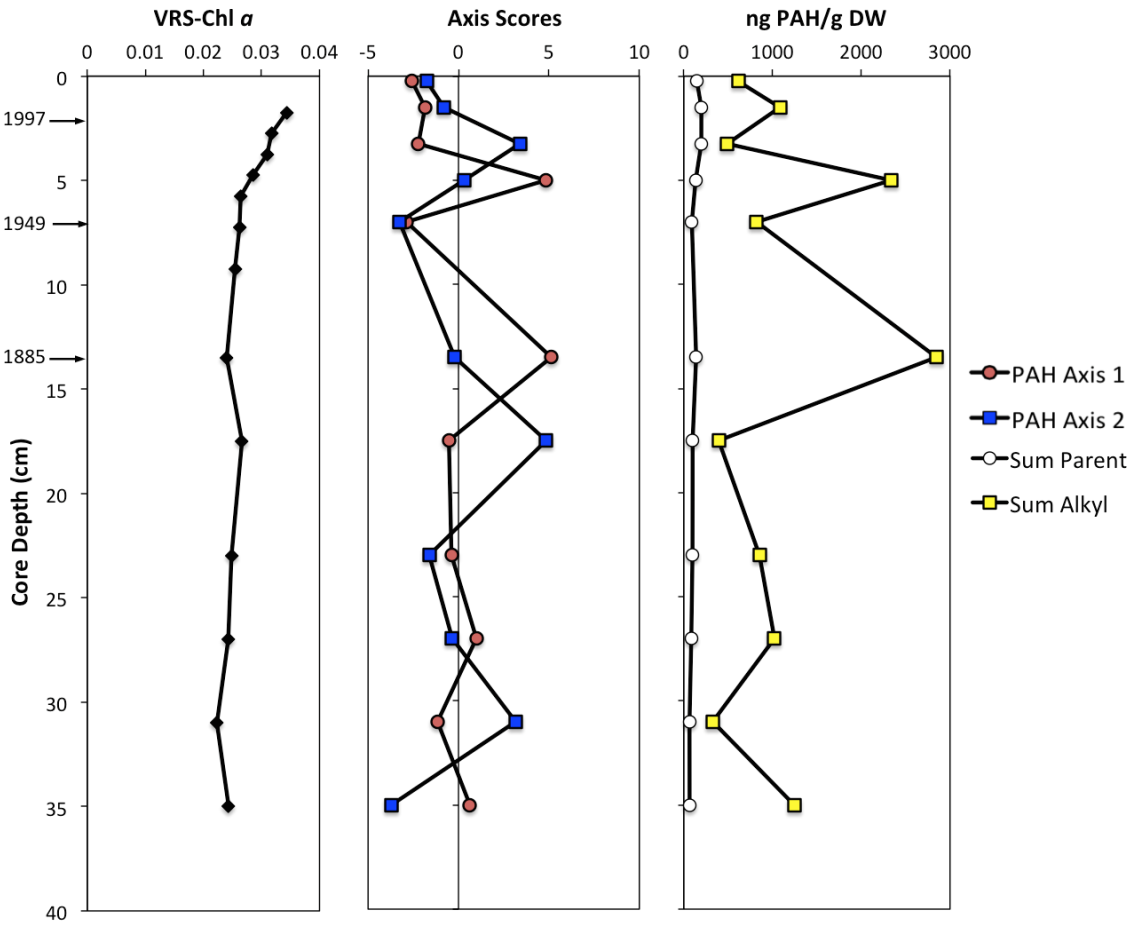

Figure 3.6: VRS-Chl *a*, axis scores from principle components analysis of relative abundance of most abundant PAHs and PAH concentrations in lake CamH7 (Cameron Hills, NWT).

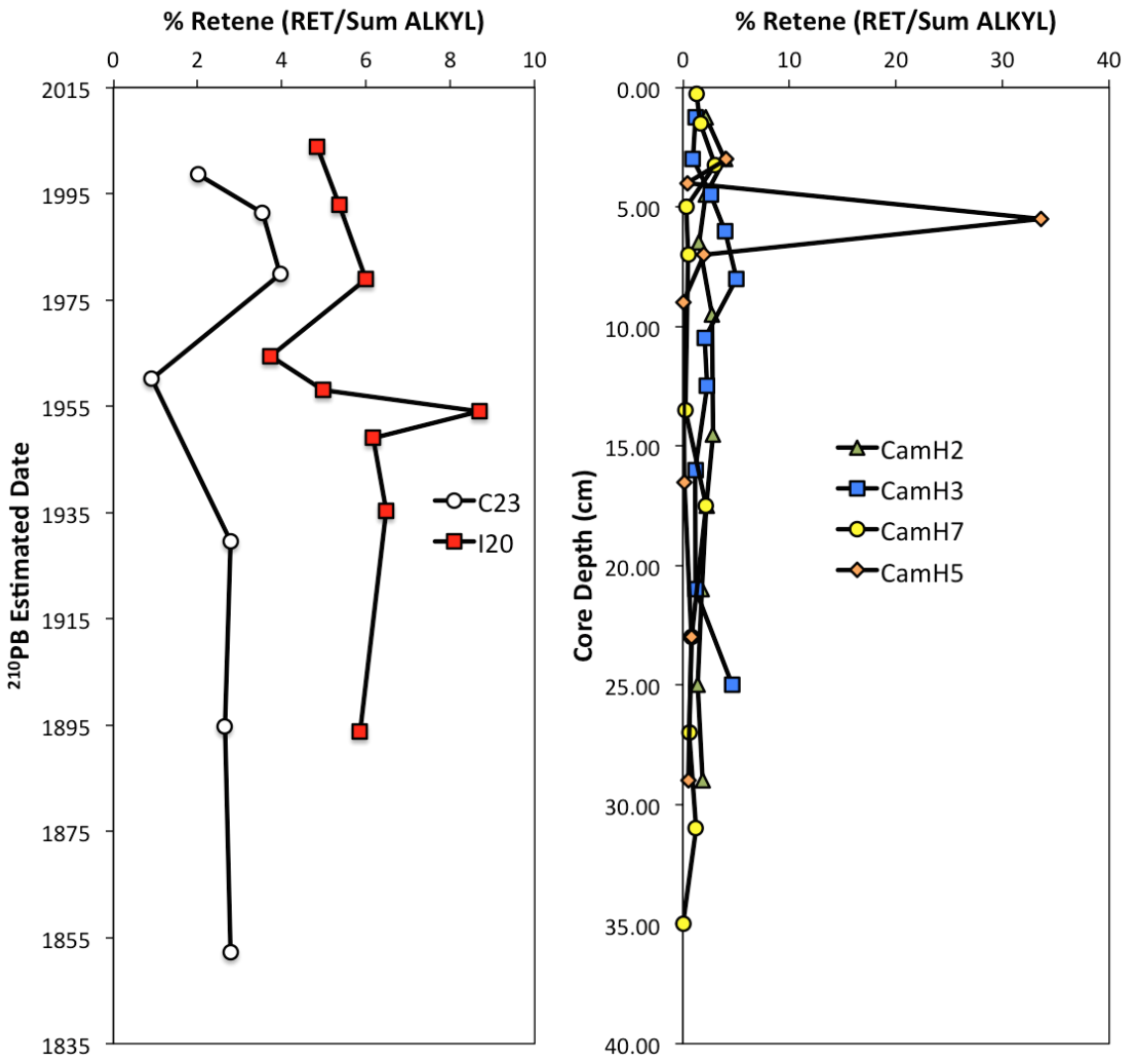

Figure 3.7: Per cent relative abundance of retene (% Retene) in the total concentration of alkyl PAHs.

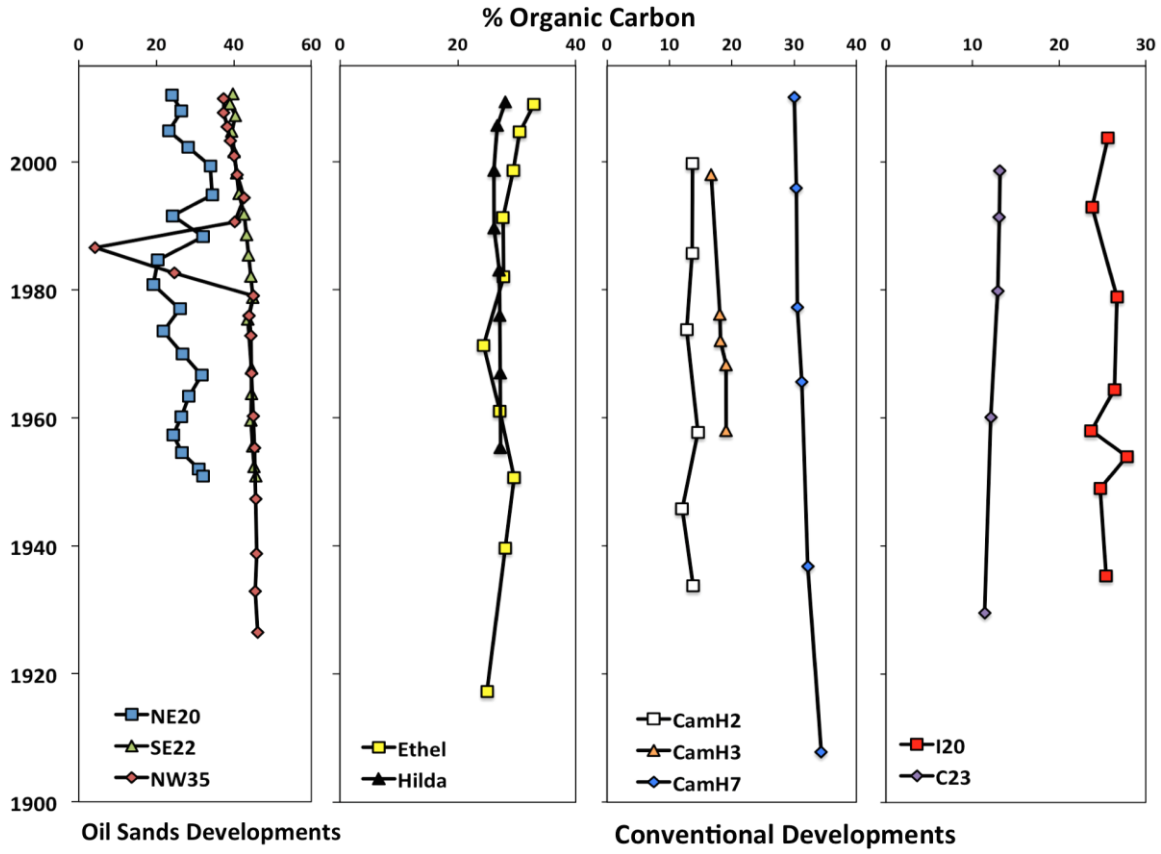

Figure 3.8: Organic carbon for all lakes, with the exception of CamH5 (due to poor dating profile).

CHAPTER 4. Conclusions

4.1. General Conclusions

4.1.1. Chapter 2

Although a few studies have looked at the impacts of petrochemical development on PAH deposition in lakes, this thesis represents the first attempt to directly compare the effects of different types of petrochemical development on the concentration and sources of PAHs in lake sediments. Sedimentary concentrations of PAHs generally declined over time in lakes of the Northwest Territories, where conventional sources are the chief deposits of petrochemicals. In the oil sands regions of Alberta, sedimentary concentrations of PAHs were generally found to be increasing, especially in the Athabasca region. The temporal analyses revealed that the major shifts seen in all three Athabasca lakes near open-pit mining, as well as the shifts in some of the Cold Lake sediments coincide with the onset of oil sands extraction in these regions, while variations in concentrations of PAHs in all 9 lakes near the different conventional developments revealed no clear trend coincident with the timing of their individual developments. The degree of impact to PAH deposition was largest in the Athabasca oil sands region, compared to the other three regions, which is likely due to the immense scale of land-use change that has occurred in the course of open-pit mining for oil sands. This result agrees with previous studies, such as Kurek et al. (2013) and Jatuzy et al. (2013), who have suggested that large-scale expansion of the area being surface mined is leading to greater scattering of oil sands particles from wind erosion. Additionally, the greater scale of oil production has led to greater amounts of bitumen being extracted and upgraded to marketable petrochemical products, which is a likely source of pyrogenic PAHs in the region. This study, as well as others, would suggest that the delivery of PAHs to aquatic ecosystems will continue to increase in the region as surface-mined areas expand.

Both spatial and temporal analyses indicate that there are regional differences in the composition of PAHs being deposited to sediments in the 14 study lakes. The lakes near conventional developments have seen virtually no changes to their composition and

have been subject to mixed sources over time. Low retene concentrations in all study lake sediments in the NWT suggest that forest fires do not contribute large amounts to the PAH profiles in these regions, but rather the chief contributors to their PAH profiles are likely global background sources that are generally pyrogenic in origin, as well as significant inputs of petrogenic PAHs from their surrounding hydrocarbon-rich catchments. It should be noted that although petrogenic PAHs are evident in the slightly elevated levels of alkylated PAHs in the sediments, they don't show any evidence of increasing over time. If these regions were experiencing significant impacts from land disturbance due to petrochemical development, we would expect to see an increase in petrogenic PAHs in particular, as PAHs were flushed in from the surrounding hydrocarbon-rich catchments, and this simply not what has been documented in these regions.

Lakes near oil sands extraction, on the other hand, have seen significant changes in their composition and sources over time, that coincide with the onset of their respective oil sands developments. Sources in the Athabasca region were historically dominated by pyrogenic sources in the early 1950s when coal and wood were the chief fuel sources. Subsequently, beginning in the first half of the 1970s, sources shifted to more petrogenically sourced PAHs. Meanwhile, in Cold Lake heavier parent PAHs had historically originated from petrogenic and lighter parent PAHs from pyrogenic sources. But as the area became populated and exploration and exploitation of the area's oil sands began, the sources of the heavier compounds shifted to more pyrogenically sourced PAHs. Vehicle exhaust and the burning of natural gas as part of the steam-assisted gravity drainage method used to extract oil sands in the area are contributing pyrogenic PAHs to the deposition to lakes in this region. In addition, the deposition of these pyrogenic PAHs is likely to increase as development of these oil sands deposits found at depth expands.

4.1.2. Chapter 3

This study also marks the first attempt to explore the relationship between climate and PAHs and the differences between regions that experience different climatic conditions. Contrary to models and laboratory experiments (Mackay and Paterson 1991,

Zheng et al. 2013), PAHs were generally not found to correlate with organic carbon, algal-derived carbon or inferred chlorophyll *a*. This would suggest that the amount and composition of the PAHs deposited to lake sediments are controlled by general background sources rather than climatic conditions, and so any changes to climatic conditions would not significantly affect PAHs through changes in carbon deposition, at least for the nature and magnitude of changes observed in our study lakes. Additionally, as previously mentioned, retene was found to be a relatively small proportion of the alkylated PAHs deposited to lake sediments, thus there is currently no evidence that climate change has greatly impacted the fire history near our study lakes, either. Therefore, our conclusion is that increases in primary production from climate change, as well as regional forest fire history, have not affected the amount of PAHs or the composition and sources of PAHs deposited to sediments over the last several hundred years. Although, if fires continue to increase in frequency and intensity it may be that in the future fire events could become significant sources of carbon and PAHs to aquatic ecosystems in the study regions.

Understanding how the deposition and sources of PAHs might change in response to the combined stresses of petrochemical development and climate change will clarify how these changes might impact the integrity of freshwater sources utilized by communities residing near petrochemical developments. Although we have shown that smaller scale petrochemical development near conventional oil and gas developments, increased primary production and the disturbance from forest fires have yet to greatly impact PAH deposition and sources in lakes in the Northwest Territories, the increasing stress from climate change and land-use change near these lake ecosystems do not preclude potential increases in PAH contamination in the future.

