

Can alluvial landforms attenuate post-wildfire lake sedimentation rates
and external phosphorus inputs?

by

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Author's Declaration

This thesis consists of material all of which I authored or co-authored: see Statement of Contributions included in the thesis. This is a true copy of the thesis, including any required revisions, as accepted by my examiners.

I understand that this thesis may be made electronically available to the public.

Statement of Contributions

This research was conducted at the University of Waterloo by Stephanie Pegler under the supervision of Dr. Mike Stone. Sediment core collection was conducted by Dr. Mike Stone and James Telford, and geochemical analyses of samples were conducted at a commercial laboratory (Actlabs). Stephanie Pegler compiled and interpreted all data, completed statistical analyses, produced all figures and tables, as well as a draft manuscript, with edits made by Dr. Mike Stone and Dr. Roland Hall. Johan Wiklund provided technical assistance in dating the sediment core.

Abstract

Severe large-scale landscape disturbance in forested source water regions by wildfire can increase the transfer of fine sediment and associated phosphorus (P) to aquatic systems. Notably, the subsequent mobilization of sediment-associated P to receiving surface waters can increase primary productivity in lakes and severely degrade water quality. The goal of this research was to examine abiotic controls on the form and mobility of particulate phosphorus (PP) in benthic sediment of an oligotrophic lake in Waterton Lakes National Park (WLNP), Alberta. In 2017, the Kenow wildfire severely burned an area of 35 000 ha which increased the transfer of pyrogenic materials to receiving streams in WLNP. The temporal distribution and concentration profiles of PP forms (non-apatite inorganic P, apatite P, organic P) and P mobility (equilibrium phosphorus concentration, EPC_0) and major elements in a sediment core collected from Lower Waterton Lake (LWL) were evaluated to assess the potential of post wildfire fine sediment inputs to influence P concentrations in the overlying water column, and the potential internal loading legacy effect that wildfires may have on lake P-dynamics. Despite elevated PP levels observed in fire impacted tributaries that flow into LWL, the particulate P fractionation data show that post-fire P loading from pyrogenic materials to the lake was extremely low and only very minor post-wildfire changes in TPP speciation were observed. The presence of a large alluvial fan impeding river flow, coupled with the ingress of fine sediment in gravel bed rivers and adjacent floodplains, most likely attenuated the influx of pyrogenic materials into LWL. The mobility of PP in lake bottom sediment corresponded with historical landscape disturbances (e.g., flooding), which are likely associated with the remobilization of deposited pyrogenic materials in the alluvial fan, demonstrating a fire-flood sequence. Although the data presented in this thesis suggests that wildfire will have a minimal effect on the internal loading of P in LWL, it is increasingly acknowledged that non-stationarity due to changing climate may produce flow conditions which will resuspend and deliver pyrogenic materials to the lake, potentially creating a post-wildfire legacy effect. This thesis provides new knowledge regarding the effects of alluvial landscape forms on the post-wildfire delivery and mobility of PP into critical source water areas.

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Dedication

I dedicate this thesis to my best friend and partner, Peter. You were always there for me. I can't wait for our next adventure.

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List of Abbreviations

Al - aluminium

AP – apatite phosphorus

BD-RP – bicarbonate-dithionite reactive phosphorus

CRS – constant rate of supply

EPC₀ – equilibrium phosphorus concentration

Fe – iron

Mn - manganese

NAIP – non-apatite inorganic phosphorus

OP – organic phosphorus

P – phosphorus

PP – particulate phosphorus

SRP – soluble reactive phosphorus

SWI – sediment water interface

SWP – source water protection

TDP – total dissolved phosphorus

TP – total phosphorus

TPP – total particulate phosphorus

WLNP – Waterton Lakes National Park

WWTP- waste water treatment plant

Chapter 1

Introduction

1.1 Problem Statement

Climate change exacerbated droughts, increases in wildland fuel loads, and heightened human presence in the wildland-urban interface have all contributed to an alarming increase in the frequency and severity of wildfires globally (Hanes et al., 2018; Mell et al., 2010; Jolly et al., 2015). Severe wildfires can have catastrophic economic and infrastructure impacts on communities (Price et al., 2013; Mell et al., 2010) and drastically alter the physical, chemical, and biological composition of soils in forested watersheds (Bodì et al., 2014). Increased erosion rates and sediment-associated contaminants transferred from burned landscapes to receiving waters can degrade water quality (Emelko et al., 2016; Robinne et al., 2016; 2019), harm aquatic ecosystem functions (Silins et al., 2009; Kunze and Stednick, 2006;) and challenge water supplies (Emelko et al., 2011; Emelko and Sham, 2014; Wilkinson et al., 2009). The impacts of wildfire on aquatic systems are largely related to the erosion and transfer of pyrogenic materials and associated contaminants which can be stored in streams or propagated downstream to receiving waters such as lakes and reservoirs (Moody and Martin, 2004; Wilkinson et al., 2009; Burd et al., 2018). There is a critical need to further our understanding of the post-wildfire effects of accelerated nutrient release from pyrogenic materials on receiving water bodies such as lakes and reservoirs (McCollough et al., 2019).

Forested watersheds are responsible for the supply, natural filtration, and storage of water, valued at \$4.1 trillion, generating most of the high-quality water for domestic, agriculture and ecological needs globally (Costanza et al., 1998). In Alberta, more than 85% of drinking water for communities originates in forested watersheds (Emelko et al., 2011). In these important watersheds, the streams, rivers, and downstream lake water quality is intrinsically linked to catchment vegetation, hydrology, land use, geology, and soils (Anderson, 2014); thus the water quality of lakes can differ significantly within and between watersheds as a function of land use. Wildfires threaten global water security through biological, chemical, and physical stressors which impact water quality, quantity, sediment transport, nutrient dynamics, and overall ecosystem health (Baker, 2003; Silins et al., 2014). Notably, lakes in forested watersheds typically have very good

water quality but these sensitive aquatic systems are under increasing threat from sediment-associated impacts following landscape disturbance (e.g., wildfire, flooding; Foley et al., 2005).

Pyrogenic materials (i.e., burned particulate matter containing ash, charcoal, burned soils) and associated contaminants (e.g., nutrients, metals) derived from burned landscapes can be propagated downstream and subsequently deposited in receiving lakes and reservoirs (Moody and Martin, 2001; McCullough et al., 2019). In particular, the delivery of sediment-associated phosphorus (P) to aquatic systems is a major concern as P is a limiting nutrient in freshwater aquatic ecosystems and increased nutrient loading can proliferate the growth of cyanobacteria (Schindler, 1977; Emelko et al., 2016). The external loading of potentially mobile sediment-associated P forms in lake bottom sediment increases the concentration of P in benthic sediments (Nurnberg, 2009). Under the appropriate environmental conditions, bioavailable P (soluble reactive phosphorus; SRP) bound to sediment can be released to the water column via desorption in a process known as internal P loading (Søndergaard et al., 2003). The release of SRP from sediments to the water column can alter the balance of nutrients in the water column, contributing to algal blooms, eutrophication, and long-lasting water quality issues (Emelko et al., 2016; Stone et al., 2014; Watt et al., 2021). Wildfire disturbance in watersheds can substantially increase external nutrient loading in lakes and may result in long-term water quality degradation due to the accumulation of P in sediment (McCullough et al., 2019).

There are several controls in watersheds which are responsible for the transport, delivery, and storage of sediment and pyrogenic materials to receiving waters (Wilkes et al., 2019). The transport of particulate matter from hillslopes to receiving streams and downstream lakes are subject to potential lags and delays along the river continuum. Wildfires often result in reduced vegetation cover, decreased infiltration, and increased overland flow, leading to an increase in the potential for surface runoff and major sedimentation events for several years following the burn (Wondzell and King, 2003). In the Rocky Mountains, net precipitation reaching the land surface increases (Williams et al., 2019) and runoff-generated debris flow and floods are the most common post-wildfire responses on severely burned slopes (Wondzell and King, 2003; Riley et al., 2015). As mobilized sediment and pyrogenic materials are transferred across the landscape and river continuum, there is potential for sorting and attenuation to influence the timing and quantity of material ultimately delivered downstream (Wilkes et al., 2019). Geomorphic landforms in Rocky

Mountain watersheds alter the delivery paths, hydraulic conditions, and pressure gradients inducing the deposition of sediment along the river continuum (Germanoski and Miller, 1995; Wilkes et al., 2019). While sedimentation events (e.g., erosion from hillslope to receiving stream) following wildfires are common (Wondzell and King, 2003) geomorphic landforms may ultimately affect the timing of delivery of these materials to downstream waters.

The potential nutrient shifts (e.g., eutrophication) in lakes and reservoirs following wildfire are of concern to watershed managers and drinking water providers (Emelko and Sham, 2014). All drinking water requires at least some treatment (Schmidt et al., 2019)—deteriorating source water quality often leads to increases in drinking water treatment costs (Ernst et al., 2004; Emelko & Sham, 2014; Price et al., 2017; Pan et al., 2022). After wildfire, solids-associated metals (Abraham et al., 2017), nutrients (Emelko et al., 2011; Silins et al., 2014; Gustine et al., 2021), and other contaminants (Crouch et al. 2006; Mansilha et al., 2017) may be elevated. The release of bioavailable phosphorus from fine benthic sediments has also been observed for many years post-fire (Stone et al., 2014; Emelko et al., 2016)—these effects are further magnified in combination with anthropogenic disturbance impacts (Watt et al., 2021). Rapidly fluctuating water is especially challenging for water providers (Emelko et al., 2011; Kundert et al., 2014; Skwaruk et al., 2022). Accordingly, techno-ecological nature-based solutions (TE-NBS) such as forest management-based source water protection approaches and biological treatment processes are increasingly advocated to mitigate these threats (Emelko and Sham, 2014; Blackburn et al., 2021; 2022). While landscape and/or ecosystem features may serve as TE-NBS for these challenges, they have not been widely investigated in this regard.

Pyrogenic materials contain dissolved-organic carbon, nutrients, and mineral ash, which have been shown to deteriorate water quality and may contribute to internal P loading in lakes (Bodí et al., 2014; Emelko et al., 2016). These effects can be especially challenging for water providers because they often promote the proliferation of algae that can produce toxins and clog treatment systems, leading to service disruptions and even outages (Emelko et al., 2011). While previous studies have focused on changes in post-wildfire stream water geochemistry (Granath et al., 2020), riverbed sediment composition (Son et al., 2015; Watt et al., 2021), and water quality in river systems (Emmerton et al., 2020), research on the effects of post-wildfire P dynamics in lakes is limited. Lakes are downstream recipients of water and material transport from the

watershed that are strongly related to the impacts of landscape disturbance in their watersheds (Williamson et al., 2008). Unlike streams and rivers, lakes have water residence times of months to several years, and therefore the effects of fire may be more persistent and lasting than in other waters (McCollough et al., 2019). These observations highlight the critical need to better understand the composition of pyrogenic materials and the related biogeochemical processes as key drivers of P dynamics in lakes.

1.2 Research Objectives

The primary goal of this work is to examine the vertical distribution of particulate phosphorus forms (NAIP, AP, OP) and P mobility (EPC_0) in benthic sediments of an oligotrophic lake with a history of previous wildfire disturbance in the watershed. The temporal distribution of PP forms and P mobility were determined and related to the geochemical composition and historic fire activity and lake outflow data in Waterton Lakes National Park, AB. The primary goal of this study was to advance knowledge of the effect of PP on lake nutrient cycling to inform the development of climate change adaptation strategies by furthering our understanding of the environmental impact after a large-scale wildfire. The main objectives of this study are to:

- (1) evaluate major element geochemical composition and the vertical distribution of particulate phosphorus forms (NAIP, AP, OP) in a Lower Waterton Lake sediment core,
- (2) characterize the mobility and potential release of P from deposited sediment to the water column in benchtop batch experiments,
- (3) discuss the role of watershed hydrology and the functional attributes of riverine landscapes on the fate of sediment associated phosphorus in Waterton Lakes National Park and,
- (4) discuss implications of the research to the management of lakes in WLNP in response to landscape disturbance.

To my knowledge, this research is the first of its kind to specifically evaluate the effect of wildfire-associated runoff and internal P loading in an oligotrophic lake. The resulting data will enhance process understanding to improve science-based decision making for source water

protection (SWP), reservoir management and water treatment strategies to improve drinking water security in Canada and globally.

1.3 Tasks

The objectives of this thesis were met by conducting geochemical analyses, particulate phosphorus fractionation and bench-top phosphorus sorption experiments on sections of a sediment core collected in Lower Waterton Lake. The sediment core was dated and linked to watershed disturbance history using sedimentation rate and lake outflow data as a proxy of changing hydrologic conditions in Lower Waterton Lake. Objective 1 utilized a sequential extraction scheme developed by Pettersson et al., (1998) to investigate the temporal distribution of PP forms. Objective 2 was achieved by conducting batch experiments described by Froelich (1988) to investigate the equilibrium phosphate concentration (EPC_0) of fine sediment from sections of the sediment core. Objective 3 was accomplished by analyzing the relationship of historical lake outflow data from Waterton Lakes National Park and measured temporal changes in the lake sedimentation rate to identify landscape features that attenuate the transfer of pyrogenic materials in rivers to the LWL. Objective 4 included a critical analysis of the hydrometric data to infer measured changes in lake P loading following landscape disturbances (e.g., wildfire, flooding).

1.4 Thesis Format

The problem statement, and research objectives are provided in Chapter 1. The literature is provided in Chapter 2. The study site description and methods are provided in Chapter 3. The vertical variation of lake sediment P forms and geochemical analysis and temporal variability in sorption dynamics are provided in Chapter 4. Chapter 5 provides a discussion of the results. A synthesis of conclusion and recommendations for future work is provided in Chapter 6.

Chapter 2

Literature Review

2.1 Watersheds and climate change

Forested watersheds are critical drinking source water regions of the globe, however natural and anthropogenic large-scale land disturbances can significantly alter water quantity and quality of aquatic systems in these regions (Baker, 2003; Emelko et al., 2011). The current state and deterioration of lake water quality in Europe and North America is primarily related to land use change primarily due to runoff from agricultural and urban land use (Carpenter et al., 1998). In lakes where agriculture, urbanization, and harvesting dominate catchment land use, the deposition of nutrient enriched sediment in aquatic systems can result in cultural eutrophication and degrade water quality (Carpenter et al., 1998; Orihel et al., 2015). Additionally, increasing climate change-exacerbated landscape disturbances, such as wildfires and flooding, can significantly alter vegetation and geophysical processes such as erosion, runoff, and stream flow (Moody and Martin, 2001; Williams et al., 2013). In North America, severe large-scale landscape disturbance by wildfires has increased in extent and severity (Flannigan et al., 2013) which threaten high quality, source water supplies of freshwater for human consumption (Emelko et al., 2011).

Wildfires are a natural phenomenon that have ecological (Smith et al., 2011; Robinne et al., 2020), cultural (Eisenberg et al., 2019), and geochemical (Moody and Martin, 2001; Murphy et al., 2006; Emelko et al., 2016) implications for landscape form and function. However, in recent years, wildfires have increased in extent and severity due to several factors such as changing climate, current forest management practices, prolonged droughts, increased fuel loads, snowpack changes, and increased lightning strikes (Wang et al., 2017; Eisenberg et al., 2019). The increase in wildfire frequency and severity poses a threat to forested watersheds across North America (Flannigan et al., 2013; Emelko et al., 2015). The term “mega-fire” has recently been coined in the literature and represents large-scale wildfires which make up less than 1% of wildfires worldwide, but account for ~95% of the global annual burn area (Williams, 2013). There is increase in global

concern for the degradation of water quality and aquatic ecosystem health in forested watersheds following mega-fires (Baker, 2003; Wondzell and King, 2003; Kelly et al., 2006; Emelko et al., 2011).

2.2 Sediment supply and transport in watersheds

Multiple dynamic processes (e.g., biologic, chemical, hydrologic, geomorphologic, climatic) occur simultaneously in watersheds contributing to the quality and quantity of water supply. Natural and anthropogenic factors control the delivery, transport, and storage of particulate matter in river networks, influencing the hydrological and geomorphological behavior of catchments (Owens et al., 2005; Wilkes et al., 2018). Excess fine sediment in aquatic systems, defined as organic and inorganic particles $< 63 \mu\text{m}$ in diameter, is a principal reason for reduced water quality in forested catchments, and is a main concern for water quality managers (Armstrong et al., 1979; DePinto et al., 1981; Emelko et al., 2011). The supply of fine sediment to receiving waters is governed by sediment transfer from terrestrial landscapes, delivery pathways, hydraulic and biologic processes, and environmental filtering (Figure 2.1) (Wilkes et al., 2018).

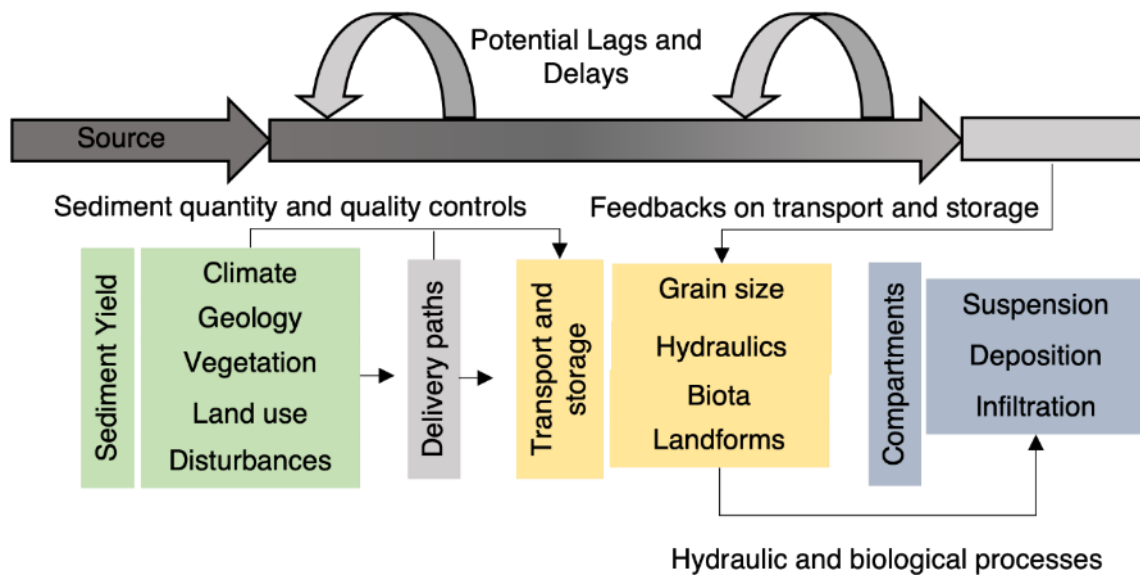


Figure 2.1 – Controls on the transport, delivery, and storage of fine sediment in aquatic systems. (After Wilkes et al., 2018.)

Sediment can be mobilized from the terrestrial landscape to the aquatic system from both natural and anthropogenic sources (e.g., arable land, road verges), and is influenced by geology, soil type, and climate (Collins & Zhang, 2016). Factors specific to each watershed can modify the amount of sediment entering waters, such as topography, land use, vegetation, and disturbances (Wilkes et al., 2019). The downstream yield of sediment depends on the delivery pathways that connect downstream waters to stream networks, as there are potential lags and delays in sediment transport in watersheds that ultimately influence the quantity of sediment delivered to receiving water bodies (e.g., lakes, reservoirs) (Wilkes et al., 2019). Generally, in forested watersheds, sediment delivery and yield are positively correlated with the magnitude of hydrologic events, and river flow intensity (Wohl, 2015). Post-wildfire erosion processes include debris flows, sheet flows, and sediment-laden floods (Wondzell and King, 2003). Once sediment is mobilized in the catchment, several controls determine the transport and deposition of fine sediment. Hydraulic controls such as water velocity and the degree of turbulence influence sediment settling and ingress along the river continuum (Van Leussen, 1988). Landscape features can influence the storage and delivery of fine sediments, as sediment can be temporarily attenuated by geomorphic forms which alter the timing and amount of sediment delivery to downstream waters (Wilkes et al., 2019). Geomorphic forms causing potential lags and attenuation include storage of sediment in gravel-bed rivers, on floodplains, deltas, man-made structures (e.g., road crossings, dams), braided rivers, wetlands, midchannel bars, alluvial fans, and riparian areas (Wohl, 2015). In watersheds located in the Rocky Mountains, most attenuation of fine sediment is in gravel bed rivers (Boano et al., 2011; Wilkes et al., 2019). Post-wildfire flows may also enter river channels, and result in channel damming or constriction and local aggradation (Meyer and Wells, 1997; Riley and Meyer, 2015). Where alluvial fans are present in watersheds, sediment attenuation is common following a wildfire as steep tributaries enter larger valleys (Riley and Meyer, 2015). Once fine sediments are deposited, they can remain on the bed surface, ingressed into the bed, or resuspended (Wildhaber et al., 2014; Wharton et al., 2017).

Once sediment is deposited in the landscape, it remains stored until it is remobilized and transported downstream to receiving waters. Sediment and pyrogenic materials can remain stored by landforms for decades or millennia (Moody and Martin, 2001; Fryirs et al., 2017). Characteristics of the watershed and geomorphic features determine the lag time of sediment

delivery (Wilkes et al., 2019). Typically, sediment is stored in river channel beds < 1 year, because the active layer of channel beds can exchange annually (Gellis et al., 2017). Fine sediment is mobilized at higher flow rates, as their small size requires a higher velocity threshold to reach a critical shear stress for erosion (Stone et al., 1993). While there may be lags and delays in sediment delivery, the frequency of severe hydrologic events is increasing with changing climate (Emelko et al., 2011). Potential increases in storm frequency and magnitude will result in greater hillslope erosion, and sediment supply to receiving waters. Large floods can scour vegetation from previously stabilized riparian areas, resulting in widening of flood plain and higher deposition rates, creating new sources of sediment that can remobilize with future high discharge events (Warrick et al., 2013). Soil erosion and alterations to sediment transport and fate is the main driver of water quality issues, impacting the future of water supply in forested watersheds (Emelko et al., 2011).

2.2 Landscape disturbance by wildfire

Wildfire is a major source of disturbance in forested watersheds and can greatly influence the hydrology of watersheds. Vegetation loss after a wildfire has been shown to increase rates of runoff and sediment erosion from terrestrial surfaces, increasing the transfer of sediment-associated contaminants (Crouch et al., 2006; Kelly et al., 2006) and nutrients (Kunze and Stednick, 2006; Emelko et al., 2011; Silins et al., 2014) to receiving streams (Wondzell and King, 2003; Williams et al., 2013). Burned soils are typically hydrophobic which reduces infiltration capacity and increases overland flow (DeBano, 2000). Increased overland flow coupled with reduced vegetation results in increased erosion rates and increased sediment production from burned landscapes (Ewing, 1996; Benavides-Solorio and MacDonald, 2001; Silins et al., 2009). Moreover, burned surface litter and topsoil can often have higher nutrient concentrations after a wildfire (Bodì et al., 2014) and cause post-wildfire catchment yields of sediment and nutrients to increase markedly (Moody, 2001; Emelko et al., 2016; Wilkinson et al., 2009). The effects of wildfire on receiving waters draining burned watersheds can persist for decades (McCollough et al., 2019).