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Appendix A. Concentrations of PAHs for all study lakes.

Table A.1: Concentrations of PAHs in lake C1A, Mackenzie Delta Uplands, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	1.00	2.00	3.00	4.00	5.00	6.00	8.00	10.00	12.00	15.00	18.00
%OC	4.71	4.67	4.31	3.76	3.87	5.09	4.95	4.81	4.51	4.19	2.77
PAH:											
NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ACY	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ACE	0.00	12.48	10.42	9.93	13.42	12.78	9.74	8.73	8.40	18.74	12.42
FLU	0.00	2.87	3.37	5.81	5.21	5.42	5.46	4.57	4.55	5.77	10.78
PHE	19.91	18.42	5.84	16.95	14.45	10.58	23.57	21.07	20.76	30.48	0.00
ANT	0.00	2.76	1.76	1.92	1.99	1.44	2.66	2.64	1.82	6.68	5.79
FLA	18.66	20.78	13.63	22.18	16.17	15.09	17.51	9.39	18.85	16.31	23.79
PYR	15.50	20.15	9.95	19.20	13.53	9.15	12.61	10.56	10.92	18.28	9.87
BaANT	6.00	4.08	2.13	3.99	3.41	2.94	3.94	2.68	3.38	4.12	3.13
CHR	12.91	12.47	10.65	14.46	8.88	7.01	7.83	8.25	7.21	8.33	11.45
BbFLA	57.36	63.78	44.62	64.32	54.51	46.41	48.45	40.36	46.31	60.70	48.66
BkFLA	11.12	11.40	8.38	12.44	9.73	9.05	9.42	7.94	9.13	11.15	9.68
BaPYR	19.28	19.76	15.52	24.87	18.04	17.90	17.73	14.49	17.53	20.07	28.57
IcdP	23.46	28.40	22.14	37.30	27.22	27.78	26.90	25.95	29.82	31.41	26.03
DahA	11.24	12.16	10.49	14.77	11.89	10.61	11.37	8.70	10.78	12.08	10.21
BghiP	68.73	89.23	68.37	98.48	77.37	72.60	75.91	61.14	72.93	83.35	71.98
DBT	3.96	10.23	8.81	8.15	8.52	8.99	6.39	7.59	4.85	9.35	9.20
C1-DBT	29.08	29.07	34.94	40.72	47.34	36.75	23.79	40.99	19.20	1.94	43.13
C2-DBT	4.44	1.27	2.26	2.88	2.44	4.24	1.43	6.07	3.84	0.67	8.46
C3-DBT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-DBT	0.00	0.00	0.00	0.00	1.07	0.00	0.00	0.00	0.00	0.00	0.00
1-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

2-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3-MePHE	19.17	23.88	22.54	20.63	15.12	9.33	13.26	16.09	10.85	36.17	14.06
2-MePHE	24.82	19.50	8.51	22.85	24.90	15.12	20.26	28.38	15.97	49.00	14.92
9/4-MePHE	29.95	22.10	9.87	26.20	30.24	19.28	23.45	34.57	18.26	59.47	19.02
1-MePHE	25.45	18.79	8.27	18.86	19.71	13.17	17.33	20.14	15.05	34.55	16.25
3,6-DiMePHE	55.87	30.36	13.41	30.66	52.45	25.00	24.90	48.44	21.63	76.52	24.24
2,6-DiMePHE	22.60	15.43	7.55	13.79	21.11	9.94	10.77	18.97	10.27	33.10	10.05
1,7-DiMePHE	25.26	19.21	9.12	16.06	21.52	11.12	12.93	18.47	11.70	28.72	13.01
1,8-DiMePHE	22.75	16.64	8.06	15.98	22.80	12.10	12.33	20.37	10.18	33.57	11.73
Retene	16.40	17.50	11.45	14.29	9.67	22.57	13.00	5.97	13.73	11.14	16.55
C2-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-NAPH	21.44	14.72	5.26	13.86	9.07	4.42	11.23	5.81	13.05	0.33	8.03
C4-NAPH	57.00	28.92	16.77	43.63	31.67	18.50	25.15	20.38	26.97	1.40	28.80
C1-FLU	15.88	21.68	25.11	39.69	39.90	42.19	37.84	43.52	27.43	1.77	41.31
C2-FLU	131.67	186.83	171.32	248.64	307.36	266.94	214.41	351.13	133.47	15.31	202.82
C3-FLU	143.41	257.20	213.39	301.67	425.04	334.37	193.77	483.27	122.45	20.91	272.16
C1-PHE/ANT	112.84	90.28	56.51	101.87	110.11	66.10	87.54	126.60	71.44	7.43	74.91
C2-PHE/ANT	0.00	0.00	0.00	0.00	63.26	0.00	0.00	89.56	0.00	3.14	0.00
C3-PHE/ANT	88.57	151.60	77.89	119.75	139.49	88.20	93.87	109.58	80.89	5.88	92.95
C4-PHE/ANT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-FLA/PYR	63.31	65.90	48.19	66.07	49.78	47.32	51.86	29.13	51.73	1.73	77.11
C2-FLA/PYR	118.78	185.19	147.21	156.30	123.34	120.49	119.69	73.92	112.08	4.85	168.95
C3-FLA/PYR	55.18	124.94	100.31	99.85	97.27	89.63	88.81	46.07	66.36	2.97	96.66
C4-FLA/PYR	33.98	78.28	70.71	69.58	47.59	59.86	50.69	30.13	46.82	2.15	63.55
C1-BaA/CHR	20.36	16.98	8.03	12.58	15.03	12.68	16.04	10.76	12.43	0.64	10.70
C2-BaA/CHR	22.99	17.82	9.50	13.07	14.15	13.65	15.89	11.76	12.88	0.66	11.58
C3-BaA/CHR	9.53	9.87	5.10	7.64	8.34	7.34	9.78	9.18	7.21	0.35	9.53
C4-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-BFLA/BPYR	35.15	38.35	30.91	50.50	38.16	37.55	36.18	32.07	39.52	1.50	42.02
C2-BFLA/BPYR	5.50	8.35	9.74	12.08	9.28	8.59	8.40	9.22	8.89	0.38	8.19

Table A.2: Concentrations of PAHs in lake C23, Mackenzie Delta Uplands, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	1.75	2.75	3.75	4.75	5.75	6.75	8.50	11.75	15.50
%OC	13.17	13.10	12.91	12.12	11.40	12.61	12.11	12.44	12.14
PAH:									
NAPH	21.09	53.46	46.43	65.06	64.08	53.71	0.00	0.00	40.15
ACY	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ACE	0.00	0.98	0.00	0.00	4.16	3.22	2.21	0.00	4.24
FLU	3.23	4.92	3.03	6.94	5.03	4.43	4.71	5.15	5.14
PHE	11.10	21.14	15.65	31.57	28.09	24.26	26.80	23.12	30.05
ANT	0.00	0.00	0.00	5.77	0.00	0.00	4.00	0.00	5.89
FLA	3.03	4.77	3.99	6.78	5.17	5.40	5.77	6.05	5.97
PYR	3.09	5.24	3.72	6.40	5.24	5.77	5.61	6.25	5.46
BaANT	0.66	0.98	0.93	1.36	1.26	0.87	1.17	1.13	1.45
CHR	1.62	2.24	1.87	2.78	2.55	2.52	3.12	2.75	2.54
BbFLA	7.11	13.51	10.48	13.58	15.59	14.04	15.45	15.37	15.31
BkFLA	1.14	2.17	1.99	3.01	3.57	2.42	2.60	3.09	3.80
BaPYR	1.97	4.13	3.79	4.53	5.06	3.97	5.43	4.80	4.12
IcdP	4.12	9.22	6.16	7.83	8.40	6.25	10.66	10.36	7.73
DahA	1.48	3.61	2.72	3.11	4.54	3.19	4.80	2.40	4.27
BghiP	11.46	23.29	20.23	24.73	23.87	24.65	27.08	28.62	24.72
DBT	0.00	0.00	0.00	2.96	0.00	0.00	0.00	0.00	0.42
C1-DBT	6.01	0.00	0.00	0.00	5.87	3.56	5.97	4.63	3.76
C2-DBT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-DBT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-DBT	5.96	0.00	0.00	50.13	0.00	0.00	0.00	0.00	0.00
1-MeNAPH	10.96	23.72	22.39	27.08	29.59	22.45	0.00	0.00	22.45
2-MeNAPH	22.49	42.41	44.26	49.98	56.28	45.07	0.00	0.00	40.98
3-MePHE	4.99	8.04	5.71	12.82	10.43	9.66	10.66	10.97	9.28
2-MePHE	6.27	9.99	6.77	16.46	11.74	10.86	13.01	14.77	10.62
9/4-MePHE	5.41	9.83	6.88	14.39	10.23	11.41	12.45	12.77	11.26

1-MePHE	6.97	9.93	6.70	15.08	10.67	10.64	12.19	12.85	11.12
3,6-DiMePHE	7.58	12.25	7.57	19.60	12.98	13.91	16.02	15.86	14.41
2,6-DiMePHE	2.90	5.33	3.83	9.47	4.63	7.07	8.37	7.66	7.98
1,7-DiMePHE	3.91	5.68	4.20	10.43	7.27	8.78	9.64	8.41	8.05
1,8-DiMePHE	3.74	3.82	2.77	8.97	5.12	5.40	6.13	6.73	6.12
Retene	5.15	8.84	7.31	9.13	9.70	10.08	11.26	10.74	11.11
C2-NAPH	43.73	163.55	175.42	103.04	184.71	156.72	0.00	0.00	123.79
C3-NAPH	27.53	207.37	270.94	118.22	289.27	248.89	0.00	0.00	125.50
C4-NAPH	26.29	200.09	227.93	131.63	279.89	232.16	0.00	0.00	116.37
C1-FLU	14.31	18.98	13.66	52.97	27.68	25.31	31.74	38.27	36.84
C2-FLU	31.42	23.04	16.11	126.40	33.89	30.65	28.00	32.46	32.12
C3-FLU	52.41	29.53	20.22	186.96	36.07	33.95	30.10	32.60	36.91
C1-PHE/ANT	42.59	58.11	39.03	81.97	62.86	65.06	62.83	66.00	59.41
C2-PHE/ANT	25.98	12.00	4.06	152.26	41.54	49.86	59.38	59.08	51.70
C3-PHE/ANT	34.75	38.12	29.55	66.95	49.76	51.37	60.55	49.36	55.11
C4-PHE/ANT	0.00	0.00	0.00	190.00	0.00	0.00	15.41	0.00	0.00
C1-FLA/PYR	8.62	11.41	9.37	17.19	14.70	16.00	17.28	16.40	16.14
C2-FLA/PYR	15.02	21.16	11.47	37.51	18.27	20.37	19.76	20.80	22.72
C3-FLA/PYR	6.35	0.00	2.70	19.23	7.30	20.37	7.50	8.17	7.75
C4-FLA/PYR	5.43	0.00	5.09	18.99	8.03	20.37	7.66	5.95	7.75
C1-BaA/CHR	3.41	6.84	5.29	8.39	7.47	6.53	7.40	7.08	7.29
C2-BaA/CHR	3.64	7.13	5.98	10.58	8.42	7.35	8.24	7.42	8.52
C3-BaA/CHR	0.00	0.00	1.73	3.97	3.55	2.39	2.85	2.46	3.05
C4-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-BFLA/BPYR	6.55	13.98	12.65	18.61	18.92	20.41	34.35	32.71	36.62
C2-BFLA/BPYR	0.12	0.56	0.30	0.52	0.42	0.54	0.54	0.00	0.66

Table A.3: Concentrations of PAHs in lake I17, Mackenzie Delta Uplands, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	2.25	3.25	4.25	5.5	6.5	7.5	8.75	10	11	12.5
%OC	21.97	21.95	22.05	21.48	22.33	21.99	22.30	22.83	24.19	24.21
PAH:										
NAPH	0.00	10.29	0.00	0.00	0.00	11.85	36.04	0.00	0.00	0.00
ACY	0.94	0.45	1.46	1.09	1.34	1.65	1.29	0.41	1.47	1.36
ACE	1.70	1.10	1.86	1.47	1.76	1.64	1.16	1.77	1.71	1.85
FLU	4.42	8.92	5.25	4.50	5.43	5.50	6.50	6.72	7.51	8.00
PHE	13.27	16.01	18.11	18.09	15.12	18.57	16.48	23.49	17.28	19.32
ANT	1.06	2.13	1.89	1.65	1.54	2.45	2.65	3.00	2.29	1.92
FLA	4.87	0.00	5.64	6.58	7.88	7.91	5.35	9.95	8.80	9.33
PYR	4.77	5.03	5.41	6.57	6.77	8.33	7.40	9.00	7.53	7.69
BaANT	0.82	1.35	0.81	1.04	1.41	1.40	1.61	2.37	1.57	1.46
CHR	4.96	7.84	5.95	6.76	7.41	7.68	7.36	8.89	8.06	8.59
BbFLA	9.94	10.88	9.58	12.00	14.07	10.73	16.10	13.91	13.79	11.27
BkFLA	1.60	11.69	1.65	1.84	2.26	2.17	2.40	2.31	2.69	2.84
BaPYR	2.20	4.59	2.92	2.52	2.75	2.69	3.26	3.18	3.18	3.20
IcdP	2.39	3.08	3.97	4.72	0.00	3.09	3.68	5.25	3.52	4.26
DahA	1.68	0.00	1.51	3.18	3.27	3.02	5.30	3.64	3.99	4.35
BghiP	6.64	12.45	8.43	8.90	12.36	10.96	0.00	19.13	19.17	16.86
DBT	3.99	5.87	5.51	5.62	5.04	5.53	6.70	9.44	7.91	8.77
C1-DBT	9.65	16.71	13.38	13.20	12.92	12.39	12.45	18.68	15.18	14.02
C2-DBT	8.34	16.10	10.39	11.65	8.34	5.52	9.61	14.98	7.85	9.47
C3-DBT	9.53	26.68	16.36	16.89	12.56	12.59	13.11	19.49	17.94	17.50
C4-DBT	7.13	34.78	17.26	16.99	6.33	10.93	10.73	17.84	13.53	21.03
1-MeNAPH	1.03	6.69	2.31	0.93	2.45	7.88	15.95	1.83	0.69	0.41
2-MeNAPH	0.00	3.85	0.00	0.00	0.00	6.89	18.48	0.00	0.00	0.00
3-MePHE	3.74	4.75	3.71	4.34	4.27	5.94	4.07	5.15	4.57	4.90

2-MePHE	4.20	4.70	4.53	4.19	4.72	5.79	4.60	5.49	4.66	5.41
9/4-MePHE	5.04	7.17	6.22	6.04	6.07	7.08	6.63	7.73	6.58	7.12
1-MePHE	4.16	4.82	4.57	4.76	5.08	5.84	5.80	6.53	6.19	5.71
3,6-DiMePHE	5.05	7.08	5.09	5.54	6.22	6.46	5.39	5.87	5.26	5.71
2,6-DiMePHE	1.87	2.35	2.07	2.20	2.25	2.89	2.68	2.66	2.66	2.76
1,7-DiMePHE	2.01	1.78	1.94	2.30	2.30	2.79	2.42	2.30	2.24	2.40
1,8-DiMePHE	1.73	3.13	2.00	2.05	1.85	2.41	2.05	2.73	1.99	2.18
Retene	7.15	3.09	7.73	9.77	13.10	14.45	15.15	31.93	17.95	17.80
C2-NAPH	9.24	30.53	14.10	7.16	10.62	36.33	70.59	13.14	8.55	3.83
C3-NAPH	3.34	18.94	10.44	0.83	4.63	27.04	47.95	5.83	2.45	0.97
C4-NAPH	3.86	25.72	10.68	3.08	4.83	23.35	44.54	6.98	2.47	5.15
C1-FLU	16.28	22.98	21.74	17.48	19.90	19.28	26.41	29.70	26.43	25.51
C2-FLU	41.95	61.97	61.73	62.11	59.92	51.83	57.84	63.81	56.38	64.15
C3-FLU	115.95	149.08	173.86	84.27	79.24	82.73	57.26	77.86	62.05	106.51
C1-PHE/ANT	34.57	49.94	42.44	28.69	29.66	36.44	26.52	35.39	29.08	35.59
C2-PHE/ANT	21.75	34.73	28.38	25.17	27.51	29.78	26.72	30.90	28.76	30.41
C3-PHE/ANT	14.06	23.84	16.71	17.07	19.46	19.02	17.49	22.56	19.68	18.58
C4-PHE/ANT	26.15	43.30	31.42	24.28	30.56	27.66	24.54	28.45	30.24	43.63
C1-FLA/PYR	7.03	2.86	11.69	11.15	0.00	14.28	10.73	17.62	19.15	23.87
C2-FLA/PYR	11.33	3.25	11.78	16.38	0.00	20.71	13.26	20.15	22.05	25.97
C3-FLA/PYR	9.80	2.34	9.95	9.99	0.00	15.52	10.58	13.85	13.40	21.47
C4-FLA/PYR	3.61	4.21	10.72	15.07	0.00	15.13	11.59	19.81	19.17	18.58
C1-BaA/CHR	22.30	26.55	24.14	14.99	36.63	13.89	15.78	14.97	17.72	19.91
C2-BaA/CHR	21.32	37.59	20.27	22.93	28.36	26.46	41.69	55.00	42.05	36.84
C3-BaA/CHR	15.25	25.17	12.58	15.91	23.99	16.52	25.08	32.15	24.40	26.66
C4-BaA/CHR	18.68	25.80	11.46	13.63	16.88	11.81	23.59	12.65	25.83	16.47
C1-BFLA/BPYR	22.98	25.81	17.54	18.75	28.28	23.36	26.76	23.72	32.55	30.35
C2-BFLA/BPYR	7.08	9.54	6.64	7.26	11.11	8.29	12.41	7.88	8.01	11.56

Table A.4: Concentrations of PAHs in lake I20, Mackenzie Delta Uplands, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.75	1.75	2.75	3.75	4.75	5.75	6.75	7.75	9.75	11.75	14.50	18.00
%OC	25.65	23.80	26.67	26.38	23.71	27.81	24.74	25.44	26.71	25.82	28.69	24.80
PAH:												
NAPH	0.00	0.00	0.00	0.00	0.00	0.00	3.74	0.00	0.00	0.00	0.00	3.57
ACY	3.40	0.00	0.43	0.88	0.61	1.10	0.02	1.31	0.78	0.29	0.43	0.00
ACE	2.55	0.54	0.25	2.66	3.63	1.58	2.98	2.75	4.51	1.52	2.10	0.25
FLU	2.10	1.74	2.26	3.72	4.25	2.97	3.39	3.28	5.57	2.87	6.80	4.77
PHE	8.16	6.76	10.50	16.42	20.19	11.67	12.17	9.07	23.94	12.76	27.84	15.78
ANT	3.80	0.00	1.53	2.50	4.07	2.38	3.61	4.91	3.92	2.96	4.82	2.31
FLA	7.71	1.79	2.69	3.69	4.92	3.36	3.28	5.62	4.46	3.65	6.77	4.45
PYR	16.20	1.65	3.58	5.59	6.18	4.01	4.61	5.62	6.96	3.16	6.21	4.50
BaANT	2.05	0.46	0.42	0.53	0.51	0.46	0.64	1.65	0.38	0.55	1.00	0.62
CHR	7.60	0.74	0.94	1.21	1.39	1.27	1.47	2.46	1.31	1.09	2.25	1.12
BbFLA	7.66	3.41	5.01	5.66	6.15	6.57	7.19	9.38	5.40	6.50	11.11	8.13
BkFLA	1.55	0.49	0.60	0.84	0.92	0.90	1.17	2.11	1.10	1.04	1.41	1.29
BaPYR	3.79	0.95	1.10	1.93	2.04	1.80	1.94	2.86	1.77	1.85	2.53	1.79
IcdP	4.27	1.36	1.96	3.16	3.72	3.20	3.89	3.50	1.85	3.30	4.45	3.87
DahA	1.14	0.49	0.95	1.02	1.29	1.11	1.42	1.09	1.01	1.34	2.26	1.55
BghiP	10.31	4.90	6.36	8.39	9.77	10.70	9.10	9.40	8.08	9.56	12.49	8.74
DBT	0.00	0.00	0.00	0.00	0.00	0.00	0.75	0.00	0.00	0.00	1.56	0.96
C1-DBT	0.00	68.39	4.39	5.17	6.13	5.18	7.21	7.92	6.72	5.94	10.95	7.87
C2-DBT	0.00	0.00	0.00	0.00	3.14	2.20	4.82	3.75	3.84	0.00	0.00	0.00
C3-DBT	0.00	0.00	0.00	0.00	1.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-DBT	0.00	2.53	0.00	6.21	17.94	18.07	0.00	6.30	35.72	0.00	34.31	1.16
1-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	2.78	0.00	0.00	0.00	0.00	2.82
2-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	1.32	0.00	0.00	0.00	0.00	1.73
3-MePHE	4.89	2.55	4.75	7.80	8.97	5.19	47.87	3.70	8.92	6.01	10.71	19.47
2-MePHE	5.80	2.85	4.63	8.29	8.89	5.46	4.85	3.56	9.80	5.81	11.11	5.30
9/4-MePHE	4.68	2.67	4.17	7.11	8.76	5.28	5.40	3.89	7.89	5.87	10.18	5.62

1-MePHE	5.37	3.26	4.01	7.92	8.87	5.93	4.84	5.01	7.80	6.01	11.73	5.82
3,6-DiMePHE	7.79	3.84	5.98	12.73	12.20	6.46	6.23	4.84	12.57	7.92	10.73	6.23
2,6-DiMePHE	3.11	1.45	2.45	4.84	5.71	2.85	2.88	2.07	4.18	2.93	4.67	3.86
1,7-DiMePHE	3.15	1.68	2.46	4.73	4.54	3.45	3.88	3.02	4.94	4.29	6.41	3.49
1,8-DiMePHE	2.95	1.25	2.50	4.45	4.18	2.79	2.43	1.76	4.64	2.54	5.54	2.81
Retene	14.43	9.12	12.25	16.56	20.29	25.69	23.30	19.85	26.67	20.63	42.87	26.57
C2-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	15.54	0.00	0.00	0.00	0.00	11.94
C3-NAPH	0.00	0.00	0.00	0.00	11.77	0.00	16.05	0.00	0.00	0.00	0.00	15.77
C4-NAPH	0.00	0.00	0.00	0.00	5.17	0.00	16.56	0.00	0.00	0.00	0.00	16.23
C1-FLU	16.71	11.75	10.69	25.73	20.41	15.52	19.84	19.28	20.61	17.77	39.84	25.06
C2-FLU	26.14	19.95	27.25	45.79	43.63	30.45	38.08	37.88	45.63	34.42	92.09	39.73
C3-FLU	63.13	34.91	37.51	76.58	66.30	42.69	58.54	43.26	75.23	51.61	97.64	44.06
C1-PHE/ANT	37.97	18.74	27.42	46.61	53.04	35.73	86.92	26.10	50.78	37.02	66.50	57.73
C2-PHE/ANT	27.30	8.25	11.98	45.88	45.29	22.01	0.00	18.82	62.47	18.25	51.19	4.41
C3-PHE/ANT	39.36	19.28	19.82	45.88	39.52	34.54	34.23	29.72	40.66	32.10	59.49	37.91
C4-PHE/ANT	10.36	5.74	0.00	55.79	29.46	8.10	0.00	27.27	79.31	20.18	57.85	0.00
C1-FLA/PYR	14.43	6.16	8.13	11.23	10.89	9.41	12.00	9.49	10.72	10.03	15.26	11.39
C2-FLA/PYR	16.39	7.41	9.41	14.12	15.89	14.68	17.24	16.14	11.77	14.47	20.62	21.22
C3-FLA/PYR	0.00	4.10	5.05	11.03	6.78	6.42	11.87	8.32	0.00	8.44	12.25	15.56
C4-FLA/PYR	0.00	4.19	5.51	8.76	8.26	6.18	10.97	8.96	0.00	9.09	13.29	14.06
C1-BaA/CHR	6.27	2.90	3.98	4.88	5.93	4.39	6.01	5.12	3.90	3.87	6.21	4.31
C2-BaA/CHR	16.58	11.17	15.92	22.42	27.73	25.88	33.37	23.58	16.35	19.36	30.90	29.80
C3-BaA/CHR	2.27	1.88	3.15	4.65	6.50	5.33	8.38	4.96	3.84	4.41	7.26	7.54
C4-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	5.90	0.00	0.00	0.00	0.00	4.45
C1-BFLA/BPYR	6.44	4.34	6.04	7.07	7.78	8.47	9.58	7.47	7.06	8.21	12.19	9.99
C2-BFLA/BPYR	0.33	0.36	0.23	0.37	0.36	0.36	1.61	0.29	0.28	0.55	0.60	1.29

Table A.5: Concentrations of PAHs in lake CamH1, Cameron Hills, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.75	1.75	2.75	3.75	4.75	5.75	6.75	7.75	8.75	9.75
%OC	26.34	25.75	25.86	26.90	27.00	26.56	25.31	24.97	24.62	24.53
PAH:										
NAPH	14.55	1.68	5.01	26.53	0.00	0.00	0.00	0.00	0.00	0.00
ACY	1.45	1.51	1.87	1.68	1.79	2.21	3.17	2.86	2.79	2.43
ACE	6.19	6.43	9.22	11.94	10.52	11.46	13.71	9.60	7.35	8.68
FLU	56.18	46.82	54.54	49.94	44.07	35.74	23.38	17.00	12.28	14.02
PHE	119.16	88.91	108.81	110.70	112.57	97.30	70.91	60.24	49.42	56.88
ANT	15.04	11.02	17.70	17.57	18.72	17.78	21.92	22.95	14.04	16.75
FLA	62.57	35.95	53.89	62.41	57.70	54.91	69.51	79.32	54.52	61.70
PYR	29.52	18.69	22.15	26.10	23.42	20.29	28.46	28.25	25.83	31.69
BaANT	5.14	3.11	19.89	3.63	4.69	14.63	5.11	6.87	5.00	4.95
CHR	23.31	14.95	19.16	18.20	16.29	16.58	15.92	16.13	12.42	14.43
BbFLA	40.27	29.13	36.82	37.35	33.94	29.10	28.27	38.15	26.74	31.25
BkFLA	4.22	1.81	2.34	2.43	2.11	2.33	1.75	4.09	3.60	3.24
BaPYR	3.94	2.66	3.59	4.15	3.48	3.45	4.26	4.57	3.66	3.31
IcdP	0.00	5.16	0.00	27.16	6.79	0.00	5.83	6.09	2.81	5.49
DahA	0.00	0.00	2.49	1.23	1.20	1.43	0.00	1.91	2.98	2.30
BghiP	5.91	4.53	5.61	6.27	7.83	5.73	4.49	6.14	4.25	5.04
DBT	11.94	8.41	9.46	9.66	9.45	7.97	7.77	7.92	6.27	6.28
C1-DBT	11.43	9.60	9.13	8.03	9.16	7.49	7.51	7.89	6.84	5.42
C2-DBT	18.50	6.27	7.84	7.44	17.16	5.98	3.77	4.51	3.81	4.29
C3-DBT	66.51	19.18	45.69	44.29	43.35	26.66	12.07	12.25	6.48	6.29
C4-DBT	29.90	10.55	17.49	18.32	22.55	10.81	7.51	9.19	6.02	3.47
1-MeNAPH	0.74	0.46	0.10	1.43	0.00	0.00	0.00	0.00	0.00	0.00
2-MeNAPH	0.00	0.00	0.00	0.28	0.00	0.00	0.00	0.00	0.00	0.00
3-MePHE	8.99	5.86	7.45	7.50	8.27	7.40	8.10	8.65	7.60	8.48

2-MePHE	18.90	13.48	16.15	16.95	7.68	15.19	15.79	15.13	13.07	14.99
9/4-MePHE	9.71	6.78	7.49	7.80	8.02	7.59	8.90	11.12	8.56	9.45
1-MePHE	33.92	24.15	29.10	30.26	30.52	27.11	20.81	18.92	15.68	17.86
3,6-DiMePHE	4.20	2.84	3.18	3.63	3.58	3.99	2.97	2.87	2.65	2.94
2,6-DiMePHE	5.61	3.71	4.29	4.50	4.86	4.34	4.35	5.05	3.70	4.75
1,7-DiMePHE	24.66	16.42	20.51	21.99	21.46	18.93	15.67	13.84	10.98	11.83
1,8-DiMePHE	2.46	1.24	1.98	1.79	1.75	1.86	1.88	1.83	1.56	1.84
Retene	166.80	104.31	190.41	171.21	177.96	159.60	126.48	117.32	71.92	102.76
C2-NAPH	16.17	16.85	15.39	22.26	11.81	10.64	8.99	7.27	6.48	11.92
C3-NAPH	11.21	11.49	11.27	15.33	7.75	8.28	8.20	6.86	6.31	11.10
C4-NAPH	8.17	9.17	9.18	13.39	6.19	7.56	6.96	5.32	5.32	9.70
C1-FLU	50.30	37.21	41.23	40.80	39.43	38.78	31.85	33.01	27.49	28.53
C2-FLU	67.89	54.32	54.35	50.43	51.54	46.49	36.71	38.38	30.97	31.35
C3-FLU	80.17	52.44	45.14	43.26	45.59	37.49	26.25	27.22	20.34	18.76
C1-PHE/ANT	80.35	56.57	69.36	71.98	73.07	67.42	62.19	64.75	55.10	65.13
C2-PHE/ANT	68.07	46.92	56.86	65.36	58.94	62.19	57.52	54.47	47.30	53.97
C3-PHE/ANT	68.92	51.78	71.81	65.06	69.53	33.33	55.32	47.28	39.09	47.96
C4-PHE/ANT	53.99	39.22	48.94	48.80	60.81	61.10	63.65	52.50	49.83	54.88
C1-FLA/PYR	56.28	35.17	51.22	58.76	58.14	52.30	78.64	95.02	69.79	73.67
C2-FLA/PYR	40.21	26.86	37.07	40.61	44.16	35.10	47.19	56.58	39.28	42.47
C3-FLA/PYR	17.33	9.81	16.52	16.02	16.57	14.66	21.88	22.03	17.45	14.26
C4-FLA/PYR	15.56	8.60	12.77	16.23	16.44	15.47	18.78	24.76	14.09	17.63
C1-BaA/CHR	21.69	11.06	72.93	16.51	23.28	72.22	25.52	21.65	18.62	19.32
C2-BaA/CHR	34.08	19.39	114.46	20.21	28.43	101.69	34.69	33.17	28.45	32.43
C3-BaA/CHR	32.76	15.89	110.64	17.17	28.09	97.32	24.98	25.97	21.14	24.67
C4-BaA/CHR	12.40	0.00	75.44	0.00	9.80	88.53	2.13	3.38	2.55	6.09
C1-BFLA/BPYR	0.00	0.00	0.00	0.01	0.00	0.96	0.00	8.45	3.77	6.59
C2-BFLA/BPYR	2.09	2.16	1.41	2.25	2.61	1.99	1.54	4.48	3.88	3.59