Wildfire can alter the physical, chemical, and biological composition of litter on the forest floor (vegetation, biomass, soil organic matter) and can convert organic matter to pyrogenic materials, such as ash and charcoal (Bodí et al., 2014). Wildfire ash contains a mixture of partially burned organic matter, inorganic material (referred to as mineral ash), and particulate matter, that are chemically modified or transformed by fire (Bodi et al., 2014). The amount and kind of pyrogenic materials post fire are dependent on the maximum fire temperature (Boerner, 1982). As fire severity increases, combustion temperature rises, resulting in more organic matter converted to pyrogenic materials (Bodi et al., 2014; Son et al. 2015). Intense mineralization of the soil after fire can be enriched with nutrients such as phosphorus, that accumulate in ash and surface runoff and transferred from hillslopes to receiving streams (Bodí et al., 2014). Carbon is the dominant element in ash, however other elements are also present in varying concentrations depending on maximum fire temperature (Bodí et al., 2014). Elements with high volatilization temperatures include phosphorus ($> 774^{\circ}\text{C}$), magnesium ($> 1107^{\circ}\text{C}$), calcium ($> 1240^{\circ}\text{C}$), and manganese ($> 1962^{\circ}\text{C}$; Weast, 1980; Wright and Bailey, 1982). These elements have been found to increase in the soil after a wildfire as their high volatilization temperatures are seldom reached. Major elements with low volatilization temperatures include mercury (Hg; $> 300^{\circ}\text{C}$), nitrogen ($> 500^{\circ}\text{C}$), sulfur (S; $> 575^{\circ}\text{C}$; Hungerford et al., 1991; Bodí et al., 2014) and may decrease in proportion following wildfire. Ash can have profound geochemical effects on terrestrial and aquatic ecosystems, increasing concentration of nutrients into burned watersheds (Bodí et al., 2014). Forested watersheds are particularly vulnerable to contaminant fluxes associated with pyrogenic materials, and the composition, transport, and mobility of these materials to downstream lakes is poorly understood (Emelko et al., 2011; Stone et al., 2011).

2.3 The role of phosphorus in lake productivity

Forested watersheds are a critical source of drinking water globally, however following wildfire increased erosion rates can greatly increase the amount of sediment, nutrients, and pyrogenic materials to lakes (Moody and Martin, 2009; McCollough et al., 2019; Robinne et al., 2020) decreasing the quality of downstream water supplies. Phosphorus (P) has a high volatilization temperature and, is therefore, of particular concern due to the increased P concentration in surface waters following a wildfire (Bodí et al., 2014). Phosphorus is a limiting nutrient in freshwater

aquatic systems and when total phosphorus concentrations exceed a threshold of $30 \mu\text{g P L}^{-1}$, primary producers proliferate in aquatic systems (Schindler, 1977; Barlow-Busch et al., 2006). Due to the increase in productivity of primary producers, the excess plant and algal material can reduce light penetration, increase hypoxia, increase turbidity, and cause eutrophication in lakes (Carpenter, 1998; Corell, 1998; Chorus and Bartram 1999). Eutrophication increases the risk of toxin-producing cyanobacteria proliferation in the water column, with implication for ecosystem function (Hauer and Spencer, 1998), human health (brain and liver toxins; Chorus and Bartram, 1999), and water treatability (taste and odor problems; Emelko et al., 2011). In oligotrophic lakes, the input of post-fire nutrient enriched particulate matter has the potential to accelerate primary production

Historically, anthropogenic disturbances like urbanisation and agriculture, have led to the P enrichment in aquatic systems which have accelerated the rate of eutrophication through point and nonpoint discharge sources. Point discharge sources are commonly cited as discharge outlets of wastewater effluents and surface runoff that are highly concentrated in soluble P, such as those from wastewater treatment plants (WWTP; Edwards and Whithers, 2008). Diffuse sources of P are associated primarily with agricultural runoff which can transfer suspended solids and dissolved P (Whither and Jarvie, 2008). Natural sources of P in the environment include the common weathering of materials, atmospheric deposition, vegetation litter, and erosion (Holtan et al., 1988; Walling et al., 2008; Whithers and Jarvie, 2008). However, natural large-scale landscape disturbances (i.e., wildfire) can deliver substantial amounts of sediment (Silins et al., 2009) and P (Emelko et al., 2016) to receiving waters. This increase can change not only the biogeochemical processing of P within the river continuum but can also have water quality implications for aquatic systems downstream (Arp and Baker, 2007; Whithers and Jarvie, 2008).

2.4 Particulate phosphorus in watersheds

The total phosphorus concentration in aquatic systems is operationally defined as the sum of total dissolved phosphorus (TDP) and total particulate phosphorus (TPP) forms (Jarvie et al., 2002). Dissolved P forms are the most bioavailable of any species of P and dissolved P, commonly termed soluble reactive phosphorus (SRP), is predominantly found as orthophosphates (H_2PO_4^- , HPO_4^{2-} , or PO_4^{3-}) (Boström et al., 1988; Whithers and Jarvie, 2008). Due to the bioavailability of

SRP, benthic communities and primary producers can readily metabolize this nutrient, contributing to eutrophication and water quality concerns in lakes (Corell, 1998). The relative role of abiotic and biotic controls on sediment-associated P dynamics are complex (Figure 2.2). While SRP is generally the most bioavailable form of P, some forms of particulate phosphorus (PP) such as non-apatite inorganic P (NAIP) can sorb and release SRP between the water column and benthic sediment (Froelich, 1988).

Particulate phosphorus (PP) is associated with fine sediment (i.e., $< 63 \mu\text{m}$), and is the primary vector for P transport in aquatic systems (Stone and English, 1993; Davies-Colley and Smith, 2001; Dodds, 2003). Fine sediment has a large surface area per unit mass and metal oxyhydroxide coatings preferentially bind P (Horowitz and Elrick, 1987; Stone and Murdoch, 1989; Wood and Armitage, 1997). The increase in surface area with decreasing particle size allows for greater P sorption to/from the surfaces of sediment. Fine sediment is the most abundant fraction of eroded material present in many river systems and this sediment fraction is generally considered to be the most geochemically active and a key driver of nutrient transport in aquatic systems (Peart and Walling, 1982; Stone and Murdoch, 1989; Allan, 1986). This sediment size fraction has a low settling velocity and therefore can remain suspended in the water column, propagating sediment and associated phosphorus across the river continuum to receiving waters such as lakes and reservoirs (Emelko et al., 2016; Armstrong et al., 1979; DePinto et al., 1981; Owens et al., 2005).

Particulate phosphorus has various forms and can pose different ecological significance depending on their bioavailability. In aquatic systems PP can be fractionated into three forms: (1) non-apatite inorganic phosphorus (NAIP), (2) apatite phosphorus (AP), and (3) particulate organic phosphorus (OP). NAIP is associated with aluminium, iron-hydroxides, and manganese on particle surfaces, which can be readily desorbed back into the water column as SRP (De Pinto et al., 1981; Pettersson et al., 1981). AP forms are susceptible to low pH and are bound primarily to Mg and Ca carbonates. AP is considered the most stable and a permanent sink of P in sediments (Kaiserli et al., 2002). OP has many forms, including organic molecules such as nucleic acids, phospholipids, phosphonates, and nucleotides (Froelich, 1988). OP has low potential bioavailability; however, some fractions can be mineralized with hydrolysis (Sharpley et al., 1995).

The form and availability of P in freshwater systems is influenced by a range of physical, biological, and geochemical controls (Boström et al., 1988). Iron (Fe), manganese (Mn), and aluminium (Al) ion coatings are often correlated with PP concentrations (House and Denison, 2002; Evans et al., 2004). As phosphate ions sorb to metal complexes (Klotz, 1991), the availability of metal oxyhydroxides in the environment will result in the binding of P to metal coatings on sediment (Golterman, 2004). Therefore, the availability of P and P fluxes may be influenced by the transformation of these metals (Klotz, 1991). Further, environmental conditions such as pH and redox can influence PP form and sediment geochemistry (Golterman, 1995). In oxic environments, SRP can precipitate with Fe and Ca to form stable P minerals (Jensen et al., 1992). Additionally, OP can undergo hydrolysis, increasing bioavailability (Golterman, 1995). The presence of biofilms in oxic environments can influence chemical gradients that may alter the availability of metal, calcite, and organic carbon (Boano et al., 2014). Consequently, abiotic, and biotic controls are coupled, resulting in an interplay on sediment geochemistry and the dynamics of PP forms in aquatic environments (Withers and Jarvie, 2008).

The input of sediment-associated phosphorus to aquatic systems has implications for water quality and drinking water security (Silins et al., 2009; Blandon et al., 2014; Emelko et al., 2016). The external loading of sediment-associated P to lakes can increase the pool of potentially available bioavailable P accumulated in lake bottom sediment. Under the proper conditions, the release of soluble reactive P from deposited fine-grained benthic sediments to the water column can increase (Stone and English, 1993; Søndergaard et al., 2003). Internal P loading has been identified as the main driver for algae growth in aquatic systems (Paytan et al., 2017; Stoddard et al., 2016). Internal P loading can create a legacy effect in lakes, where benthic sediments can release P for decades (Søndergaard et al., 2001; Sharpley et al., 2013). Notably, it follows that if watershed managers can limit total phosphorus concentrations in the water column in source water, water quality issues associated with cyanobacteria blooms could be mitigated (Emelko et al., 2011).

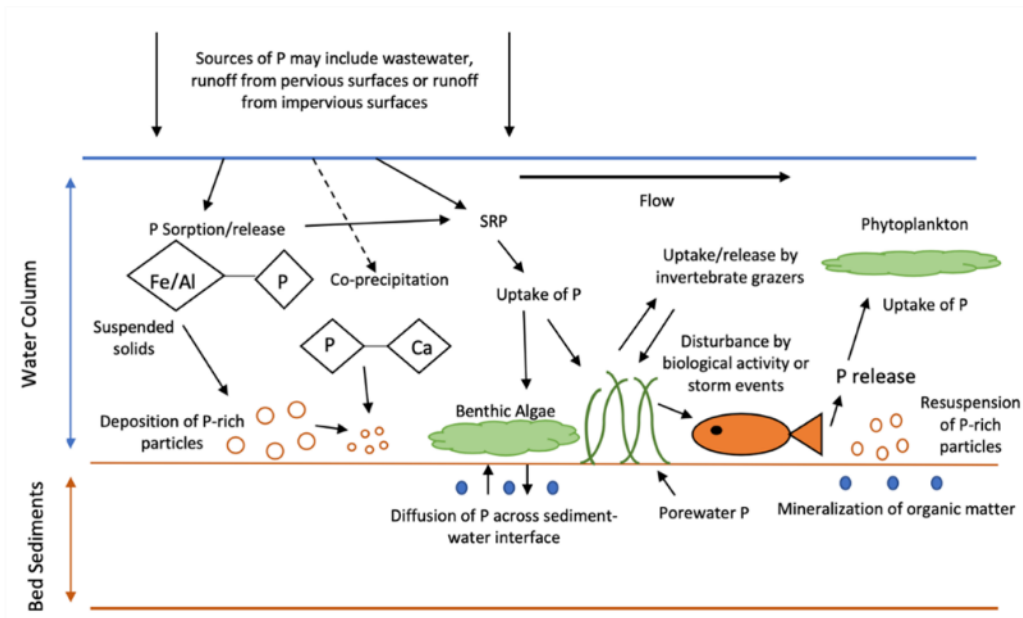


Figure 2.2 – Conceptual diagram of water column processes influencing P concentrations in lakes. (Source: Withers and Jarvie, 2008.)

2.5 Implications of landscape disturbance on internal loading of phosphorus in lakes

The bioavailability of particulate P forms is dependent on their ability to desorb from surfaces of particle matter. The “zero equilibrium phosphorus concentration” (EPC_0) is defined as the dissolved phosphorus concentration of a sediment sample at the point where it neither adsorbs nor desorbs P (Froelich et al., 1988). The EPC_0 can be used as a proxy to determine whether sediments are sources or sinks of P and if the sediment-associated P is in equilibrium with the water column. When the P concentration in the water column is higher than the EPC_0 , sediment will act as a sink to P. Conversely, when the P concentration in the water column is less than the sediment EPC_0 , sediments will act as a source of P to the water column. Previous studies have used EPC_0 as an indicator of nutrient loading with respect to agriculture, non-point pollution, wastewater treatments plants (WWTPs), and timber harvesting (logging) in surface water systems such as rivers and lakes (Søndergaard et al., 2001; Krogrus and Ekholm, 2002; James et al., 2004; Macrae et al., 2005). Studies that have been conducted in lake systems using pore water peepers, have described the changes in dissolved P above, at, and below, the sediment-water interface

(James et al., 1995; Murphy et al., 2001; Matisoff et al., 2016). These studies show that in lakes where benthic sediments are anoxic there is a release of dissolved P from the pore water into the overlying water column (Mortimer, 1941; Nürnberg, 1988). The process by which P is adsorbed or released in an aquatic system is dynamic and is a function of biotic and abiotic processes that influence the ability of sediment to adsorb P from the water column to sediment, until a threshold (EPC_0) condition is reached, where it will desorb back into the water column (Withers and Jarvie, 2008). In contrast few studies have been conducted to evaluate the vertical release potential in lake bottom sediments using EPC_0 in sediment cores. Even fewer studies have been conducted to evaluate P mobility in lake sediment with a history of severe wildfire activity in adjacent landscapes.

Previous studies of phosphorus dynamics in benthic sediments have shown that phosphorus can be released from sediment depths as low as 20 cm (Søndergaard et al., 2003) but the top 5cm immediately below the sediment water interface is generally the most geochemically active (Böstrom et al., 1988). In vertical variations of lake sediments, generally the concentration of Fe is much higher at the sediment-water interface during anoxic conditions as redox conditions control Fe in labile P cycling at the sediment-water interface (Yuan et al., 2020). An increase in pH can precipitate Al as $Al(OH)_3$, as alkalinity decreases the solubility of $Al(OH)_3$ and can result in the uptake of P from the water column to bottom sediment (Kopáček et al., 2000). During anoxia, unlike Fe, Al immobilizes P. This is because the precipitation of Al and P is pH-dependent, not redox-dependent (Kopáček et al., 2000; Wilson et al., 2008). Compared to Al and P, Fe and Mn are redox sensitive, and reflect lake oxygen conditions (Davison, 1993). Mn concentrations in sediments are controlled by physical and geochemical processes in a cyclic pattern, where Mn is released into the water column by reductive dissolution and when re-oxidized subsequently redeposited (Balistrieri et al., 1992; Davison, 1993).

2.6 Phosphorus mobility in sediments

The adsorption/desorption of P to/from sediment is a result of a two-step process where there is a primary rapid surface adsorption followed by a slow ‘solid-state diffusion’ (Froelich, 1988). Phosphorus isotherm experiments and sorption models describe the ability of sediment to

buffer phosphorus (e.g., Langmuir, Freundlich, Tempkin). EPC_0 allows the determination of the dissolved phosphorus concentration where neither adsorption nor desorption occurs (Froelich, 1988). EPC_0 batch experiments conducted in the laboratory typically use the mass of sediment mixed with a series of SRP concentrations until equilibrium between sediment and solution can be assumed (Golterman, 2004). Data are then plotted with initial P concentration on the x-axis and P adsorption/desorption on the y-axis, calculated by equation 2.1.

$$P_{ads} = [(P_{initial} - P_{final}) * 0.025L] * wt_{sed}^{-1} \quad (2.1)$$

The adsorption coefficient is represented by the slope of the buffer diagram and EPC_0 , where steep slopes show greater ability to buffer P (Froelich, 1988; See Figure 2.3). P sorption studies can provide a proxy to the environmental state, as temperature, pH, ionic strength, and ratio of sediment to solution all have the potential to influence sorption characteristics (Barrow, 1980; Klotz, 1988; Lucci et al., 2010). Understanding lake biogeochemical processes coupled with sorption dynamics is necessary to develop an understanding of P dynamics and internal loading.

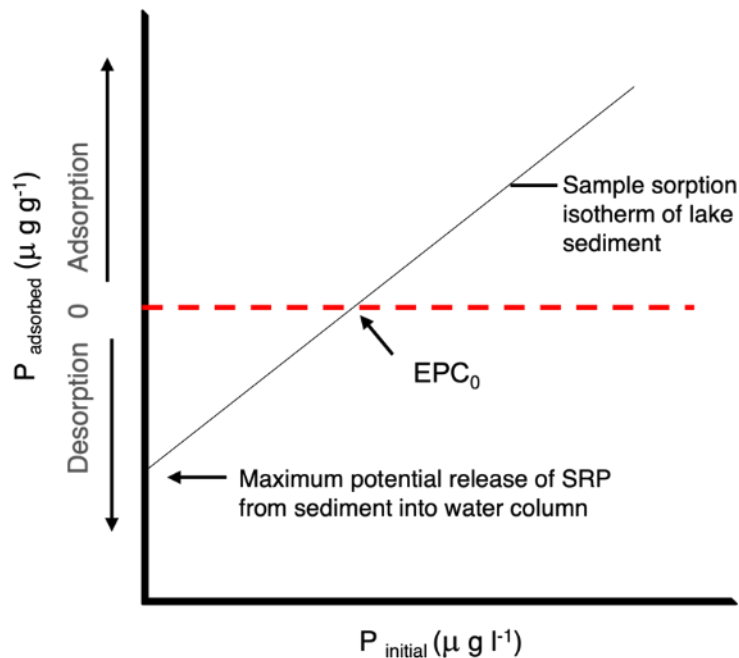


Figure 2.3 – Sorption profile of benthic sediment, where $x = 0$ (dotted line) denotes the equilibrium between benthic sediments and overlying water column (adapted from Froelich, 1988).

EPC₀ concentrations reported in the literature can vary greatly in different aquatic environments and within the same study (Table 2.1). Studies which determine the EPC₀ for lakes and reservoirs affected by landscape disturbance are generally focused on anthropogenic landscape disturbances (e.g., agriculture, WWTPs, urbanization), while much fewer studies focus on natural landscape disturbances (e.g., wildfire). The varying degree of EPC₀ is displayed in the Ekbatan Dam delta, Iran, (Shoja et al., 2017) where EPC₀ concentrations ranged from <1 to 470 µg L⁻¹. Due to the complexity of lake P cycling, there can be a wide range of results across studies and even within the same study. For example, agriculture disturbances in lake catchments show varying EPC₀ concentrations from <1 µg L⁻¹ (Krogerus and Ekholm, 2003; Belmont et al., 2009; Yin et al., 2016; James, 2017; Khanal et al., 2021) to concentrations >470 µg L⁻¹ (Shoja et al., 2017). Lake trophic state does not reflect EPC₀ concentrations, as other environmental factors such as pH, alkalinity, and SWI (sediment water interface) pumping mechanisms are dominant (Table 2.1). For example, although Lake Vörtsjärv and Lake Karhijärvi are eutrophic and commonly have blue-green algae blooms, their maximum EPC₀ is < 2 µg L⁻¹. However, bottom sediments in these lakes are rich in CaCO₃, pointing to the importance of environmental factors controlling algal growth. There is no explicit relationship between trophic level and the P mobility of sediments (Boström et al., 1982). To date, few studies have been conducted to evaluate variability of EPC₀ in oligotrophic lakes or on the variability of EPC₀ in a lake impacted by wildfire. Most studies analyzing the wildfire impacts on sediment are focused on riverbed sediment (not lake or reservoir sediment) and results of these studies vary considerably. . For example, maximum EPC₀ in the wildfire impacted Crowsnest River was 213.5 µg P L⁻¹ (Emelko et al., 2016), while the EPC₀ in the wildfire-impacted Cache la Poudre River ranged from 300 to 1200 µg P L⁻¹ (Son et al., 2015). Variability in measured EPC₀ is a product of varying effects of land use, sediment sources, sediment geochemistry, and biogeochemical processes that regulate P sorption dynamics at the sediment water interface. (Stone and English, 1993; Stutter and Lumsden, 2008).

Citation	Location	Impacts	Lake	Trophic status	Min EPC ₀	Max EPC ₀
This study	Alberta, Canada	Wildfire	Lower Waterton Lake	Oligotrophic	2	23
Spears et al., 2007	Kinross, Scotland	Nutrient loading	Loch Leven	Eutrophic	< 1	12
Cyr et al., 2009	Algonquin Park, Ontario	Tourism, fishing	Lake Opeongo	Oligo-mesotrophic	23	114
Yin et al., 2016	China	WWTP, agriculture	Lake Taihu	Hypereutrophic	72	244
Belmount et al., 2009	Florida, USA	Agriculture	Kissimmee Upper Chain of Lakes	Eutrophic	<1	110
Krogerus and Ekholm, 2003	Southern Estonia	Agriculture	Lake Võrtsjärv	Eutrophic	0	<2
Krogerus and Ekholm, 2003	SW Finland	Agriculture	Lake Karhijärvi	Eutrophic	0	<2
James, 2017	Minnesota, USA	Agriculture, tourism	Lake of the Woods	Mesotrophic	<1	130
Khanal et al., 2021	Combodia	Agriculture, paddy	Tonle Sap Lake	Meso-eutrophic	3	32
Shoja et al., 2017	Iran	Erosion, grasslands, agriculture	Ekbatan Dam delta	Mesotrophic	<1	470
Łukawska-Matuszewska and Xie, 2013	Morsa, Norway	Agriculture	Lake Sæbyvannet	Eutrophic	1.8	22.3
Zhu and Yang, 2018	Hunan, China	WWTP	Dongting Lake	Meso-eutrophic	5.8	345.9

Table 2.1- EPC₀ ($\mu\text{g L}^{-1}$) concentrations for different lakes across a variety of land use and levels of trophic status.

2.7 Utility of sediment cores to evaluate environmental change

A wide range of physical, and biogeochemical factors affect sediment associated P sorption dynamics in lake ecosystems and improved knowledge of lake response to landscape disturbance is critical for improved lake management. Reconstructing historical changes to lakes can be difficult to evaluate due to the lack of direct-long term monitoring, and the absence of knowledge regarding reference conditions. However, these obstacles can be overcome with the application of methods used in paleolimnological studies (Smol, 2008). The examination of lake sediment cores provides a temporal perspective on the temporal changes in the quantity and duration of contaminant deposition, as well as an archive of past environment history and atmospheric inputs (Smol, 2008). Sediment cores provide a method to obtain long-term information regarding biological, hydrological, and sedimentological processes using information stored in the sediment which settles at the bottom of lakes and reservoirs. Once sediment cores are adequately dated, ecosystem response to past disturbances can be evaluated. This is beneficial to lake managers, as sediment cores provide a utility which *in situ* experiments cannot provide. For example, through the collection of a single core, pre-impact, reference conditions can be obtained to compare lake conditions from different time eras, a benefit towards understanding a lakes “natural” state. Lake sediment and nutrient budgets can provide useful and novel frameworks for determining the effect of wildfire-derived sediment on lake nutrient dynamics. Paleolimnological methods allow the comparison of past and present lake histories, which is beneficial for understanding environmental states and developing proper management strategies.