Table A.6: Concentrations of PAHs in lake CamH2, Cameron Hills, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	1.25	3.00	4.50	6.50	8.00	9.50	14.50	17.50	21.00	25.00	29.00
%OC	13.71	13.64	12.75	14.58	12.03	13.81	12.61	15.89	15.58	15.88	16.70
PAH:											
NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ACY	0.08	0.24	1.01	0.20	0.00	0.30	0.37	0.25	0.00	0.22	0.05
ACE	2.62	2.11	3.43	5.45	2.42	3.77	2.80	1.97	2.09	1.83	2.42
FLU	10.24	7.49	14.04	3.46	4.31	6.09	5.09	4.86	4.95	3.73	4.94
PHE	24.55	20.77	42.09	11.81	13.78	28.21	21.08	15.70	14.17	11.06	12.96
ANT	0.95	2.26	1.20	0.38	0.66	1.20	1.46	1.11	0.56	0.54	0.65
FLA	4.82	7.54	17.79	3.02	2.65	5.02	6.11	6.39	5.82	7.71	7.04
PYR	7.15	8.96	18.70	3.84	4.66	8.95	10.53	9.08	7.43	7.31	8.02
BaANT	0.78	0.99	2.39	0.56	0.74	0.93	1.48	1.38	1.23	1.46	1.45
CHR	1.29	1.50	9.10	2.64	3.47	5.82	9.31	1.12	1.45	1.42	1.33
BbFLA	8.68	10.37	19.47	4.74	7.69	10.47	17.11	13.09	11.20	14.86	13.81
BkFLA	1.41	1.41	2.33	0.68	0.91	1.69	2.68	2.10	1.69	2.18	2.43
BaPYR	1.63	1.20	4.70	1.46	1.43	2.76	4.82	3.88	3.23	4.77	3.85
IcdP	4.27	4.56	7.56	2.19	3.63	5.24	9.79	6.54	5.24	7.23	7.08
DahA	0.58	1.02	1.69	0.30	0.78	1.17	2.04	1.62	1.40	1.92	1.61
BghiP	7.78	8.12	18.54	3.80	11.07	10.81	15.66	11.98	12.57	13.80	15.50
DBT	9.94	7.01	14.22	6.30	4.73	7.80	7.14	6.39	5.45	7.34	6.89
C1-DBT	18.90	15.64	51.96	17.54	0.00	14.93	10.15	14.14	18.63	19.06	13.93
C2-DBT	65.74	21.74	91.46	24.60	0.00	19.57	41.52	9.61	23.86	17.61	21.07
C3-DBT	15.74	20.75	98.00	17.25	0.00	23.18	16.33	18.60	22.56	20.54	19.47
C4-DBT	44.77	35.66	173.59	47.45	0.00	37.70	28.32	35.92	44.40	54.47	39.28
1-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2-MeNAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3-MePHE	15.85	11.37	39.03	13.29	20.60	16.93	3.16	3.64	5.58	3.53	4.51
2-MePHE	5.96	5.17	10.97	1.96	2.60	7.12	3.45	3.21	3.67	2.33	2.45
9/4-MePHE	6.28	3.70	11.07	1.48	2.37	8.14	3.63	3.37	4.14	2.42	3.01

1-MePHE	6.22	5.02	11.21	2.63	2.77	7.32	6.03	4.17	3.30	2.47	3.18
3,6-DiMePHE	3.95	3.31	5.95	0.91	2.33	4.44	2.42	1.61	2.56	1.86	1.49
2,6-DiMePHE	1.46	1.47	3.34	0.52	4.46	2.43	1.61	1.20	1.70	1.09	0.96
1,7-DiMePHE	3.00	2.98	5.85	1.48	2.25	2.97	2.31	1.30	1.56	1.15	1.43
1,8-DiMePHE	1.86	1.22	2.94	0.82	0.98	1.54	0.86	0.74	1.17	0.78	0.78
Retene	9.88	12.85	15.99	4.37	5.19	8.24	8.73	5.72	5.00	4.10	5.06
C2-NAPH	8.58	3.09	4.00	3.56	0.00	4.53	2.09	3.26	5.37	3.51	5.77
C3-NAPH	8.95	2.92	2.61	6.33	0.00	0.30	0.00	0.00	0.81	0.00	2.02
C4-NAPH	4.88	0.00	2.00	0.49	0.00	3.16	1.52	1.20	2.28	0.27	2.36
C1-FLU	24.42	34.28	0.00	0.00	0.00	13.89	11.93	13.34	14.25	10.95	12.77
C2-FLU	86.37	53.24	0.00	83.65	0.00	29.02	25.15	24.80	22.54	26.38	25.85
C3-FLU	65.98	16.85	0.00	48.62	0.00	31.31	28.23	25.16	29.63	40.52	40.79
C1-PHE/ANT	43.58	30.01	121.28	40.24	0.00	50.35	20.14	19.07	23.71	14.39	18.80
C2-PHE/ANT	24.04	20.59	45.46	0.00	0.00	25.90	20.27	15.55	18.10	29.29	22.49
C3-PHE/ANT	28.89	19.13	31.10	6.84	0.00	18.16	15.92	11.93	17.56	15.58	13.31
C4-PHE/ANT	135.87	54.96	327.16	70.41	0.00	54.95	48.89	35.25	83.34	59.52	42.56
C1-FLA/PYR	0.00	7.63	29.85	4.18	0.00	16.19	23.94	20.14	22.31	19.70	19.84
C2-FLA/PYR	0.00	11.01	31.55	0.00	0.00	11.87	18.72	15.50	0.00	23.82	16.38
C3-FLA/PYR	0.00	0.00	0.00	0.00	0.00	0.00	10.31	8.30	0.00	0.00	0.00
C4-FLA/PYR	0.00	8.24	18.87	3.60	0.00	0.67	9.63	13.85	0.00	5.21	2.60
C1-BaA/CHR	2.68	3.81	12.56	1.55	0.00	4.03	6.45	6.51	5.90	6.20	6.40
C2-BaA/CHR	7.04	11.02	18.41	5.13	0.00	6.33	8.44	10.69	7.87	8.63	7.78
C3-BaA/CHR	19.76	20.51	51.37	10.03	0.00	4.45	5.79	5.36	3.23	3.43	4.43
C4-BaA/CHR	0.48	6.56	9.53	0.00	0.00	1.06	1.76	1.86	5.03	2.67	1.87
C1-BFLA/BPYR	9.80	15.43	36.70	9.30	0.00	24.78	40.72	26.50	21.51	24.90	31.45
C2-BFLA/BPYR	0.00	2.45	2.92	0.84	0.00	1.47	4.19	1.90	2.01	2.29	2.23

Table A.7: Concentrations of PAHs in lake CamH3, Cameron Hills, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	1.25	3.00	4.50	6.00	8.00	10.50	12.50	16.00	21.00	25.00
%OC	16.69	18.11	18.19	19.06	19.09	19.22	19.34	22.72	22.06	21.77
PAH:										
NAPH	0.00	0.00	0.00	0.00	2.94	0.00	35.39	0.00	0.00	0.00
ACY	0.52	0.66	0.51	0.56	0.76	0.64	0.56	0.87	0.00	0.00
ACE	0.00	2.40	7.72	4.06	4.61	6.76	4.60	0.00	3.72	0.00
FLU	7.45	6.25	12.08	7.98	9.30	14.23	7.55	15.06	10.01	0.00
PHE	28.01	26.45	59.69	36.14	45.85	57.33	29.11	47.44	25.40	0.00
ANT	1.23	2.40	5.96	4.73	5.80	5.00	4.24	3.84	4.46	0.65
FLA	10.69	12.86	15.45	11.41	19.00	14.81	11.61	22.15	12.75	0.00
PYR	6.65	7.17	14.92	10.27	13.51	17.58	12.43	22.50	17.59	0.00
BaANT	0.95	1.05	1.31	1.32	2.11	1.43	11.34	2.16	1.95	0.82
CHR	3.52	3.99	10.33	3.65	6.54	3.69	3.41	6.10	4.81	2.21
BbFLA	9.34	10.06	11.14	10.45	15.24	9.44	11.24	20.33	11.85	3.88
BkFLA	1.48	1.58	1.20	1.36	2.24	1.44	1.44	2.89	1.64	4.44
BaPYR	1.36	1.34	3.47	2.12	2.57	1.54	1.76	2.85	2.33	1.09
IcdP	3.62	3.89	2.93	4.37	4.36	3.12	3.28	7.67	4.79	3.42
DahA	0.66	0.77	0.58	0.65	0.99	1.03	0.83	1.26	0.93	0.00
BghiP	3.61	4.23	5.03	4.21	6.59	4.72	5.80	8.86	7.36	2.15
DBT	6.50	6.98	9.45	5.70	6.08	10.04	6.34	15.63	13.47	0.00
C1-DBT	16.55	19.39	12.56	4.21	5.45	15.93	6.04	60.87	20.36	0.00
C2-DBT	29.72	14.61	15.67	6.22	4.87	16.53	4.45	62.05	26.46	0.00
C3-DBT	46.08	34.61	7.32	7.67	6.63	4.82	5.20	103.10	24.62	0.00
C4-DBT	58.74	34.99	9.85	15.64	9.06	8.33	3.84	133.13	107.05	0.00
1-MeNAPH	0.00	0.00	0.00	0.00	1.19	0.00	7.83	0.00	0.00	0.00
2-MeNAPH	0.00	0.00	0.26	0.01	1.05	0.55	4.45	0.00	0.00	0.00
3-MePHE	10.78	14.01	9.47	4.57	4.49	11.39	4.02	12.05	5.22	0.00
2-MePHE	3.98	3.01	15.21	7.92	9.85	16.74	7.39	11.17	7.34	0.00
9/4-MePHE	1.64	1.08	12.70	4.76	4.63	17.28	4.95	7.79	4.63	0.00

1-MePHE	4.77	4.72	14.43	8.02	10.41	14.65	6.71	9.53	5.94	0.00
3,6-DiMePHE	0.82	0.50	7.36	2.85	3.19	9.92	2.58	2.86	2.88	0.00
2,6-DiMePHE	0.80	0.86	4.04	2.31	2.66	4.60	2.14	2.01	1.54	0.00
1,7-DiMePHE	4.00	4.29	11.39	7.67	10.09	8.56	5.01	5.13	4.01	0.00
1,8-DiMePHE	0.47	0.36	2.71	0.95	1.40	3.87	0.95	1.70	1.35	0.00
Retene	3.93	2.69	8.36	9.06	12.13	7.51	8.15	6.43	3.30	3.15
C2-NAPH	2.09	0.93	1.11	0.00	2.18	3.00	26.89	0.00	0.00	22.89
C3-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	17.30	0.00	0.00	4.62
C4-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	5.29	0.00	0.00	0.00
C1-FLU	17.03	13.58	31.36	20.44	0.00	35.37	22.12	38.13	0.00	0.00
C2-FLU	55.50	55.52	30.82	22.00	0.00	42.66	23.19	86.61	0.00	0.00
C3-FLU	86.74	84.84	11.15	15.15	0.00	27.08	0.00	97.77	0.00	0.00
C1-PHE/ANT	27.58	31.97	56.05	27.97	31.69	66.41	25.21	51.43	26.64	0.00
C2-PHE/ANT	11.13	6.97	44.22	27.80	32.69	48.99	23.20	30.80	27.44	0.00
C3-PHE/ANT	16.22	3.99	25.55	19.71	25.99	21.48	16.25	29.48	14.77	0.00
C4-PHE/ANT	68.47	39.45	28.73	36.67	42.86	28.08	19.69	98.13	71.46	0.00
C1-FLA/PYR	11.32	12.57	25.72	20.76	29.75	30.56	21.89	35.07	21.80	0.00
C2-FLA/PYR	19.35	19.75	17.44	12.45	19.23	19.35	15.31	46.13	12.38	0.00
C3-FLA/PYR	0.00	0.00	8.01	0.00	10.20	8.51	6.32	0.00	0.00	0.00
C4-FLA/PYR	0.00	0.00	7.20	0.00	9.09	6.27	5.93	0.00	62.31	0.00
C1-BaA/CHR	3.49	2.88	5.01	3.86	6.28	6.60	40.51	6.76	5.90	3.62
C2-BaA/CHR	3.75	3.96	5.91	4.38	7.49	7.42	57.43	9.43	9.31	24.63
C3-BaA/CHR	6.99	5.52	4.50	2.61	4.77	5.46	44.62	13.43	10.57	6.83
C4-BaA/CHR	1.06	0.00	0.71	0.00	0.54	0.50	21.32	0.00	3.29	10.54
C1-BFLA/BPYR	2.71	2.85	5.89	5.17	7.33	4.86	6.96	13.00	9.46	10.34
C2-BFLA/BPYR	1.03	0.96	3.79	1.84	1.75	1.96	1.15	1.72	2.93	9.06

Table A.8: Concentrations of PAHs in lake CamH5, Cameron Hills, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	1.00	3.00	4.00	5.50	7.00	9.00	16.50	23.00	29.00
%OC	33.73	33.64	33.44	33.44	32.34	33.87	32.86	32.67	32.96
PAH:									
NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ACY	9.64	0.82	0.00	0.00	0.00	0.00	0.00	0.00	0.93
ACE	0.00	7.39	13.74	0.00	1.62	17.96	0.00	0.00	0.00
FLU	0.00	29.04	12.65	0.00	15.46	0.00	11.20	0.00	0.00
PHE	0.00	50.45	30.74	0.00	0.00	0.00	0.00	26.22	24.33
ANT	1.05	1.11	1.30	0.59	0.91	2.89	0.00	1.59	1.88
FLA	222.71	39.31	26.55	91.49	27.67	0.00	0.00	14.74	24.51
PYR	0.00	11.19	6.64	7.71	6.98	3.73	0.00	6.44	4.94
BaANT	4.33	2.41	2.42	2.27	0.99	6.99	0.00	2.32	1.43
CHR	6.43	4.04	1.51	3.38	1.01	1.87	0.00	1.26	1.10
BbFLA	257.59	14.03	5.43	69.74	4.05	30.24	0.00	9.13	11.47
BkFLA	21.31	1.34	0.72	5.17	0.96	2.16	0.00	0.50	0.76
BaPYR	0.00	2.59	1.34	0.00	0.88	3.01	0.00	0.00	1.64
IcdP	10.77	13.96	10.02	4.93	0.97	46.34	0.00	11.42	5.98
DahA	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
BghiP	0.00	18.50	0.00	0.00	3.14	0.00	0.00	7.45	4.83
DBT	0.00	8.59	7.27	0.00	4.50	0.00	3.79	0.00	0.00
C1-DBT	0.00	31.81	82.01	0.00	89.80	0.00	83.07	0.00	0.00
C2-DBT	0.00	43.63	175.51	0.00	127.10	0.00	93.75	0.00	0.00
C3-DBT	0.00	35.35	193.85	0.00	194.73	0.00	107.86	0.00	0.00
C4-DBT	0.00	55.74	658.63	0.00	422.22	0.00	397.67	0.00	0.00
1-MeNAPH	0.00	4.95	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2-MeNAPH	0.00	2.88	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3-MePHE	0.00	6.66	94.31	0.00	0.00	0.00	15.07	36.10	31.22
2-MePHE	0.00	8.01	5.58	0.00	0.00	0.00	2.02	4.83	4.32
9/4-MePHE	0.00	6.78	0.00	0.00	0.00	0.00	1.68	0.00	0.00