The study of paleolimnology and paleoecology uses sediment cores to infer climate history and vegetation at Holocene-scale (Smol, 2008). These millennial-scale records of wildfire history patterns can provide a baseline for ecosystem management and infer the prediction of future variations in wildfires (Gavin et al., 2007). Lake sediment records from southwestern British Columbia at high elevation and forest-meadow parkland, show large-scale fire intervals over 400 years apart (Lartzman et al., 2002). While paleoecology studies have long temporal histories, they are generally focused on pollen and charcoal proxies (Brown et al., 2017) and much less is known about the form or mobility of particulate P.

Most post-wildfire studies of surface water quality have focused on the measurement of major nutrients (N and P), exchangeable metal cations (Al, Ca, Mn) and organic carbon alterations. Other studies have used tracers to examine the the input of post-fire particulate matter in lake sediment (Owens et al., 2005). Blake et al., (2006) demonstrated the potential of mineral magnetic properties as sediment tracers in wildfire impacted basins. Another approach to characterize wildfire activity in lake sediments is to examine the vertical variation in sediment geochemical composition using sediment cores. Analysis of sediment cores can be used to test hypotheses about factors that control lake response to fire (McCollough et al., 2019). Further research is required to understand the potential of post-fire sediment indicators in sediment cores.

2.8 Novelty of Research

Understanding the dynamics of wildfire-derived contaminants and their relationship to internal P loading is critical to mitigate water quality concerns and potentially toxic cyanobacteria blooms. More research is required to understand the vertical distribution patterns of major elements in lake sediment with wildfire disturbance. Previous studies of the effects of wildfire have focused on landscape vegetation recovery, erosion, soil properties (i.e., water repellency) and river P cycling (Alexis et al., 2007; Bodì et al., 2014). Results show that substantial increases in total phosphorus and total nitrogen were detected in rivers after a wildfire (Blake et al., 2010; Son et al., 2015; Emelko et al., 2016). In landscapes where particulate phosphorus (PP) is transferred from hillslopes to aquatic systems, the flux of potentially bioavailable PP from burned sediment was two orders of magnitude greater than dissolved P yields (Blake et al., 2010; Son et al., 2015). Although post-fire changes of TP concentration in lakes and reservoirs have not been widely reported, sediment deposition post-wildfire is correlated to wildfire severity (Moody and Martin, 2004). Post-wildfire surface water quality in reservoirs can decline due to increased suspended solids, SRP, and nitrate concentrations (Spencer and Hauer, 1991; Basso et al., 2021). Currently, no post-wildfire research has been conducted to evaluate the particulate form and mobility of P in benthic sediment.

Understanding the dynamics of PP in the water column in relation to influx of pyrogenic materials and associated P is an important knowledge gap required to enable modelling post-fire

P dynamics in lakes and reservoirs. While there is an abundant literature on lake P-dynamics, with focus on agriculture, non-point pollution, and urban impacts, consideration of the impacts of wildfire-derived pyrogenic matter and associated nutrients remains limited (Smith et al., 2011;; Robinne et al., 2020). Wildfire is increasing in extent and severity in Western North America (Wang et al., 2017) and forested watersheds under severe threat from wildfire are critical source water regions for water supply (Emelko et al., 2011). To date, little research has been conducted to determine the post-wildfire sediment-associated P dynamics in lake systems. Lake systems on the Eastern slope of the Rocky Mountains have historically been oligotrophic and are sources of high-quality fresh water (Emelko et al., 2016). Sediment geochemistry, geology, and land use characteristics all contribute to biogeochemical cycling of P in lakes (Dean, 1999; Koski-Vähälä and Harikainen, 2001; Jarvie et al., 2005; Lucci et al., 2010; Bhadha et al., 2012). Increased knowledge of post-wildfire P dynamics in lakes and reservoirs is critical to understand the short and longer term impacts of climate change on water quality.

The storage of P in lake systems and interactions in the water column have been intensively studied in systems where the catchment is impacted by anthropogenic stressors such as agriculture, urbanization, and timber harvesting. While much of the literature on sediment-associated P focuses on point and nonpoint P inputs into the catchment and water column dynamics (e.g., McDowell et al., 2002; Ellison and Brett, 2006; Ballantine et al., 2009; Emelko et al., 2016; Watt et al., 2021), much less research has focussed on the post-wildfire P dynamics and specifically the role of pyrogenic sediment as a factor that contributes to internal loading of P to the water column.

Chapter 3

Study Area and Methods

3.1 Site Description

Lower Waterton Lake (N 49° 05' 24.2", W 113° 51' 14.0") is in Waterton Lakes National Park, Alberta (WLNP; Figure 3.1). The Waterton Lakes are composed of three water bodies: Upper Waterton Lake, Middle Waterton Lake, and Lower Waterton Lake. Large alluvial fans located at the mouth of the Sofa and Blakiston Creeks have separated Middle Waterton Lake from Lower Waterton Lake by a narrow channel called the Dardanelles, from the presence of the Blakiston Alluvial Fan (Figure 3.1). Lower Waterton Lake is the smallest and most northern of the three lakes. Lower Waterton Lake is oligotrophic (Canadian Council of Ministers of the Environment, 2004). Maximum depth of the Lower Waterton Lake is 7.3 m.

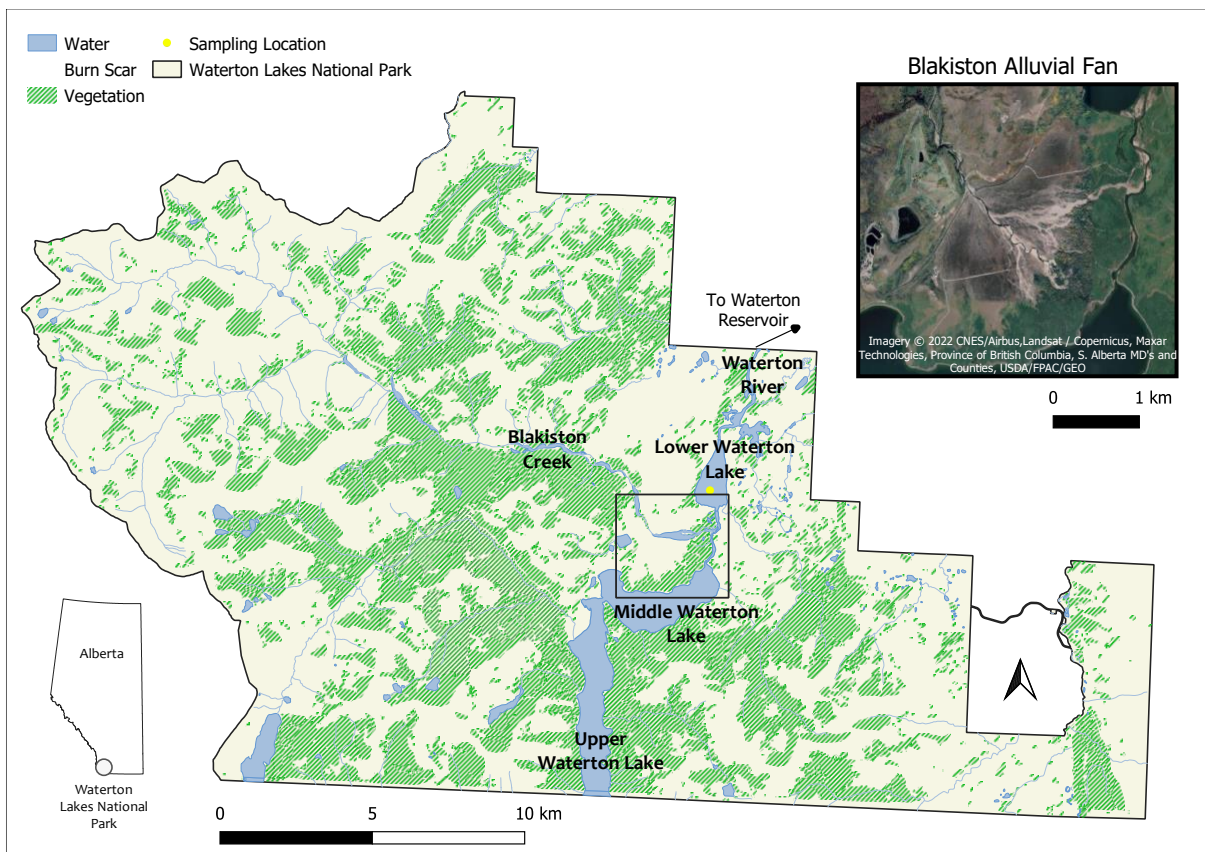


Figure 3.1 – Map of Waterton Lakes National Park. Sediment coring location denoted by yellow point.

Waterton Lakes National Park (WLNP) has a large elevation gradient (600m to 2500m) and a diverse semi-arid ecosystem that includes coniferous and mixed wood forests, open grasslands, and rich riparian areas (Parks Canada, 2018a). Dominant forest species include lodgepole pine (*Pinus contorta* var. *latifolia*), subalpine fir (*Abies lasiocarpa*), and Engelmann spruce (*Picea engelmanni*). Proterozoic sedimentary materials, forced over the younger Mesozoic rocks, make up the foothills of WLNP (Coen and Holland, 1976). Surficial geology of the watershed includes glacial deposits, colluvium, and till (Parks Canada, 2018b). Gravel bed rivers are predominant in the study area due to high slope and abundant glacial-fluvial deposits. Currently there are no glaciers in WLNP.

Southwest Alberta has a long history of fire activity. Prior to the 20th century, wildfires were common in the WLNP area where small fires occurred every 5-10 years, and large fires every 100-150 years (Parks Canada, 2021). Due to fire suppression efforts, wildfire activity has been less frequent since the 1935 Boundary Creek fire (903 ha; Parks Canada, 2019). Prescribed burns (a controlled fire set intentionally as a fuel management tool) were first used in the park in 1989. During the last 25 years, there have been several large (>1000 ha) wildfires and prescribed burns due to increased fuel loading from suppression activities by Parks Canada. In 1993, the Sofa Mountain wildfire burned 1521 ha on the East side of WLNP. In 2006 and 2015 large, prescribed burns (~1200-1600 ha) were conducted at the entrance to and in the upper Red Rock areas of the park, burning the foothills and prairie regions of the Blakiston creek watershed. These prescribed fires utilized the natural fire guards (fuel breaks) such as the Waterton River, Lower Waterton Lake, and Sofa Creek (Parks Canada, 2018a).

In 2017, the Kenow wildfire severely burned a contiguous area of ~35 000 ha, of which 19 303 ha fell within in WLNP. A wet spring season resulted in abundant fuel, while a hot, dry summer and strong winds created conditions for fire ignited by lightning which exhibited extreme fire behaviour and spread rapidly (Eisenberg et al., 2019). The Kenow fire burned approximately 75% of the organic material, resulting in a high severity burn, which is an uncommon behaviour for wildfires in the park (Figure 3.2; Parks Canada, 2019).