1-MePHE	0.00	10.57	6.26	0.00	0.00	0.00	1.94	5.65	0.00
3,6-DiMePHE	0.00	3.35	0.00	0.00	0.00	0.00	0.77	0.00	2.51
2,6-DiMePHE	0.00	1.66	3.90	0.00	0.00	0.00	0.73	8.05	2.98
1,7-DiMePHE	0.00	6.39	4.82	0.00	0.00	0.00	1.41	5.60	4.35
1,8-DiMePHE	0.00	2.69	0.00	0.00	0.00	0.00	0.00	3.11	0.00
Retene	84.12	25.43	10.21	39.25	9.53	0.00	1.03	8.20	5.85
C2-NAPH	43.95	50.48	26.31	37.49	19.72	49.67	15.23	51.52	56.19
C3-NAPH	17.65	34.16	14.95	28.08	7.95	19.00	13.03	20.17	17.87
C4-NAPH	22.08	13.87	0.00	40.80	2.03	0.00	3.71	9.32	10.69
C1-FLU	0.00	39.30	206.28	0.00	0.00	0.00	55.62	0.00	0.00
C2-FLU	0.00	109.81	509.05	0.00	196.03	0.00	291.47	0.00	0.00
C3-FLU	0.00	101.80	487.06	0.00	225.48	0.00	225.86	0.00	0.00
C1-PHE/ANT	0.00	49.91	673.11	0.00	0.00	0.00	57.76	310.81	386.19
C2-PHE/ANT	0.00	42.41	0.00	0.00	0.00	0.00	21.83	151.12	102.56
C3-PHE/ANT	0.00	36.41	176.83	0.00	0.00	0.00	26.00	193.43	165.67
C4-PHE/ANT	0.00	66.78	444.95	0.00	0.00	0.00	96.28	290.18	422.70
C1-FLA/PYR	0.00	13.65	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-FLA/PYR	0.00	22.15	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-FLA/PYR	0.00	1.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-FLA/PYR	0.00	8.12	46.37	0.00	0.00	0.00	0.00	9.71	0.00
C1-BaA/CHR	0.00	7.15	15.13	0.00	3.14	35.41	1.78	8.79	8.78
C2-BaA/CHR	0.00	17.54	31.41	34.29	11.90	132.03	6.90	33.84	37.97
C3-BaA/CHR	0.00	36.42	31.63	0.00	28.87	55.99	10.88	3.97	17.43
C4-BaA/CHR	0.00	30.97	33.75	20.69	3.14	119.21	4.41	16.89	28.39
C1-BFLA/BPYR	21.95	11.77	12.63	22.66	7.05	20.23	5.47	12.28	9.20
C2-BFLA/BPYR	0.54	0.00	0.00	0.00	0.00	0.00	0.59	0.00	0.00

Table A.9: Concentrations of PAHs in lake CamH7, Cameron Hills, NWT (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.25	1.50	3.25	5.00	7.00	13.50	17.50	23.00	27.00	31.00	35.00
%OC	30.02	30.34	30.50	31.23	32.19	34.33	34.42	35.57	35.58	37.63	38.79
PAH:											
NAPH	0.00	0.00	0.00	0.00	0.00	6.18	0.00	0.00	0.00	0.00	0.00
ACY	0.69	1.38	1.15	1.25	1.05	0.00	0.58	0.00	0.00	0.82	0.00
ACE	3.30	2.90	2.45	0.00	0.80	0.00	6.02	7.80	4.14	2.44	2.90
FLU	20.56	27.78	34.34	20.09	16.50	19.50	14.82	12.68	14.04	10.56	0.00
PHE	66.22	80.26	76.26	0.00	37.82	0.00	25.24	25.92	14.95	18.71	23.97
ANT	2.60	3.94	4.74	3.69	3.20	4.26	3.29	2.99	2.36	2.56	2.47
FLA	13.78	22.43	23.21	19.36	8.00	23.44	16.47	11.33	10.28	7.30	0.00
PYR	16.67	21.18	21.35	34.59	5.80	27.09	8.90	8.56	5.30	7.15	4.44
BaANT	1.83	2.67	2.25	2.21	0.89	3.29	1.33	1.13	0.77	0.88	1.15
CHR	3.94	6.11	4.66	5.78	2.32	17.21	3.48	3.60	1.81	1.89	4.89
BbFLA	7.18	1.06	10.79	12.78	6.34	18.64	8.36	2.67	5.08	5.16	6.14
BkFLA	1.54	11.75	2.42	2.90	0.98	1.36	1.21	6.47	0.68	0.76	8.89
BaPYR	1.96	3.73	2.37	2.99	1.14	2.97	1.76	1.09	0.93	0.88	0.61
IcdP	3.03	5.93	4.63	16.07	3.15	11.27	3.96	8.01	7.11	2.64	6.32
DahA	0.59	0.89	0.75	0.00	0.00	0.00	0.52	0.00	0.00	0.52	0.00
BghiP	4.47	6.72	6.94	12.51	3.31	10.90	4.57	6.53	17.88	3.04	4.48
DBT	17.81	15.15	15.75	14.42	6.93	5.33	12.17	7.52	9.02	9.61	0.00
C1-DBT	33.88	72.51	24.39	153.90	45.77	136.75	19.93	74.65	69.75	27.42	0.00
C2-DBT	30.36	100.87	29.11	171.24	64.42	179.83	19.81	83.97	101.67	32.92	0.00
C3-DBT	0.00	166.14	34.94	339.92	122.25	165.93	27.72	157.12	132.09	41.49	0.00
C4-DBT	165.99	176.42	29.97	397.46	150.24	304.30	63.94	153.01	189.07	91.20	0.00
1-MeNAPH	0.00	0.00	0.00	0.00	0.00	3.07	0.00	0.00	0.00	0.00	0.00
2-MeNAPH	0.00	0.00	0.00	0.00	0.00	1.05	0.00	0.00	0.00	0.00	0.00
3-MePHE	20.92	57.39	12.96	0.00	31.66	0.00	5.11	15.25	8.43	5.51	11.97
2-MePHE	16.39	18.00	17.77	0.00	7.11	0.00	5.27	6.29	4.72	4.71	16.91
9/4-MePHE	12.93	8.28	7.96	0.00	1.55	0.00	2.67	4.73	2.62	3.01	3.21

1-MePHE	14.25	12.53	14.22	0.00	6.38	0.00	6.21	7.42	4.26	4.06	5.94
3,6-DiMePHE	7.14	6.68	6.90	0.00	1.05	0.00	0.71	1.54	1.00	0.81	2.96
2,6-DiMePHE	3.91	4.60	4.23	0.00	1.12	0.00	0.90	1.73	1.27	0.77	5.25
1,7-DiMePHE	7.04	9.03	7.61	0.00	4.68	0.00	3.20	4.41	2.40	1.76	5.28
1,8-DiMePHE	3.07	3.42	3.13	0.00	1.31	0.00	0.71	2.02	0.80	0.80	3.75
Retene	7.79	17.87	15.21	7.17	3.94	6.51	8.71	6.95	5.93	4.08	0.00
C2-NAPH	0.00	0.00	0.00	14.53	0.00	59.03	0.00	16.96	8.35	0.00	19.73
C3-NAPH	0.00	0.00	0.00	0.00	0.00	21.34	0.00	0.00	1.31	0.00	1.19
C4-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-FLU	33.03	46.08	50.29	113.88	39.40	133.81	20.18	46.62	48.91	18.30	0.00
C2-FLU	116.70	153.33	91.61	1529.07	131.16	2102.22	46.74	218.03	325.74	46.69	0.00
C3-FLU	103.91	226.75	50.15	480.34	209.31	379.86	69.96	182.80	225.26	64.98	0.00
C1-PHE/ANT	71.81	130.75	60.33	0.00	71.00	0.00	23.22	64.25	44.04	25.73	376.25
C2-PHE/ANT	44.58	47.95	46.88	0.00	27.58	0.00	17.35	14.22	25.03	18.36	115.34
C3-PHE/ANT	49.23	47.25	29.62	0.00	18.35	0.00	16.43	50.61	15.66	16.81	127.09
C4-PHE/ANT	126.29	302.12	69.43	0.00	250.33	0.00	122.39	192.79	227.70	79.21	552.08
C1-FLA/PYR	24.03	16.72	19.57	26.36	13.83	37.12	20.02	15.67	23.69	13.93	0.00
C2-FLA/PYR	25.03	37.52	26.02	46.39	21.31	41.38	15.48	18.03	26.48	12.17	0.00
C3-FLA/PYR	0.00	0.00	7.91	6.56	2.16	19.01	8.39	5.67	7.95	3.72	0.00
C4-FLA/PYR	0.00	10.72	7.84	12.00	7.32	0.00	7.62	0.00	9.47	4.95	0.00
C1-BaA/CHR	3.83	7.50	6.00	3.99	3.18	16.54	3.18	3.38	3.80	1.79	6.08
C2-BaA/CHR	7.57	23.50	9.02	57.07	13.45	91.47	6.53	17.79	21.82	5.58	34.06
C3-BaA/CHR	6.48	28.04	10.62	18.78	9.66	0.00	9.43	6.29	4.98	5.77	13.53
C4-BaA/CHR	0.00	0.78	0.00	32.44	0.00	0.00	0.00	8.41	4.11	1.55	0.00
C1-BFLA/BPYR	0.16	3.02	1.46	6.31	1.99	20.56	5.11	6.74	6.47	3.41	16.09
C2-BFLA/BPYR	0.81	2.37	2.32	3.01	1.14	5.44	1.28	1.49	1.16	4.34	4.16

Table A.10: Concentrations of PAHs in Ethel Lake, Cold Lake, AB (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.50	1.50	2.50	4.50	5.50	6.50	7.50	8.50	10.50	11.50	18.50	26.00	32.00	44.00
%OC	32.94	30.50	29.43	27.76	24.38	27.14	29.58	26.50	24.96	16.74	19.22	13.11	21.59	13.18
PAH:														
NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	35.32	37.67	0.00	0.00
ACY	0.00	0.00	0.00	0.00	25.09	44.66	0.00	313.98	116.32	0.00	17.57	129.80	247.91	0.00
ACE	8.40	0.19	0.70	0.15	7.04	0.00	0.82	38.96	21.64	0.56	4.10	14.09	25.04	66.74
FLU	2.59	9.17	8.42	10.13	11.76	13.65	15.60	19.57	21.42	12.41	4.76	7.58	6.90	55.00
PHE	43.36	13.18	9.25	14.54	18.69	13.54	16.27	43.32	27.80	15.75	7.46	19.52	21.17	162.15
ANT	0.00	0.11	2.44	2.81	4.88	7.29	3.47	13.83	12.91	3.49	5.30	14.75	12.11	134.82
FLA	13.36	10.46	12.67	12.66	15.61	13.74	15.92	16.06	13.46	9.81	6.09	5.60	7.04	26.93
PYR	10.54	0.00	6.44	6.04	10.24	6.83	6.42	12.28	6.82	2.50	0.29	1.94	3.50	22.40
BaANT	0.00	1.98	2.73	2.69	2.78	3.61	3.17	0.00	3.79	1.92	0.13	2.04	0.13	0.13
CHR	0.00	4.64	5.69	5.46	4.64	5.60	6.16	0.00	8.57	4.82	3.86	5.41	5.50	14.90
BbFLA	0.00	5.04	9.80	11.27	11.69	12.37	11.87	10.03	6.77	6.73	3.73	4.33	4.30	19.04
BkFLA	0.00	0.73	3.97	4.26	4.34	4.08	4.39	0.00	3.04	2.42	0.15	0.15	0.15	0.15
BaPYR	0.00	0.00	6.18	5.07	6.50	5.31	6.73	3.40	6.75	4.34	0.41	0.41	0.41	0.41
IcdP	0.00	0.00	7.91	7.08	7.70	8.24	8.21	0.00	0.10	0.18	0.18	0.18	0.18	1.08
DahA	0.00	0.00	0.26	0.03	0.03	0.21	0.14	0.00	0.15	0.26	0.26	0.26	0.26	0.26
BghiP	0.00	7.54	8.42	9.76	12.94	11.08	16.14	11.61	11.65	8.94	3.29	4.56	3.38	17.10
DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C1-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C2-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C3-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C4-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1-MeNAPH	27.29	14.19	8.23	9.10	0.00	0.00	7.06	0.00	0.00	8.81	32.00	56.29	0.00	0.00
2-MeNAPH	6.47	5.69	4.06	4.46	0.00	0.00	2.81	0.00	0.00	3.53	19.55	34.78	0.00	0.00
3-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
2-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
9/4-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A

1-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
3,6-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
2,6-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1,7-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1,8-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Retene	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C2-NAPH	76.43	65.72	0.35	0.03	2.12	2.12	39.24	2.12	2.12	35.18	91.18	0.35	2.12	2.12
C3-NAPH	96.08	71.61	0.35	0.03	2.12	2.12	54.16	2.12	2.12	26.98	0.35	0.35	2.12	2.12
C4-NAPH	0.00	0.00	0.35	0.03	2.12	2.12	3.63	2.12	2.12	5.04	0.35	0.35	2.12	2.12
C1-FLU	0.00	0.00	0.00	0.00	0.00	0.00	6.18	0.00	0.00	3.84	0.00	8.04	9.21	46.00
C2-FLU	15.05	8.68	13.93	10.89	11.55	5.31	6.27	46.46	23.36	3.76	0.00	0.00	3.01	0.00
C3-FLU	0.00	0.00	0.00	0.00	0.00	0.00	0.00	59.88	0.00	0.00	0.00	19.05	13.97	0.00
C1-PHE/ANT	56.53	15.43	9.91	12.33	29.20	21.58	4.05	59.29	20.66	0.00	18.57	7.40	16.49	21.55
C2-PHE/ANT	0.00	20.50	0.82	7.94	18.15	10.56	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-PHE/ANT	0.00	3.19	0.00	0.00	13.73	3.91	2.68	0.00	0.00	0.00	3.89	0.00	0.00	0.00
C4-PHE/ANT	0.00	0.00	0.43	0.04	0.05	0.35	0.23	0.00	0.25	0.42	0.43	0.43	0.43	0.42
C1-FLA/PYR	0.00	0.00	0.91	1.14	0.03	0.20	0.13	0.00	0.15	0.25	0.25	0.25	0.25	0.25
C2-FLA/PYR	0.00	0.00	0.25	0.02	0.03	0.20	0.13	0.00	0.15	0.25	0.25	0.25	0.25	0.25
C3-FLA/PYR	0.00	0.00	0.25	0.02	0.03	0.20	0.13	0.00	0.15	0.25	0.25	0.25	0.25	0.25
C4-FLA/PYR	0.00	0.00	0.25	0.02	0.03	0.20	0.13	0.00	0.15	0.25	0.25	0.25	0.25	0.25
C1-BaA/CHR	0.00	0.00	0.13	0.01	0.01	0.11	0.07	10.41	15.08	0.13	0.13	0.13	0.13	0.13
C2-BaA/CHR	0.00	0.00	0.13	0.01	0.01	0.11	0.07	0.00	0.08	0.13	0.13	0.13	0.13	0.13
C3-BaA/CHR	0.00	0.00	0.13	0.01	0.01	0.11	0.07	0.00	0.08	0.13	0.13	0.13	0.13	0.13
C4-BaA/CHR	0.00	0.00	0.13	0.01	0.01	0.11	0.07	0.00	0.08	0.13	0.13	0.13	0.13	0.13
C1-BFLA/BPYR	0.00	0.00	0.41	0.04	0.04	0.34	0.22	0.00	0.24	0.41	0.41	0.41	0.41	0.41
C2-BFLA/BPYR	0.00	0.00	0.41	0.04	0.04	0.34	0.22	0.00	0.24	0.41	0.41	0.41	0.41	0.41

Table A.11: Concentrations of PAHs in Hilda Lake, Cold Lake, AB (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.50	1.50	2.50	3.50	4.50	5.50	6.50	7.50	12.50	16.50	37.00
%OC	28.07	26.70	26.13	26.15	27.04	27.11	27.18	27.24	30.51	26.39	23.48
PAH:											
NAPH	294.84	37.37	0.00	28.80	0.00	0.00	35.03	22.32	36.75	29.26	0.00
ACY	260.06	0.00	1082.75	0.00	27.76	0.00	0.00	0.00	0.00	0.00	0.00
ACE	52.76	0.00	183.31	0.00	25.43	0.00	0.00	0.00	0.00	0.00	0.00
FLU	14.30	3.44	4.60	1.15	8.48	6.99	10.33	7.30	15.37	17.33	26.62
PHE	94.63	0.00	45.27	0.00	10.57	0.00	0.00	0.00	0.86	6.94	15.60
ANT	13.83	0.00	0.00	0.11	5.09	0.00	8.46	1.42	4.77	6.68	12.57
FLA	18.04	3.55	0.00	4.31	9.27	6.05	6.38	8.41	9.41	8.94	7.67
PYR	24.20	0.00	16.70	0.00	4.60	1.20	0.00	0.00	0.00	0.00	0.00
BaANT	0.00	0.00	0.00	0.00	1.62	0.00	2.32	2.16	1.11	1.71	3.27
CHR	0.00	0.00	0.00	0.00	5.63	0.00	4.06	4.48	3.87	1.73	4.59
BbFLA	0.00	0.00	0.00	0.00	3.51	0.00	12.73	9.43	3.02	4.19	3.72
BkFLA	0.00	0.00	0.00	0.00	0.00	0.00	6.11	3.20	0.62	1.22	0.00
BaPYR	0.00	0.00	0.00	0.00	0.00	0.00	14.98	4.21	0.42	1.31	0.00
IcdP	0.00	0.00	0.00	0.00	0.00	0.00	64.61	6.88	10.08	24.59	0.00
DahA	0.00	11.02	0.00	13.54	0.00	0.00	68.59	0.00	13.44	25.75	0.00
BghiP	40.49	26.65	0.00	23.82	0.00	0.00	75.99	7.36	17.67	29.68	1.91
DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C1-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C2-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C3-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C4-DBT	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1-MeNAPH	84.77	13.70	0.00	9.51	0.00	0.00	9.50	6.73	7.92	5.23	0.00
2-MeNAPH	51.85	4.87	0.00	3.64	0.00	0.00	4.12	2.96	3.05	2.08	0.00
3-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
2-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
9/4-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A

1-MePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
3,6-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
2,6-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1,7-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1,8-DiMePHE	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Retene	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
C2-NAPH	0.00	19.11	0.00	16.52	0.00	0.00	13.88	10.94	13.89	0.00	0.00
C3-NAPH	0.00	0.00	0.00	10.34	0.00	0.00	0.00	0.00	6.25	0.00	0.00
C4-NAPH	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-FLU	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-FLU	0.00	0.00	97.95	22.12	213.50	0.00	0.00	0.00	0.00	0.00	0.00
C3-FLU	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-PHE/ANT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-PHE/ANT	0.00	0.00	236.81	0.00	287.18	0.00	0.00	0.00	0.00	0.00	0.00
C3-PHE/ANT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-PHE/ANT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-FLA/PYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-FLA/PYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-FLA/PYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-FLA/PYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C3-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C4-BaA/CHR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C1-BFLA/BPYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C2-BFLA/BPYR	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table A.12: Concentrations of PAHs in lake NE20, Athabasca oil sands, AB (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.25	1.25	2.25	3.25	4.25	5.25	6.25	7.25	8.25	9.25	10.25
%OC	24.10	26.50	23.20	28.20	33.90	34.40	24.20	32.10	20.40	19.20	26.07
PAH:											
NAPH	31.07	25.45	23.91	17.64	17.83	17.10	13.63	11.39	7.29	5.83	8.19
ACY	1.06	2.01	2.11	0.64	0.80	1.54	1.38	2.02	0.34	0.75	0.33
ACE	1.41	2.72	1.57	1.02	1.37	2.14	2.52	2.08	2.36	0.96	1.01
FLU	8.71	8.29	10.00	7.76	6.76	7.03	6.45	5.13	4.17	3.21	3.00
PHE	79.22	65.63	68.04	47.70	38.87	34.09	28.60	20.33	15.13	11.65	11.04
ANT	30.00	21.50	23.60	17.40	12.80	10.70	9.08	6.15	4.08	2.88	2.91
FLA	16.88	14.72	18.09	11.90	9.92	9.14	7.51	5.66	4.80	3.96	4.10
PYR	86.85	70.46	72.23	52.96	42.48	34.49	29.10	19.78	13.38	10.72	9.40
BaANT	123.00	98.50	102.00	76.50	51.70	49.70	41.40	26.00	15.60	11.50	9.67
CHR	188.87	152.24	161.21	120.33	89.76	77.06	61.67	40.34	25.44	19.18	15.48
BbFLA	45.30	38.50	38.00	31.00	23.20	18.50	16.70	11.70	7.87	7.13	6.38
BkFLA	23.01	14.90	21.47	15.69	9.32	10.43	7.74	3.01	3.02	2.11	0.37
BaPYR	128.00	105.00	107.00	81.10	62.00	52.10	38.80	27.10	17.00	11.50	10.80
IcdP	42.20	36.30	37.00	27.20	21.00	17.80	14.00	9.54	6.85	5.04	4.13
DahA	28.40	23.80	24.50	19.20	14.80	12.50	10.80	8.06	5.95	5.25	5.65
BghiP	87.70	76.80	74.30	60.20	39.80	36.10	29.10	19.80	13.80	10.50	9.46
DBT	67.60	54.30	53.90	37.40	30.60	26.30	21.50	15.50	10.70	8.46	6.91
C1-DBT	254.00	213.00	211.00	157.00	115.00	95.60	76.70	52.20	31.00	24.80	18.90
C2-DBT	512.51	419.00	449.96	386.12	303.15	195.16	168.16	105.26	74.57	50.62	37.31
C3-DBT	439.78	332.47	355.26	287.14	213.29	170.36	148.38	99.94	66.74	50.12	39.40
C4-DBT	244.99	191.92	142.61	129.89	114.12	75.21	58.04	33.46	21.07	14.88	11.74
1-MeNAPH	26.91	18.22	19.74	15.47	14.53	12.95	9.76	7.47	6.28	4.75	4.91
2-MeNAPH	61.11	45.62	46.02	33.12	28.29	27.02	23.03	16.68	11.68	10.50	9.09
3-MePHE	59.60	43.80	47.00	33.90	29.10	24.00	20.40	14.00	8.81	6.62	5.40
2-MePHE	93.10	80.10	80.50	58.70	42.40	35.00	28.50	19.00	13.00	9.12	7.96
9/4-MePHE	40.60	30.20	32.80	25.00	22.20	20.00	16.60	12.10	8.38	6.72	5.68

1-MePHE	34.70	31.80	34.90	25.30	18.20	13.90	12.60	8.38	6.70	5.17	4.27
3,6-DiMePHE	19.10	13.80	15.40	11.10	9.89	7.48	6.76	4.58	3.40	2.85	2.41
2,6-DiMePHE	28.65	23.68	26.31	19.61	13.37	11.29	9.60	5.94	3.48	2.58	1.68
1,7-DiMePHE	51.25	34.56	39.24	27.34	24.76	20.96	20.37	13.53	11.83	7.71	6.87
1,8-DiMePHE	8.83	6.84	6.48	4.59	3.98	4.32	3.44	1.80	0.71	0.81	0.61
Retene	71.60	29.70	30.10	26.30	41.90	61.10	88.50	61.70	40.50	27.80	28.90
C2-NAPH	814.97	168.36	283.17	113.16	218.07	144.44	174.56	128.74	191.77	81.39	116.22
C3-NAPH	70.34	61.23	55.35	48.61	41.02	43.31	34.94	29.18	25.29	21.96	24.42
C4-NAPH	0.00	19.90	3.80	3.82	2.98	0.00	0.00	9.54	0.00	0.00	0.00
C1-FLU	32.21	36.98	40.55	33.42	27.79	17.76	20.39	16.80	9.51	13.02	8.09
C2-FLU	87.19	61.50	77.18	51.37	50.86	44.20	32.41	28.42	21.82	24.58	18.87
C3-FLU	88.68	86.16	100.00	75.82	61.00	46.61	38.05	27.31	25.52	17.42	15.17
C1-PHE/ANT	262.00	214.00	226.00	165.00	127.00	106.00	88.50	60.30	41.20	30.90	25.70
C2-PHE/ANT	269.78	203.15	229.96	167.73	123.87	111.92	95.94	65.13	44.24	34.48	29.06
C3-PHE/ANT	202.09	157.37	161.27	113.69	104.76	78.09	71.30	45.17	38.27	26.50	22.29
C4-PHE/ANT	2113.78	1699.09	1718.68	1210.40	954.71	802.84	657.88	402.98	259.99	198.55	152.49
C1-FLA/PYR	495.62	422.40	421.34	317.59	234.64	194.66	164.66	114.82	73.33	54.11	43.10
C2-FLA/PYR	517.26	420.48	515.39	355.78	245.86	197.89	185.90	117.15	74.75	55.18	52.47
C3-FLA/PYR	83.11	161.37	221.24	120.81	66.01	75.05	61.47	32.23	29.04	16.12	7.80
C4-FLA/PYR	31.82	84.29	108.50	91.98	44.85	39.48	54.59	29.63	24.73	20.75	12.94
C1-BaA/CHR	635.28	537.51	554.49	414.57	300.59	256.59	214.59	134.64	85.44	65.67	53.86
C2-BaA/CHR	467.43	364.27	309.21	249.48	204.53	163.55	126.56	93.13	54.73	41.92	34.01
C3-BaA/CHR	70.55	56.55	59.25	51.84	40.01	30.34	27.95	18.60	9.70	9.42	5.21
C4-BaA/CHR	38.31	26.70	24.15	24.59	10.21	14.86	11.37	8.29	6.14	4.31	2.78
C1-BFLA/BPYR	468.64	459.41	416.35	310.60	209.65	194.66	149.67	103.83	66.13	51.91	39.50
C2-BFLA/BPYR	282.23	236.14	208.07	151.36	89.81	95.84	67.34	42.13	37.93	29.23	20.72

NE20, cont'd

Interval Mid-Pt. depth (cm)	11.25	13.25	14.25	15.25	16.25	17.25	19.25	19.75
%OC	21.80	31.70	28.40	26.40	24.40	26.60	31.00	32.10
PAH:								
NAPH	3.87	4.82	4.79	7.80	6.59	5.21	8.17	5.06
ACY	0.31	0.65	0.49	0.29	0.52	0.82	0.58	0.46
ACE	2.15	3.23	1.15	1.88	1.54	1.38	1.21	2.18
FLU	4.05	5.97	5.96	5.41	7.02	6.51	4.77	3.72
PHE	10.48	13.23	10.84	12.00	12.54	11.75	10.27	9.17
ANT	2.12	4.38	1.83	1.78	2.49	2.12	1.97	1.99
FLA	4.71	7.22	6.47	6.31	7.71	7.94	7.78	6.54
PYR	7.08	7.86	8.31	7.06	8.24	7.60	7.39	6.42
BaANT	6.18	5.74	4.54	4.74	5.05	4.34	4.54	4.56
CHR	10.46	8.27	6.79	7.79	7.90	5.21	5.82	4.95
BbFLA	4.22	6.26	5.42	5.91	6.23	6.98	7.99	6.79
BkFLA	1.91	0.94	1.65	2.03	0.70	2.81	3.06	3.58
BaPYR	7.84	6.76	5.23	5.38	5.43	4.98	5.94	5.14
IcdP	1.77	1.75	2.16	2.24	1.95	2.03	1.75	1.84
DahA	5.09	7.10	6.35	5.75	7.57	8.73	10.30	9.61
BghiP	7.92	7.58	7.49	8.01	8.63	8.64	10.70	9.18
DBT	5.08	6.97	3.84	3.53	3.65	3.65	3.75	3.30
C1-DBT	13.00	12.60	7.70	5.79	7.90	2.78	5.86	5.93
C2-DBT	30.09	24.68	17.63	17.95	17.55	17.45	13.36	14.11
C3-DBT	23.37	22.26	11.22	11.76	15.16	12.58	12.92	11.36
C4-DBT	8.65	3.76	3.94	2.47	12.54	4.76	2.53	1.84
1-MeNAPH	3.06	3.34	2.91	3.48	3.19	3.11	3.24	2.40
2-MeNAPH	5.36	4.82	4.60	5.33	5.56	5.39	5.55	3.91
3-MePHE	4.21	3.92	3.05	3.30	3.27	3.25	3.12	3.31
2-MePHE	5.68	7.00	3.97	4.07	4.50	3.79	3.59	3.54

9/4-MePHE	4.99	6.04	5.47	5.38	6.56	6.55	5.93	5.77
1-MePHE	3.86	6.78	5.26	5.03	6.50	6.07	5.42	5.22
3,6-DiMePHE	1.66	3.59	2.08	2.30	2.35	2.40	2.23	2.12
2,6-DiMePHE	1.03	0.85	0.50	0.42	1.08	0.21	0.22	0.24
1,7-DiMePHE	4.12	6.50	7.08	8.31	9.29	9.50	6.45	4.35
1,8-DiMePHE	0.49	0.57	0.56	0.43	0.57	0.53	0.70	1.29
Retene	26.50	35.60	42.20	42.30	45.30	34.20	27.30	22.90
C2-NAPH	37.59	62.69	47.09	135.33	113.00	94.10	76.36	29.65
C3-NAPH	20.25	17.34	15.19	18.48	18.91	21.13	16.99	12.52
C4-NAPH	0.00	0.00	0.00	0.00	0.00	3.08	0.31	0.00
C1-FLU	5.85	15.37	9.76	13.62	11.80	12.62	14.87	9.64
C2-FLU	20.49	25.65	23.30	22.48	24.26	20.27	22.90	20.53
C3-FLU	16.54	17.58	8.84	17.09	10.86	19.39	7.57	11.90
C1-PHE/ANT	20.70	26.80	17.80	19.20	22.60	19.70	20.20	14.50
C2-PHE/ANT	15.55	21.11	19.04	16.07	23.40	20.31	17.15	12.97
C3-PHE/ANT	19.23	28.94	29.56	33.83	37.01	30.72	21.54	21.15
C4-PHE/ANT	115.24	138.03	108.55	115.84	123.66	99.69	95.78	92.26
C1-FLA/PYR	32.46	36.68	33.66	34.70	36.28	28.59	29.30	27.77
C2-FLA/PYR	27.81	33.13	31.35	31.02	27.61	28.92	30.74	23.05
C3-FLA/PYR	3.67	3.72	5.35	5.42	0.00	5.63	14.00	3.42
C4-FLA/PYR	10.39	10.72	5.18	9.80	16.32	15.02	10.84	5.62
C1-BaA/CHR	35.55	31.40	24.52	25.94	28.04	20.84	23.74	19.46
C2-BaA/CHR	18.17	15.98	15.66	12.51	12.76	13.57	13.58	9.86
C3-BaA/CHR	4.23	4.96	3.05	4.54	3.83	4.09	3.53	2.22
C4-BaA/CHR	5.28	3.63	0.00	7.23	4.90	4.22	6.56	2.91
C1-BFLA/BPYR	20.67	22.99	13.67	14.81	13.69	7.31	13.81	5.54
C2-BFLA/BPYR	16.27	14.07	9.86	14.41	11.43	13.34	12.45	6.90