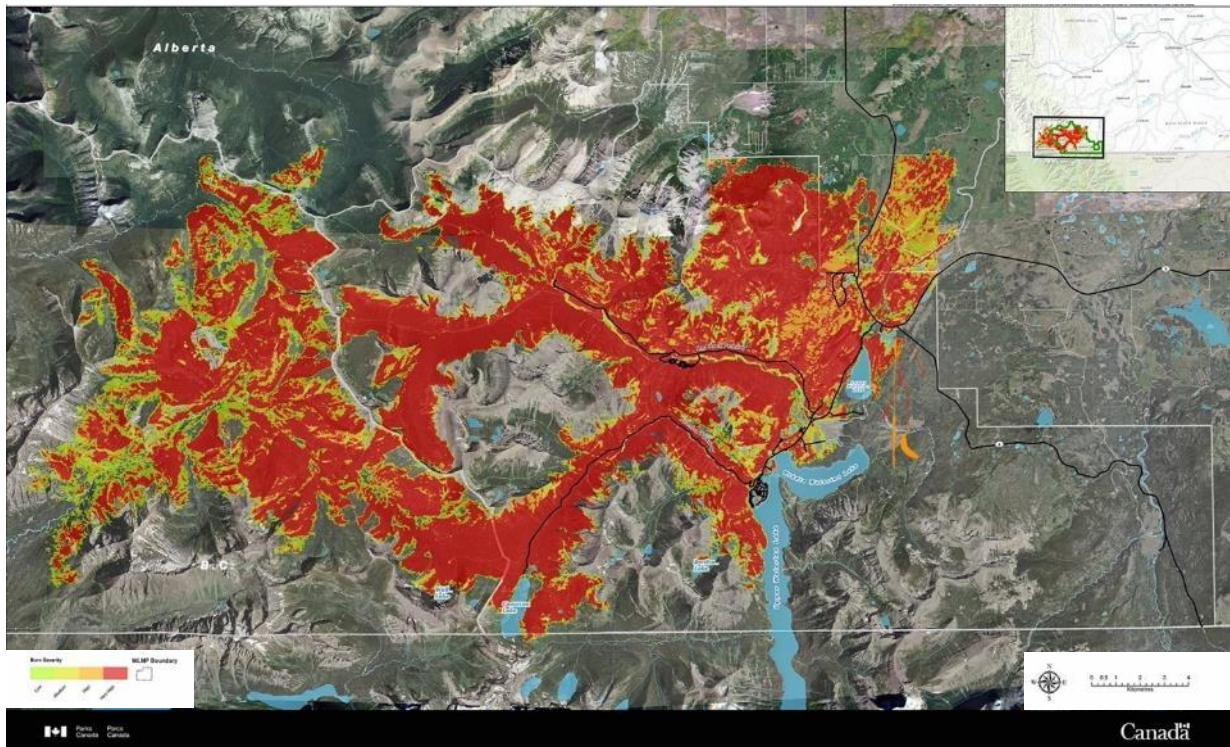


Figure 3.2 - Kenow wildfire burn area in WLNP showing fire severity and lake systems. Source: Parks Canada (2019).

3.2 Field Methods

Five sediment cores were collected using a Uwitec gravity corer in February 2020 at a depth of ~7 m from the deepest basin of Lower Waterton Lake (Fig. 3.1). All sediment cores were sectioned at 0.5 cm intervals using a vertical core extruder (Glew, 1988). Each interval of the sediment core was stored in plastic bags then immediately frozen until analysis. The longest core was selected to complete following laboratory methods.

3.3 Laboratory Methods

Loss-on-ignition (LOI) was used to measure the content of water, organic matter (%OM), mineral matter (% dry weight) and carbonate (%CaCO₃) in sediments following standard methods

(Heiri et al., 2001). LOI was measured for every 0.5 cm interval of sediment using ~ 0.5 g of wet sediment per interval (n=105). Sediment samples were first placed into pre-weighted porcelain crucibles and heated in an oven at 90°C oven for 24 hours, and then placed in a desiccator for at least 2 hours to cool to room temperature before weighing to 3 decimal places. The weight loss after heating to 90°C was used to determine percent water content (% wet weight). Organic matter content (% dry weight) was calculated by placing the dried samples in a muffle furnace and heating to 550°C for 2 hours and calculating the weight loss (Heiri et al., 2001). Mineral matter (% dry weight) is the total dry weight subtracted by the organic matter weight. The amount of carbonate-free mineral matter in samples was then determined by combusting the samples at 950°C for 2 hours, followed by a desiccator for 2 hours, then weighed. Mineral matter content (% dry weight) was calculated by dividing the post-950°C sediment weight by the post-90°C sediment weight (Heiri et al., 2001).

Stratigraphic analysis of activities of Pb^{210} and Cs^{137} was used to develop age-depth relations and temporal variation in sedimentation rate (Schelske et al., 1994) for selected intervals. Each of the top 10 intervals then every fourth interval thereafter was analyzed and results were interpolated between data points. A subsample of approximately 4.35 g (n= 28) of each freeze-dried interval of the core was packed tightly into pre-weighed, polystyrene tubes to a standard height of 35 mm. A thin silicone septum was placed on top of the sediment, followed by 1cm³ of Epoxy resin to seal the samples. Samples were then left for a minimum of 20 days to allow Rn^{222} and its decay products to equilibrate with Ra^{226} prior to analysis of Pb^{210} , Bi^{214} , and Pb^{214} activity. Samples were analyzed for radioisotope activity using a Ortec co-axial HPGe Digital Gamma Ray Spectrometer (Ortec GWL-120-15) interfaced with Maestro 32 software at the University of Waterloo Environmental Change Research (WATER) laboratory. Measured Pb^{210} activities were decay-corrected to the date when the core was collected and corrected for density (total sediment and bag weights for each sample) (Schelske et al., 1994). The Constant Rate of Supply (CRS) model was used for sediments with unsupported Pb^{210} concentrations (Appleby, 2001). A Cs^{137} peak was assumed to correspond to the ~1963 nuclear testing in the Northern Hemisphere and was used to assess the accuracy of Pb^{210} ages. Every fourth interval was analyzed and intervals between each were dated by interpolation through linear regression.

The major element composition of selected core sections was analyzed at a commercial laboratory, Act Labs (Hamilton, Ontario) using ICP-MS. Elements were determined after partial digestion using EPA digestion method 350. Quality assurance and quality control for metal analysis was ensured by including method blanks and duplicate analysis of samples. The elemental composition within discrete sediment intervals were analyzed using mass spectrometry and the results were reported as percent dry weight. To confirm analytical accuracy and validate data, Canadian Reference Materials were compared with stated reference values for major elements (see Appendix B).

Particulate phosphorus (PP) was fractionated into five operationally defined forms, using the sequential extraction scheme described by Petterson et al. (1988) at a commercial laboratory (Act Labs, Burlington). Various extracts are used to remove phosphorus fractions from sediment in order of decreasing bioavailability. First, a three-step reactive phosphate fractionation method removes non-apatite inorganic phosphorus (NAIP) which is the sum of three reactive phosphate fractions. Loosely sorbed P is removed by NH_4Cl , bicarbonate-dithionite (DB) is used to remove reductant soluble phosphorus (NaOH-RP), and NaOH to removed metal oxide bound P (Boström and Pettersson, 1982). Apatite P (AP) is then extracted with 25mL of 0.5M HCl for 24 hours. OP is extracted with 1M NaOH at 85°C for 24 hours. Samples were run in triplicate for QA/QC, see Appendix B.

Batch experiments were used to determine Equilibrium Phosphorus Concentration (EPC_0) which is a measure of the potential of sediment to release or absorb P either to or from the water column (Froelich et al., 1988). A 25 mL aliquot of phosphorus concentrations (25, 50, 100, and 200 $\mu\text{g/P L}^{-1}$) and 25 mL of deionized (DI) water was added in triplicate to 0.25 g of freeze-dried sediment from each interval in 50 mL centrifuge tubes. Tubes were then placed on a shaker table at room temperature for 18-hours at 100 rpm. Aliquots were then passed through a 0.45 μm filter into 20mL scintillation vials and refrigerated. A Bran-Luebbe AutoAnalyzer III system multi-test MT18 was used to measure SRP concentrations using the ascorbic acid reduction colorimetric method (detection limit of 1 $\mu\text{g P L}^{-1}$) at the Biogeochemistry Lab at the University of Waterloo. QA/QC results of EPC_0 experiments are presented in Appendix B. To calculate the amount of phosphorus adsorbed to or desorbed from lake bottom sediment the following equation was used,

$$P_{ads} = \frac{[(P_{initial} - P_{final}) * 0.025L]}{wt_{sed}} \quad \text{eqn. 2.3.3}$$

Where P_{ads} = P adsorbed by sediment ($\mu\text{g P g}^{-1}$ dry weight sediment), $P_{initial}$ = initial phosphate concentration of solution ($\mu\text{g P L}^{-1}$), P_{final} = final phosphate concentration ($\mu\text{g P L}^{-1}$), wt_{sed} = weight of dry sediment (g). A sediment buffer diagram was used to plot P_{ads} against $P_{initial}$. The EPC_0 of sediment is the point on the buffer diagram in which the solids display the maximum capacity for buffering solution P. Positive values of EPC_0 indicate sediment is adsorbing phosphorus from the overlying water column to the sediment, and accordingly negative EPC_0 values indicate that sediment is desorbing phosphorus from sediment to the water column (Froelich, 1988). The maximum potential release of SRP by sediment to the water column is indicated by the x-intercept of the sediment buffer diagram.

To estimate the SRP diffusive flux across the sediment-water interface, SRP data from the Waterton River (Southern Rockies Watershed Project, 2020) were considered a proxy for lake SRP concentrations and data from the sediment buffer diagrams were used to provide estimates of the mass of phosphorus released or adsorbed per gram of benthic sediment. Water samples were collected downstream of Lower Waterton Lake, biweekly, during the ice-free season from April to November in 2018 and 2019. Assumptions used to calculate the total mass of phosphorus release from the entire lake bottom sediments include 1) assume that downstream water column SRP concentrations reflect lake water column SRP concentrations, 2) the area of the sediment core and the release of P from the top layer sediment layer is representative of lakebed phosphorus release and 3) approximately 80% of the lakebed area consists of the fine sediment fraction active in geochemical cycling. To calculate the estimated total lake phosphorus concentration from the potential SRP desorption the following equation was used,

$$P_{total} = \frac{(P_{release} * area_{lake})}{area_{core}} * 0.8 \quad \text{eqn. 2.3.4}$$

Where P_{total} = potential total water column phosphate ($\mu\text{g P}$), P_{release} = mass of SRP desorbed from sediment to the overlying water column ($\mu\text{g P}$), $\text{area}_{\text{lake}}$ = area of Lower Waterton Lake bottom (m^2), $\text{area}_{\text{core}}$ = area of sediment core from the internal diameter of the core.

Chapter 4

Results

4.1 Sediment core chronology and sediment composition

The stratigraphic profiles of organic matter (%OM), mineral matter (% dry weight), and carbonate content (%CaCO₃) are presented in Figure 4.1 to illustrate temporal change in core composition. Vertical distribution of organic, mineral and carbonate content were relatively stable throughout the core. Organic matter makes up < 10 % of sediment material, whereas mineral matter comprises > 90% of sediment material. Notably, there is a dramatic change in %OM and %MM in 2007 (top 9 cm), where %OM rapidly increases from ~4 % to ~9 % by 2019, and %MM rapidly decreases from ~96% to 91% by 2019.