Table A.13: Concentrations of PAHs in lake NW35 Athabasca oil sands, AB (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.25	0.75	1.25	1.75	2.25	2.75	3.25	3.75	4.25	4.75	5.25
%OC	37.40	37.40	38.28	39.15	40.03	40.90	42.70	40.20	4.16	24.63	45.10
PAH:											
NAPH	14.01	13.09	21.23	19.93	27.42	16.90	14.81	32.72	0.00	0.00	29.89
ACY	1.16	0.93	3.39	3.70	3.57	1.22	2.59	6.62	2.80	1.96	2.74
ACE	1.92	2.87	3.82	4.04	9.98	2.24	2.85	7.50	8.04	6.64	6.38
FLU	12.10	28.70	11.80	17.00	18.60	14.70	15.20	21.10	25.40	19.40	24.60
PHE	30.07	25.74	27.26	37.54	38.71	25.74	24.21	37.89	38.40	29.66	34.25
ANT	5.59	4.44	5.63	5.43	4.68	3.10	3.65	8.64	5.46	4.62	4.30
FLA	6.43	5.77	6.73	9.26	9.05	6.48	5.65	11.98	9.72	9.11	11.02
PYR	12.14	10.64	10.11	14.36	13.16	10.45	7.54	12.86	11.31	9.08	10.21
BaANT	9.93	9.05	8.68	10.24	9.72	6.69	6.02	6.01	8.53	5.47	4.69
CHR	21.50	20.21	19.59	24.23	23.23	17.92	14.34	19.41	17.88	13.06	13.38
BbFLA	7.37	7.35	6.50	6.49	7.39	7.21	5.18	6.72	7.61	6.72	4.46
BkFLA	2.82	2.57	2.69	4.53	3.68	3.09	1.77	4.14	6.45	2.52	4.28
BaPYR	13.60	12.50	11.00	14.70	13.80	9.48	9.15	9.28	9.40	5.59	5.76
IcdP	4.91	4.51	2.55	2.18	7.26	2.35	0.87	4.35	2.88	1.58	3.66
DahA	4.91	2.95	0.36	4.56	3.63	0.35	3.50	0.00	7.53	5.12	5.54
BghiP	8.41	9.74	8.54	10.79	10.00	7.15	5.93	7.52	9.97	7.03	5.09
DBT	12.50	10.40	10.40	12.40	11.70	8.26	7.44	10.90	9.86	8.37	7.88
C1-DBT	30.20	19.60	26.60	24.90	28.20	21.70	17.40	8.49	3.97	15.80	15.40
C2-DBT	59.97	47.50	52.43	61.60	62.59	41.24	35.83	38.41	25.72	16.07	19.90
C3-DBT	49.22	48.88	53.55	62.86	57.85	34.68	30.92	30.46	24.66	20.54	17.69
C4-DBT	1.10	5.46	3.73	10.46	9.03	8.63	3.42	0.00	4.01	6.93	0.00
1-MeNAPH	8.58	8.71	9.55	12.17	11.80	10.01	8.40	16.02	13.62	11.64	12.74
2-MeNAPH	15.63	15.99	16.08	18.71	20.88	15.50	14.68	24.38	19.32	19.15	19.47
3-MePHE	15.10	12.50	14.00	17.70	18.40	10.20	9.79	12.40	10.40	10.40	8.86
2-MePHE	15.80	14.30	13.50	17.70	16.90	10.90	10.70	18.30	12.20	10.90	11.30
9/4-MePHE	10.80	9.37	11.20	14.10	13.90	10.40	9.55	14.90	11.20	11.10	10.70

1-MePHE	14.10	11.80	10.90	15.50	14.50	9.88	10.10	18.20	13.80	12.50	14.80
3,6-DiMePHE	3.95	2.99	3.48	4.37	3.18	2.73	1.84	2.74	3.34	2.80	2.92
2,6-DiMePHE	3.48	2.02	3.59	4.38	4.96	3.13	2.63	2.76	3.98	2.71	4.26
1,7-DiMePHE	13.10	9.57	9.01	13.60	12.80	9.11	9.81	14.50	12.90	10.70	13.10
1,8-DiMePHE	0.00	0.00	1.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.07
Retene	93.70	93.20	104.00	124.00	122.00	98.80	99.00	150.00	112.00	158.00	125.00
C2-NAPH	141.19	85.86	1718.96	103.43	102.25	60.34	121.09	55.19	288.70	87.02	164.73
C3-NAPH	55.56	29.56	50.76	52.01	62.22	0.00	35.82	79.14	60.80	53.50	55.62
C4-NAPH	27.08	14.57	30.18	11.54	21.60	17.30	23.93	43.40	27.91	24.87	48.55
C1-FLU	48.48	37.53	47.95	53.27	73.37	50.61	62.52	56.18	47.47	32.59	33.00
C2-FLU	35.50	42.04	47.99	41.49	69.45	40.03	48.62	80.25	55.13	44.32	45.96
C3-FLU	38.12	38.12	0.00	52.07	65.16	39.63	40.56	47.46	42.33	42.17	23.08
C1-PHE/ANT	40.80	21.20	22.10	47.20	28.40	30.50	19.70	63.80	25.00	34.50	45.80
C2-PHE/ANT	33.61	52.70	39.74	52.59	44.08	39.40	42.28	42.30	50.64	26.89	31.11
C3-PHE/ANT	38.95	28.60	42.33	57.74	42.53	34.83	32.89	45.06	32.31	37.26	48.71
C4-PHE/ANT	306.42	253.23	274.81	412.43	316.46	229.13	230.23	276.19	217.89	300.45	239.75
C1-FLA/PYR	69.39	54.99	60.68	98.27	79.21	56.37	48.06	67.44	66.56	49.72	46.47
C2-FLA/PYR	85.32	70.56	76.21	110.66	99.53	72.35	72.70	87.46	72.95	60.53	63.90
C3-FLA/PYR	33.55	12.84	6.34	15.14	19.20	11.86	5.91	4.51	5.12	14.59	19.37
C4-FLA/PYR	35.67	23.70	23.43	44.13	39.94	33.36	33.76	36.96	49.36	58.98	36.03
C1-BaA/CHR	76.10	66.50	68.70	84.90	77.60	59.60	51.90	71.70	58.30	48.80	44.20
C2-BaA/CHR	62.38	53.12	56.21	69.53	76.22	59.04	48.47	64.63	51.99	49.39	48.70
C3-BaA/CHR	3.14	7.70	9.34	9.16	8.49	15.40	9.59	2.75	2.51	3.67	1.37
C4-BaA/CHR	16.20	16.40	25.00	15.20	23.70	12.00	21.40	19.90	17.90	13.20	7.04
C1-BFLA/BPYR	42.10	41.70	43.30	59.80	37.70	30.00	29.10	42.50	32.80	20.60	18.50
C2-BFLA/BPYR	22.20	33.50	54.70	31.10	20.30	26.10	19.90	40.50	30.30	36.10	25.10

NW35, cont'd

Interval Mid-Pt. depth (cm)	5.75	6.25	6.75	7.25	7.75	8.25	8.75	9.25	9.75
%OC	43.90	44.40	44.50	45.00	45.30	45.60	45.90	45.50	46.15
PAH:									
NAPH	12.94	18.26	13.88	13.02	20.87	20.61	22.60	10.92	16.97
ACY	1.47	2.57	1.16	1.27	2.37	2.68	3.95	1.32	0.92
ACE	4.29	7.05	6.10	5.46	8.44	8.10	10.80	6.96	9.32
FLU	18.10	25.40	21.40	22.50	37.20	31.00	25.30	34.70	42.70
PHE	24.09	29.43	25.77	25.80	39.34	34.50	35.62	33.21	44.00
ANT	2.93	3.50	3.23	3.54	5.42	4.79	5.66	4.61	6.21
FLA	6.92	10.19	8.02	8.46	11.55	11.35	12.09	10.44	14.39
PYR	6.58	8.11	9.25	6.40	9.62	9.40	8.99	8.37	10.24
BaANT	2.50	3.35	2.66	2.35	3.35	3.43	1.97	2.51	2.90
CHR	7.69	10.74	8.15	7.03	10.00	8.60	8.86	8.37	9.42
BbFLA	4.75	5.53	3.19	4.59	6.25	4.15	1.18	4.98	5.67
BkFLA	1.90	1.43	3.19	0.38	2.51	2.60	2.70	2.23	3.36
BaPYR	3.29	3.33	3.24	1.56	1.02	1.03	1.99	2.34	2.95
IcdP	2.50	1.34	1.34	1.12	0.97	0.94	1.58	0.81	1.17
DahA	3.38	4.39	3.54	3.16	5.16	3.60	3.19	4.92	6.21
BghiP	3.77	4.46	4.38	3.80	3.18	4.58	3.91	4.56	5.54
DBT	4.63	6.13	5.24	4.85	6.57	5.83	5.24	3.96	5.17
C1-DBT	9.82	10.70	10.90	10.60	15.20	10.60	13.60	8.76	13.50
C2-DBT	10.84	10.10	9.67	5.23	9.68	6.69	0.00	3.03	2.71
C3-DBT	12.09	13.15	7.65	5.16	9.48	2.08	2.45	2.83	3.50
C4-DBT	10.71	0.00	2.19	0.00	0.00	4.46	3.76	3.05	0.00
1-MeNAPH	6.92	8.05	6.57	6.85	8.59	6.61	9.73	5.72	7.08
2-MeNAPH	10.64	12.49	11.13	10.70	14.37	10.71	12.33	7.89	9.15
3-MePHE	6.13	6.84	6.34	4.28	6.74	4.79	5.87	4.22	5.72
2-MePHE	6.58	7.71	7.93	6.39	8.62	7.63	9.57	6.44	8.37

9/4-MePHE	7.12	9.43	9.46	7.68	10.60	9.28	10.50	7.95	8.25
1-MePHE	8.81	12.70	11.50	10.00	15.20	12.20	15.60	12.80	18.10
3,6-DiMePHE	2.12	2.81	2.68	1.78	2.70	2.54	2.39	2.40	3.01
2,6-DiMePHE	3.48	3.22	2.96	2.79	4.12	3.22	3.30	3.35	4.46
1,7-DiMePHE	7.51	10.30	9.34	7.81	12.10	12.90	12.80	11.90	17.10
1,8-DiMePHE	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.21
Retene	85.70	110.00	112.00	106.00	166.00	164.00	168.00	149.00	215.00
C2-NAPH	46.61	76.97	41.68	37.55	58.45	89.10	29.84	42.08	63.38
C3-NAPH	30.29	42.15	36.05	31.06	37.57	44.87	45.98	28.64	42.60
C4-NAPH	16.06	19.83	8.55	9.45	19.29	16.99	30.76	13.91	28.34
C1-FLU	26.52	30.86	22.37	31.49	38.47	97.19	22.04	25.98	36.34
C2-FLU	27.65	43.22	22.54	25.85	29.37	34.37	40.33	33.12	37.54
C3-FLU	15.38	16.51	21.83	20.10	21.78	22.21	10.28	14.39	22.00
C1-PHE/ANT	22.10	28.90	35.30	28.30	41.10	33.90	41.50	31.40	40.50
C2-PHE/ANT	26.37	32.95	38.70	23.21	29.16	25.17	28.66	22.84	31.23
C3-PHE/ANT	27.37	36.88	45.07	31.31	43.17	41.84	44.25	44.48	56.35
C4-PHE/ANT	134.89	194.14	178.97	163.34	288.78	247.49	240.76	232.18	301.76
C1-FLA/PYR	28.75	41.91	43.50	30.77	71.51	37.24	34.26	37.70	52.58
C2-FLA/PYR	40.39	42.62	39.26	29.55	79.17	40.43	38.80	34.96	39.90
C3-FLA/PYR	2.61	18.86	4.27	7.63	11.97	15.44	12.02	10.26	12.76
C4-FLA/PYR	23.39	30.84	17.70	18.73	22.91	36.15	24.76	30.00	29.86
C1-BaA/CHR	28.40	34.40	28.70	26.50	31.80	30.30	28.10	26.80	34.50
C2-BaA/CHR	32.88	37.37	30.58	35.33	46.63	40.63	38.37	36.62	46.68
C3-BaA/CHR	6.78	2.09	3.65	3.61	3.47	2.08	1.96	2.30	3.17
C4-BaA/CHR	12.50	13.30	6.08	10.10	1.45	13.90	3.13	13.50	8.19
C1-BFLA/BPYR	19.10	15.20	11.40	4.18	6.32	8.31	3.66	8.20	10.70
C2-BFLA/BPYR	17.30	17.90	13.20	15.00	13.00	21.00	21.00	13.50	18.60

Table A.14: Concentrations of PAHs in lake SE22, AB (ng/g dry weight).

Interval Mid-Pt. depth (cm)	0.25	1.25	2.25	4.25	5.25	6.25	7.25	8.25	9.25	10.25	11.25
%OC	39.70	38.90	40.40	39.50	40.10	40.70	41.30	42.70	43.25	43.80	44.35
PAH:											
NAPH	14.64	8.31	21.70	15.50	12.63	9.60	7.93	9.92	9.02	8.12	9.20
ACY	4.65	1.82	1.54	1.29	2.15	1.60	1.07	1.38	0.82	2.67	0.67
ACE	4.88	4.24	1.96	2.78	2.24	2.77	2.13	1.76	1.56	6.26	1.93
FLU	5.20	4.31	4.82	27.80	7.04	6.09	5.46	7.35	4.60	6.46	7.39
PHE	30.30	28.70	22.00	25.40	21.90	17.90	15.60	21.10	29.12	19.30	17.20
ANT	3.36	5.21	3.96	3.26	4.49	1.51	3.01	2.46	2.51	2.60	1.98
FLA	7.63	5.78	4.40	6.94	7.13	5.63	5.21	6.46	10.14	5.19	4.07
PYR	14.18	16.50	9.04	15.81	12.05	10.42	9.65	10.13	14.28	8.30	5.49
BaANT	21.40	19.60	12.80	15.30	12.80	12.00	8.72	10.80	5.22	8.86	6.89
CHR	27.93	38.13	23.26	28.60	20.25	19.54	14.70	19.25	21.44	15.30	9.86
BbFLA	3.61	8.97	6.10	8.72	6.80	5.10	5.35	6.65	2.35	5.10	3.81
BkFLA	4.46	3.75	3.31	6.28	6.35	2.81	2.36	4.19	3.24	3.40	3.42
BaPYR	22.30	17.80	13.90	18.80	14.00	11.80	9.60	10.70	3.99	8.70	6.86
IcdP	4.77	3.86	4.59	6.00	3.62	2.48	2.19	1.07	2.79	1.30	1.74
DahA	12.50	8.01	8.73	11.80	7.92	8.75	5.21	7.47	3.28	7.76	6.77
BghiP	20.30	22.60	11.10	11.00	18.40	17.80	6.39	18.00	13.81	17.80	7.47
DBT	12.10	11.60	7.58	9.76	8.64	7.84	5.81	7.62	4.38	6.45	6.13
C1-DBT	3.94	13.60	13.40	7.35	17.20	17.50	14.20	8.92	6.69	4.25	3.55
C2-DBT	108.23	120.24	50.15	50.12	73.36	66.16	46.91	50.42	112.90	40.35	21.55
C3-DBT	111.67	106.81	41.49	49.09	70.18	55.63	46.06	40.33	57.63	37.40	17.79
C4-DBT	58.15	43.14	26.60	24.37	27.21	28.45	12.62	8.20	142.40	7.60	13.00
1-MeNAPH	18.30	10.60	9.26	17.20	9.31	8.89	7.63	9.99	9.73	8.32	6.00
2-MeNAPH	31.50	20.70	18.60	24.10	12.10	13.00	14.00	17.10	12.82	13.40	21.70
3-MePHE	2.93	11.30	9.25	10.60	8.25	8.27	6.22	7.09	2.43	5.46	5.44
2-MePHE	7.87	20.70	10.00	12.90	11.40	10.90	7.08	13.20	13.15	7.80	6.19
9/4-MePHE	10.90	12.70	9.06	11.00	8.68	8.74	8.06	9.80	20.23	7.47	6.69

1-MePHE	12.10	12.30	8.95	11.30	9.87	7.69	6.62	7.55	16.19	8.20	5.09
3,6-DiMePHE	4.53	2.19	3.02	3.33	4.47	3.54	1.18	1.11	8.12	1.83	1.30
2,6-DiMePHE	4.73	6.27	4.40	4.80	4.51	4.21	4.77	4.42	9.56	3.59	2.86
1,7-DiMePHE	10.30	10.70	5.77	6.74	6.71	6.77	5.70	6.57	17.13	5.92	3.57
1,8-DiMePHE	4.73	2.29	1.81	2.85	1.32	1.25	1.23	1.15	6.03	1.77	1.46
Retene	25.00	25.60	14.50	19.60	29.80	32.80	22.40	36.50	32.77	28.20	12.00
C2-NAPH	578.74	235.52	58.50	54.02	349.24	143.86	83.58	132.85	1.48	210.80	44.50
C3-NAPH	0.00	4.09	18.20	20.40	7.88	7.00	10.20	12.52	0.00	1.29	19.30
C4-NAPH	0.00	0.00	59.10	65.73	0.00	14.35	5.97	0.00	0.00	14.20	62.90
C1-FLU	1.33	5.90	14.50	1.94	11.92	13.34	9.98	8.78	6.07	5.47	0.00
C2-FLU	19.79	16.01	36.25	69.37	12.14	16.81	15.21	16.90	43.98	9.14	24.55
C3-FLU	71.93	5.94	24.80	11.37	0.00	9.45	9.53	7.16	0.00	11.19	0.00
C1-PHE/ANT	2.86	44.30	9.06	32.90	26.80	28.40	21.40	37.60	49.90	23.50	23.40
C2-PHE/ANT	43.04	41.82	18.39	38.80	35.77	35.05	31.26	31.23	133.48	21.18	8.79
C3-PHE/ANT	41.66	45.65	25.59	38.23	22.87	23.02	35.89	30.26	91.78	22.40	7.19
C4-PHE/ANT	327.00	392.00	218.00	274.00	243.00	266.00	208.00	205.00	226.55	205.00	114.00
C1-FLA/PYR	100.00	107.00	66.60	89.40	67.80	72.60	59.80	65.50	91.15	47.40	37.20
C2-FLA/PYR	139.32	122.25	82.57	109.58	70.93	81.25	51.06	73.02	61.65	58.16	38.17
C3-FLA/PYR	73.17	16.39	9.35	19.81	13.80	14.37	12.13	12.82	58.17	9.50	9.75
C4-FLA/PYR	17.20	24.80	7.80	21.30	24.20	8.55	9.99	12.50	23.65	11.30	11.80
C1-BaA/CHR	101.00	108.00	71.50	84.00	69.40	62.30	45.00	55.60	60.80	47.10	38.00
C2-BaA/CHR	5.19	8.48	45.13	55.19	6.33	5.45	0.91	14.17	6.96	4.20	16.93
C3-BaA/CHR	1.50	1.50	7.35	9.80	1.50	5.41	4.47	1.50	1.50	1.50	1.50
C4-BaA/CHR	9.49	1.64	7.31	15.70	7.47	1.13	1.99	6.32	8.96	1.38	4.58
C1-BFLA/BPYR	0.00	22.30	28.75	42.23	20.39	9.79	27.83	0.35	11.56	6.48	18.55
C2-BFLA/BPYR	3.25	10.40	19.10	34.20	2.37	9.65	2.50	2.19	2.67	6.24	15.60

SE22, cont'd

Interval Mid-Pt. depth (cm)	12.25	13.25	15.25	16.25	17.25	18.25	19.25	19.75
%OC	44.90	43.60	44.50	44.50	44.40	44.80	45.20	45.60
PAH compound:								
NAPH	12.82	8.60	14.10	9.97	12.71	10.77	8.82	10.39
ACY	0.80	1.24	0.61	1.87	0.58	0.78	0.66	0.82
ACE	5.26	7.85	6.28	8.81	7.72	7.75	9.99	8.09
FLU	6.51	8.49	9.96	9.73	10.20	7.16	8.89	7.99
PHE	17.30	25.60	15.70	33.70	25.20	14.50	18.10	17.20
ANT	2.37	2.53	1.73	2.37	2.76	1.44	0.65	1.54
FLA	4.96	7.27	5.84	6.10	7.99	5.26	7.01	6.57
PYR	7.17	1.29	3.49	3.93	9.09	5.04	5.59	5.57
BaANT	6.61	5.17	4.40	2.41	6.59	2.02	1.97	1.40
CHR	9.24	14.20	4.86	5.71	12.32	4.63	4.19	4.20
BbFLA	3.75	5.05	4.04	2.10	3.48	2.72	3.61	2.83
BkFLA	3.83	3.11	1.66	2.45	1.69	2.37	1.57	1.76
BaPYR	6.00	7.47	3.94	3.42	2.11	3.42	2.01	2.15
IcdP	1.90	3.19	0.77	0.42	1.56	0.94	1.27	0.82
DahA	6.51	8.46	6.27	4.98	5.77	4.55	4.61	3.83
BghiP	14.40	13.20	5.34	8.20	8.02	16.90	7.46	6.61
DBT	5.48	4.74	3.41	3.37	4.16	2.00	2.45	1.70
C1-DBT	6.85	3.19	3.63	4.00	7.31	2.05	2.60	2.02
C2-DBT	22.68	43.98	9.05	13.13	45.35	13.96	9.66	9.83
C3-DBT	26.25	93.83	5.29	12.62	39.65	11.49	11.32	9.49
C4-DBT	8.28	28.48	5.30	0.13	50.04	6.37	4.40	4.34
1-MeNAPH	7.29	9.19	6.12	11.20	9.61	3.49	4.66	3.99
2-MeNAPH	14.30	14.60	10.30	18.60	10.30	4.60	6.47	5.23
3-MePHE	5.29	9.07	4.02	2.85	0.31	0.34	4.19	3.21
2-MePHE	7.43	9.97	4.16	3.96	9.53	3.58	3.51	4.66

9/4-MePHE	8.16	11.90	6.85	5.67	18.70	5.56	6.20	6.69
1-MePHE	5.23	13.70	5.07	6.52	11.70	5.38	6.34	6.00
3,6-DiMePHE	2.42	4.25	1.57	1.36	4.72	0.60	1.65	1.58
2,6-DiMePHE	2.16	5.16	1.72	1.78	7.00	1.49	1.74	1.66
1,7-DiMePHE	5.61	11.60	3.57	4.00	15.00	4.11	4.88	5.74
1,8-DiMePHE	0.89	4.24	2.65	1.89	5.24	2.82	2.41	2.20
Retene	37.30	0.00	14.20	47.70	48.20	24.30	50.10	48.60
C2-NAPH	104.35	206.01	0.00	107.68	141.04	148.58	196.07	175.99
C3-NAPH	7.08	0.00	0.00	13.55	13.40	0.04	14.25	13.98
C4-NAPH	0.00	0.00	62.70	0.00	0.99	0.00	0.00	3.35
C1-FLU	1.84	8.55	35.30	3.67	18.50	8.27	13.02	10.47
C2-FLU	14.05	23.46	19.75	14.59	38.22	10.56	15.29	12.64
C3-FLU	0.00	46.74	7.40	11.04	39.71	13.32	10.62	20.83
C1-PHE/ANT	26.10	35.60	20.10	19.00	42.70	15.50	22.10	15.90
C2-PHE/ANT	19.40	55.80	11.49	12.25	76.78	11.33	15.16	22.22
C3-PHE/ANT	22.91	55.50	11.59	13.01	74.03	14.56	17.78	29.67
C4-PHE/ANT	156.00	0.00	87.40	112.00	138.00	73.80	106.00	97.80
C1-FLA/PYR	38.70	98.90	31.30	33.80	54.70	31.00	31.60	36.70
C2-FLA/PYR	40.45	64.36	23.37	26.02	39.84	22.00	26.55	25.61
C3-FLA/PYR	11.92	8.97	8.55	8.84	0.00	8.12	9.38	7.28
C4-FLA/PYR	15.60	9.04	5.14	10.20	2.26	1.02	9.83	9.95
C1-BaA/CHR	30.40	44.20	23.40	18.60	31.90	16.10	15.60	11.10
C2-BaA/CHR	1.62	2.75	12.63	0.76	0.00	0.00	0.18	0.00
C3-BaA/CHR	1.43	1.50	0.55	1.74	1.50	1.55	0.70	1.50
C4-BaA/CHR	0.83	1.10	0.66	2.40	8.05	1.98	0.74	0.72
C1-BFLA/BPYR	2.80	3.38	10.25	3.31	0.00	1.43	2.01	0.00
C2-BFLA/BPYR	6.10	0.62	12.70	7.57	1.61	4.37	4.35	1.79

Appendix B. Rock Eval pyrolysis results for all lakes I20, C23, CamH2, CamH5 in the Northwest Territories.

Table B.1: Rock Eval pyrolysis analysis results for lake I20, Mackenzie Delta Uplands, NWT.

Interval Mid-Pt. depth (cm)	S1	S2	S3	TOC	RC%
0.75	16.77	64.78	46.30	23.54	14.68
1.75	17.23	59.98	42.91	22.28	13.89
2.75	14.70	47.50	33.88	17.92	11.19
3.75	17.54	64.99	46.57	24.35	15.35
4.75	15.77	60.28	39.20	21.93	13.82
5.75	13.99	58.68	36.43	20.81	13.11
6.75	15.70	66.52	40.52	23.43	14.76
7.75	15.11	62.88	38.80	22.18	13.94
9.75	15.87	63.86	39.68	22.86	14.42
11.75	16.00	63.46	39.05	22.52	14.14
14.50	15.80	64.59	40.47	23.27	14.73
17.75	14.96	65.66	39.52	23.37	14.85

Table B.2: Rock Eval pyrolysis analysis results for lake C23, Mackenzie Delta Uplands, NWT.

Interval Mid-Pt. depth (cm)	S1	S2	S3	TOC	RC%
1.75	5.65	18.96	27.03	10.71	7.57
2.75	0.43	2.32	9.69	3.07	2.49
3.75	4.32	17.94	25.65	10.25	7.35
4.75	3.90	16.60	24.91	9.86	7.13
5.75	3.85	16.67	24.84	9.89	7.17
6.75	3.84	17.16	25.09	10.24	7.45
8.50	3.90	16.67	24.68	10.10	7.35
11.75	3.54	16.26	23.01	9.85	7.22
15.50	3.98	18.93	23.21	10.32	7.42

Table B.3: Rock Eval pyrolysis results for lake CamH2, Cameron Hills, NWT.

Interval Mid-Pt. depth (cm)	S1	S2	S3	TOC	RC%
0.75	6.26	28.39	21.96	12.19	8.32
1.75	7.42	28.42	21.40	12.41	8.47
2.75	7.27	27.75	21.06	12.20	8.34
3.75	7.18	27.87	21.28	12.49	8.61
4.75	6.35	24.55	19.31	11.31	7.86
5.75	5.94	23.45	18.64	10.97	7.69
6.75	5.31	22.48	18.23	10.67	7.54
7.75	5.57	22.69	18.07	10.63	7.46
8.75	5.30	23.57	18.19	10.94	7.71
10.50	5.21	22.93	17.19	10.78	7.66
13.50	4.84	23.84	4.84	10.74	7.89
17.50	6.91	38.77	26.04	15.37	10.39
23.00	6.24	37.36	24.47	14.47	9.73
29.00	7.57	42.62	26.52	15.86	10.47

Table B.4: Rock Eval pyrolysis results for lake CamH5, Cameron Hills, NWT.

Interval Mid-Pt. depth (cm)	S1	S2	S3	TOC	RC%
0.75	24.09	105.43	41.32	27.93	15.34
1.75	23.70	108.29	41.10	28.76	15.97
2.75	24.85	108.34	40.99	28.79	15.90
3.75	24.50	108.04	40.55	28.52	15.70
4.75	24.21	108.32	40.20	28.52	15.71
5.75	25.39	108.60	39.44	28.59	15.69
6.75	24.20	108.95	38.79	28.43	15.63
7.75	23.20	111.21	38.54	28.68	15.79
8.75	24.81	111.26	39.30	28.88	15.81
10.50	24.71	112.70	39.43	29.04	15.86
13.50	22.46	109.50	38.07	28.71	16.03
17.50	21.46	109.38	37.59	28.74	16.18
23.00	22.32	109.68	37.50	28.48	15.83
29.00	21.95	108.20	38.27	28.58	16.04

Appendix C. Isotopic profiles of study lakes from Cameron Hills, NWT.

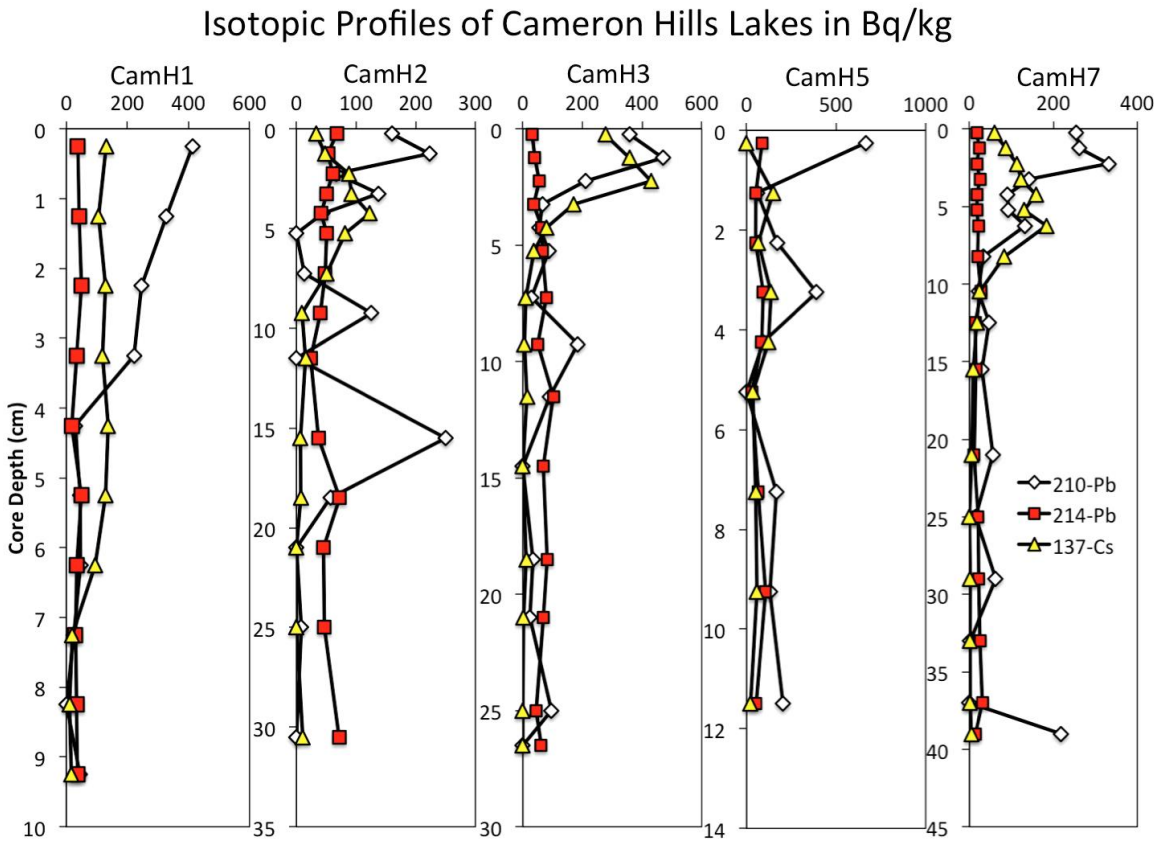

Figure C.1: Down-core profiles of ^{210}Pb, ^{214}Pb, ^{137}Cs for the five lakes in Cameron Hills, NWT.