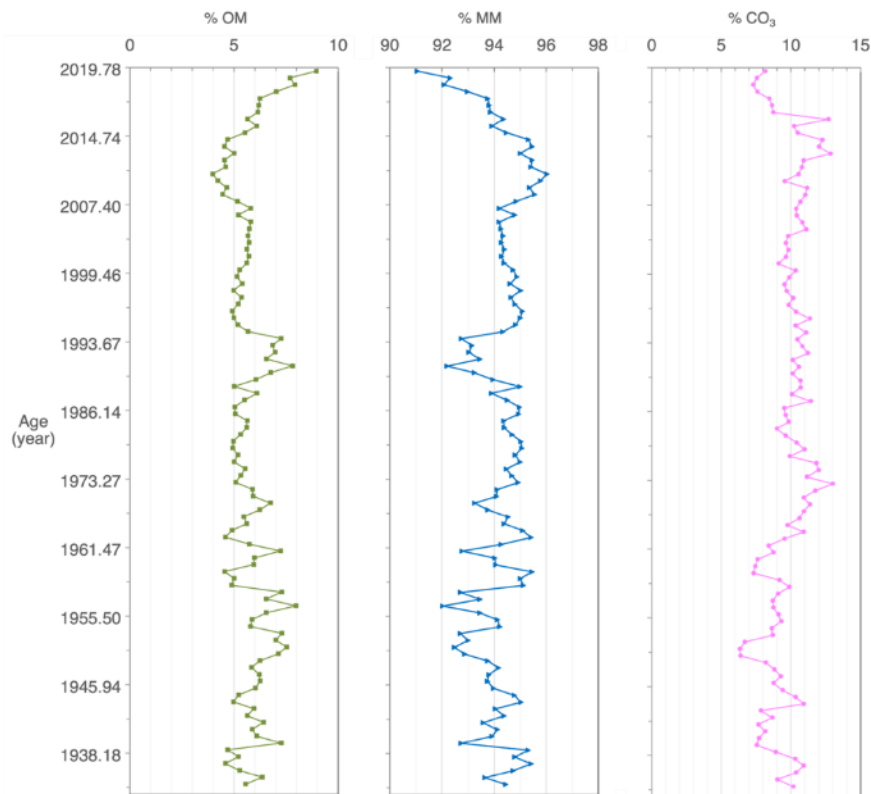


Figure 4.1- Temporal distribution in percent organic matter (%OM), percent mineral matter (%MM), and percent carbonate content (%CO₃) in Lower Waterton Lake sediment core (note scales).

Radiometric data were utilized to develop sediment core chronology for Lower Waterton Lake. The ^{210}Pb activity profiles in the lake sediment core decreased with depth, but with marked variability (Figure 4.2). The ^{226}Ra and ^{210}Pb activity profiles indicate that background ^{210}Pb levels were not reached. The age-depth relationship is plotted in the right panel of Figure 4.2. The CRS model determined the basal date of the core to be ~ 1940 (± 10 years, 2 SD, Figure 4.2). The ^{137}Cs activity profile does not decline with depth and a bifurcated peak is present at the 30.5-31.0 cm and 42.5-43.0 cm intervals corresponding to ~ 1973 (± 5 years, 2 SD) and ~ 1952 (± 9 years, 2 SD), respectively. The ^{137}Cs peak from nuclear testing in 1963 does not appear. This is likely due to massive dilution of the atmospheric ^{137}Cs fallout signal and massive change in lake sedimentation rate seen in 1961 (Figure 4.2).

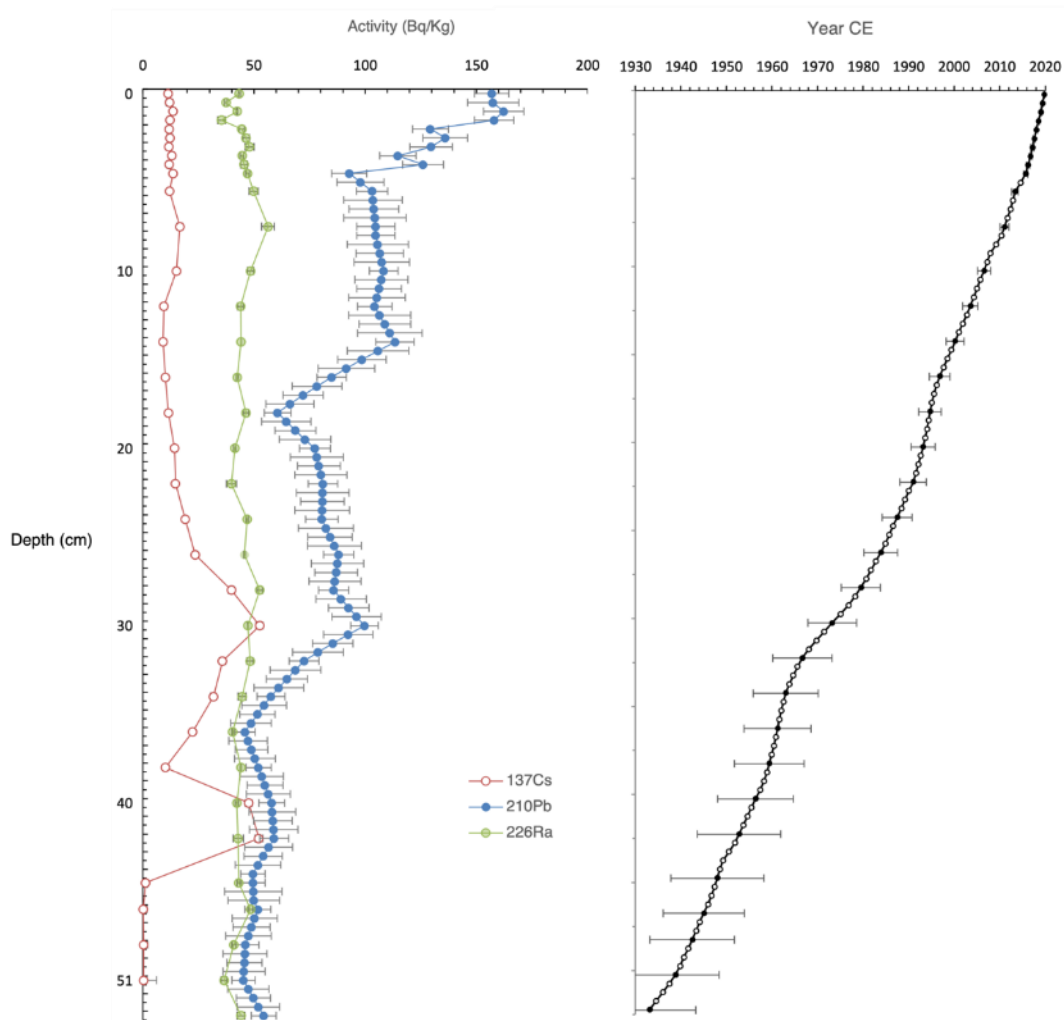


Figure 4.2 – Activity profiles of radioisotopes ^{210}Pb (blue circles), ^{137}Cs (open red circles), and ^{226}Ra (open green circles) in disintegrations per minute/gram (Bq/Kg) for sediment core from Lower Waterton Lake. The age depth relationship is plotted in the right panel. Open circles are interpolated using the CRS model. Error bars represent 1 standard deviation.

Based on the ^{210}Pb dating, the lake sedimentation rate is estimated to have fluctuated due to four episodes of elevated sedimentation above the baseline of $\sim 0.5 \text{ g/cm}^2/\text{yr}$ at ~ 1947 ($1.0 \text{ g/cm}^2/\text{yr}$), ~ 1961 ($2.0 \text{ g/cm}^2/\text{yr}$), ~ 1995 ($2.0 \text{ g/cm}^2/\text{yr}$), and ~ 2013 ($1.0 \text{ g/cm}^2/\text{yr}$) (Figure 4.3). Total sediment flux was dominated by deposition of minerogenic sediment. The highest peaks in sedimentation dates rate likely correspond to large flood events with periods of high discharge into the study lake during unusually high spring flood events (Parks Canada, 2018a).

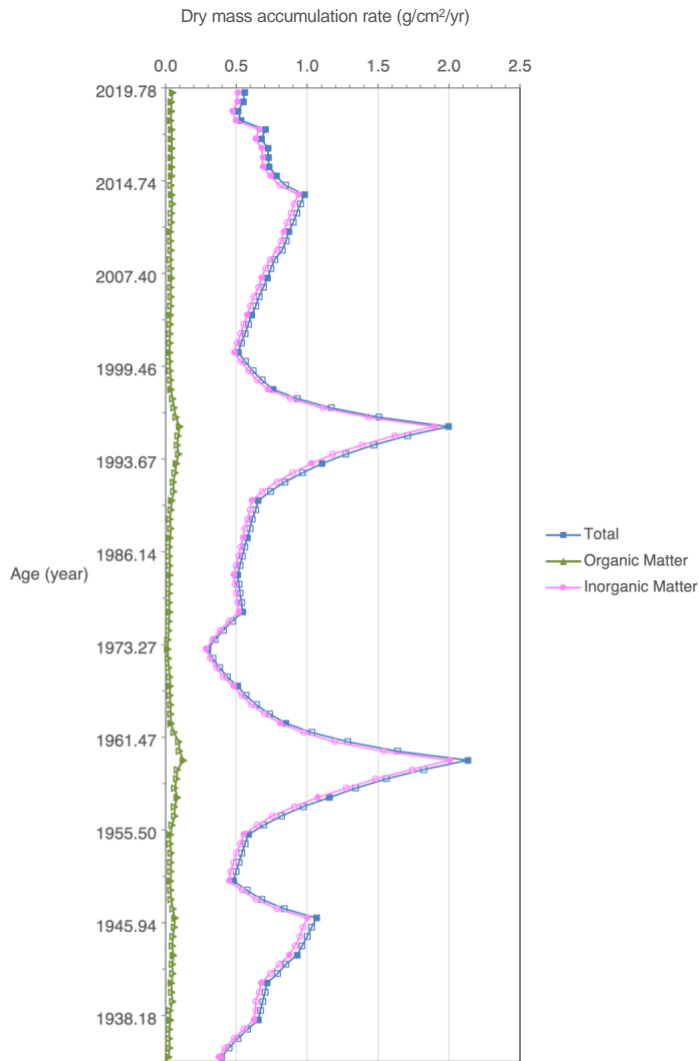


Figure 4.3 – Sedimentation rate in $\text{g/cm}^2/\text{year}$ with organic matter (green triangles), inorganic matter (pink circles), and total matter (blue squares). Pre-2015, every fourth interval was run (filled data points) and sedimentation rate via interpolation (open data points).

4.2 Temporal variations in major element concentrations

The geochemical composition of each sediment interval was analyzed to determine whether vertical differences in the distribution indicate temporal changes in sediment deposition in Lower Waterton Lake. The key indicator ions (Al, Mn, Fe, Ca, Mg) related to particulate phosphorus form are summarized in Figure 4.4. Al and Ca have relatively small fluctuations from the bottom of the core (~1940) until ~2013 where there is a decreasing trend in % dry mass. There was a relatively small increase in Mg from ~1940 to ~2017 which then declined until 2019. Fe remained relatively stable between ~2010 to 2017 where there was a small dip in % dry mass and then restabilized. Mn remained stable throughout the core. Overall, there was an increase in half a percentage in the top two intervals (2019) of the core for Al, Ca, Fe, Mg, and Mn (Figure 4.4). Metal oxide fractions (i.e., Al_2O_3 , Fe_2O_3 , and MnO) that are known adsorptive surfaces which bind P have a general inverse relationship with particle size which can influence the enrichment ratio (NAIP:TPP) of particulate P (Sharpley, 1980). Bioavailable PP forms (NAIP) are commonly understood to preferentially bind to these metal oxide surfaces. The vertical variation in enrichment ratio (Figure 4.4) was relatively stable from ~1940 – 2012, until there was a sharp decline in 2012 from 3 – 1.5. Thereafter, there was a gradual increase from 2012 to 2019 back to ~ 3 (Figure 4.4). The enrichment ratio remained below 50% for the entirety of the sediment core and was positively correlated with Fe (0.489, $p < 0.01$, Table 4.1).

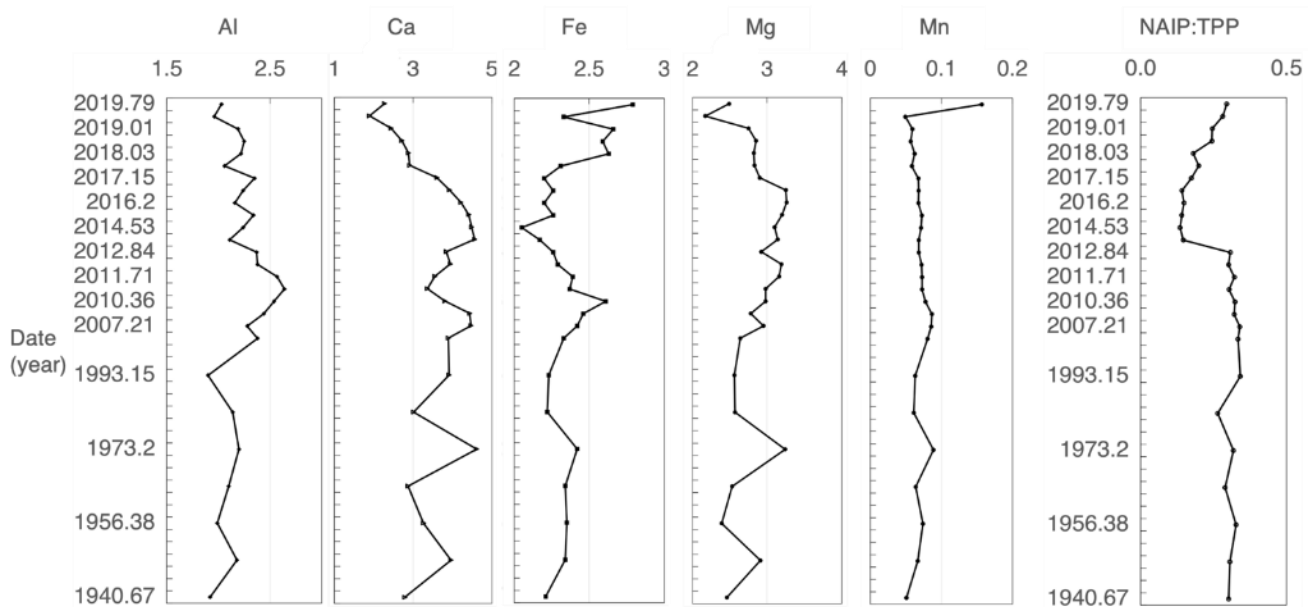


Figure 4.4 – Stratigraphic profiles of major ion concentrations (% dry mass, note scales) of aluminium (Al), calcium (Ca), iron (Fe), magnesium (Mg) and manganese (Mn) in sediment core from Lower Waterton Lake. The non-apatite inorganic phosphorus (NAIP) and total particulate phosphorus (TPP) ratio is presented as a phosphorus enrichment ratio (NAIP:TPP).

4.3 Temporal variability in form of particulate phosphorus in sediment core

Temporal variation in the sedimentary PP forms (i.e., NAIP, AP, OP) in Lower Waterton Lake sediment core are presented in Figure 4.5a. TPP concentration remained relatively constant from ~1940 – 2007, then declined from ~2007 – 2013, and then increased after 2013. The highest concentration of TPP in 2019 was $725.0 \mu\text{g/g}$ and the lowest concentration $439.2 \mu\text{g/g}$ in 2014. Overall, of the three PP forms, AP comprised the highest concentration 61% ($360.2 \mu\text{g P g}^{-1}$) on average, followed by NAIP (26%; $153.5 \mu\text{g P g}^{-1}$), and OP at 12% ($75.5 \mu\text{g P g}^{-1}$).

Temporal variability in the most bioavailable PP form (NAIP) is presented in Figure 4.5b to evaluate changes in P inputs to the lake overtime. Total NAIP ranged from a maximum of $214 \mu\text{g P g}^{-1}$ in 2019 to a minimum of $76 \mu\text{g P g}^{-1}$ in 2014. The concentration of NAIP decreased from $\sim 162 \mu\text{g P g}^{-1}$ to $\sim 84 \mu\text{g P g}^{-1}$ around 2013 which corresponds to the marked increase in AP (from $\sim 305 \mu\text{g P g}^{-1}$ to $\sim 400 \mu\text{g P g}^{-1}$). NAIP levels remained around $90 \mu\text{g P g}^{-1}$ until 2018 but by 2019

it increased to $214 \mu\text{g P g}^{-1}$. The BD-RP fractions were highest on average at 55% ($88.3 \mu\text{g P g}^{-1}$), followed by NaOH-RP at 37% ($55.2 \mu\text{g P g}^{-1}$). The NH_4Cl fraction comprised $\sim 7\%$ ($< 10 \mu\text{g P g}^{-1}$). Correlations between particulate phosphorus form and major elements are presented in Table 4.1. OP and Ca, and TPP and Fe had the strongest positive relationships ($p = < 0.01$). TPP was negatively correlated with Al, Ca, and K ($p = < 0.05$).

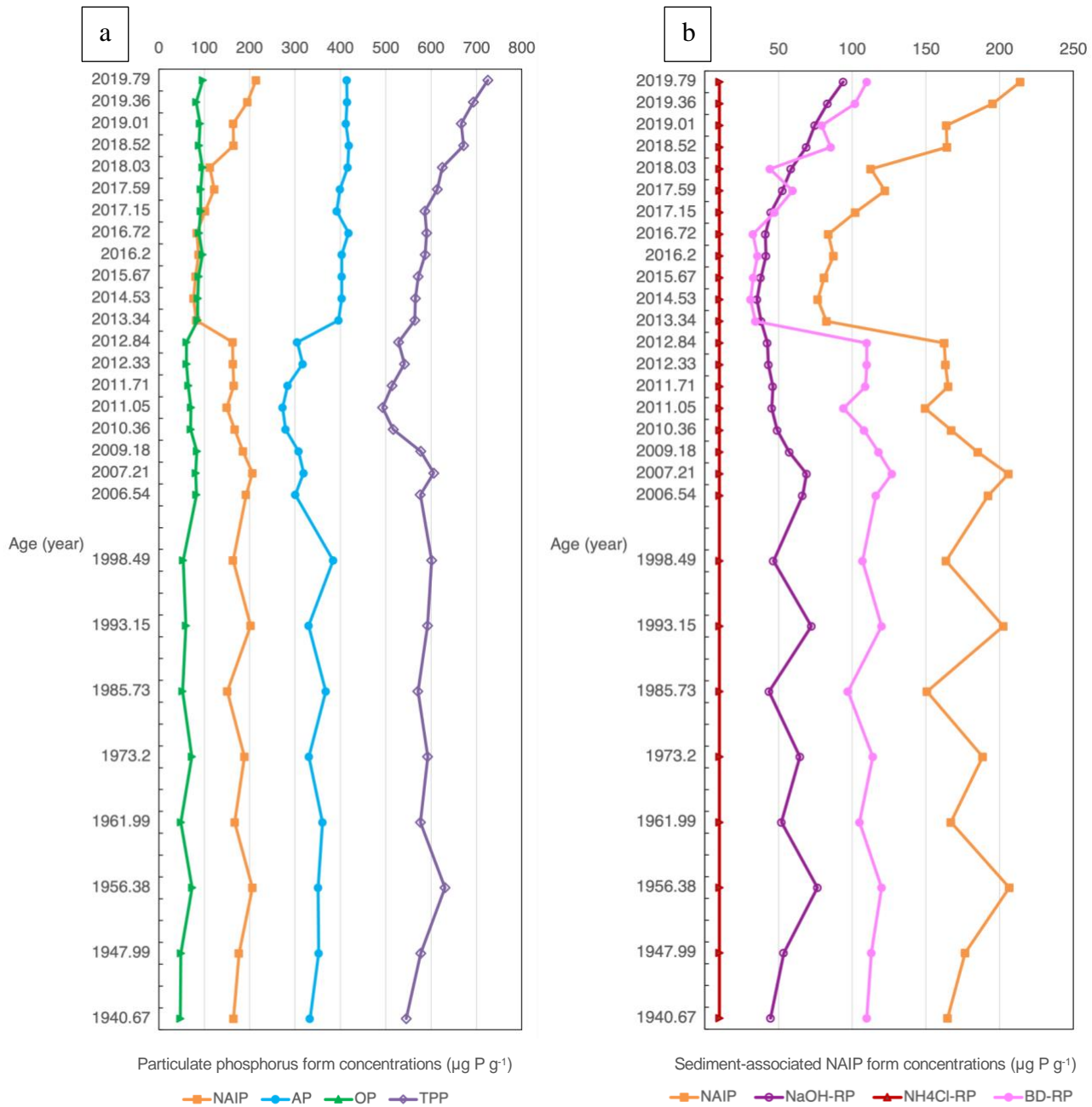


Figure 4.5 – Stratigraphic profiles of particulate phosphorus concentrations (a; $\mu\text{g P g}^{-1}$). Non-apatite inorganic phosphorus (NAIP; orange squares), apatite phosphorus (AP; blue circles), organic phosphorus (OP; green triangles), and total particulate phosphorus (TPP; purple diamonds). Stratigraphic profiles of sediment-associated NAIP forms (NaOH-RP, NH₄Cl-RP, and BD-RP) shown on b.

Table 4.1 – Correlation coefficients between particulate phosphorus form and major elements (n=28).

	NAIP	AP	OP	TPP
Al	-0.239	-0.291	-0.105	-0.578*
Fe	-0.190	0.186	-0.237	0.489**
Mn	-0.235	-0.046	-0.112	0.270*
Mg	-0.007	-0.385	-0.041	-0.469
Ca	-0.296	-0.113	0.423**	-0.557*
Na	0.024	0.246	0.319*	-0.082
K	-0.329	-0.24	0.086	-0.732*

Unmarked $p < 0.1$, * $p < 0.05$, ** $p < 0.01$

4.4 Phosphorus mobility

Temporal variability in EPC_0 is presented in Figure 4.6 to evaluate historical changes in P mobility in the lake benthic sediment in oxic conditions overtime. EPC_0 concentrations of sediment intervals varied considerably in the sediment core (see Appendix A for EPC_0 regressions). Three distinct peaks in EPC_0 concentrations were observed in 2019 ($18 \mu\text{g P L}^{-1}$), 2017 ($23 \mu\text{g P L}^{-1}$) and 2007 ($21 \mu\text{g P L}^{-1}$) (Figure 4.6). The lowest EPC_0 concentrations were associated with sediments in deposited around 2018 and 2014 (7 and $1 \mu\text{g P L}^{-1}$, respectively). The median EPC_0 value for the entire core was $7.6 \mu\text{g P L}^{-1}$ and the mean was $9.9 \mu\text{g P L}^{-1}$. Elevated peaks in EPC_0 correspond to increases in disturbance (e.g., wildfires, floods) in the Lower Lakes watershed. Historical wildfire activity (1998 – 2008) and flood events (2005 and 2017) show the largest spikes in EPC_0 , $> 20 \mu\text{g P L}^{-1}$.

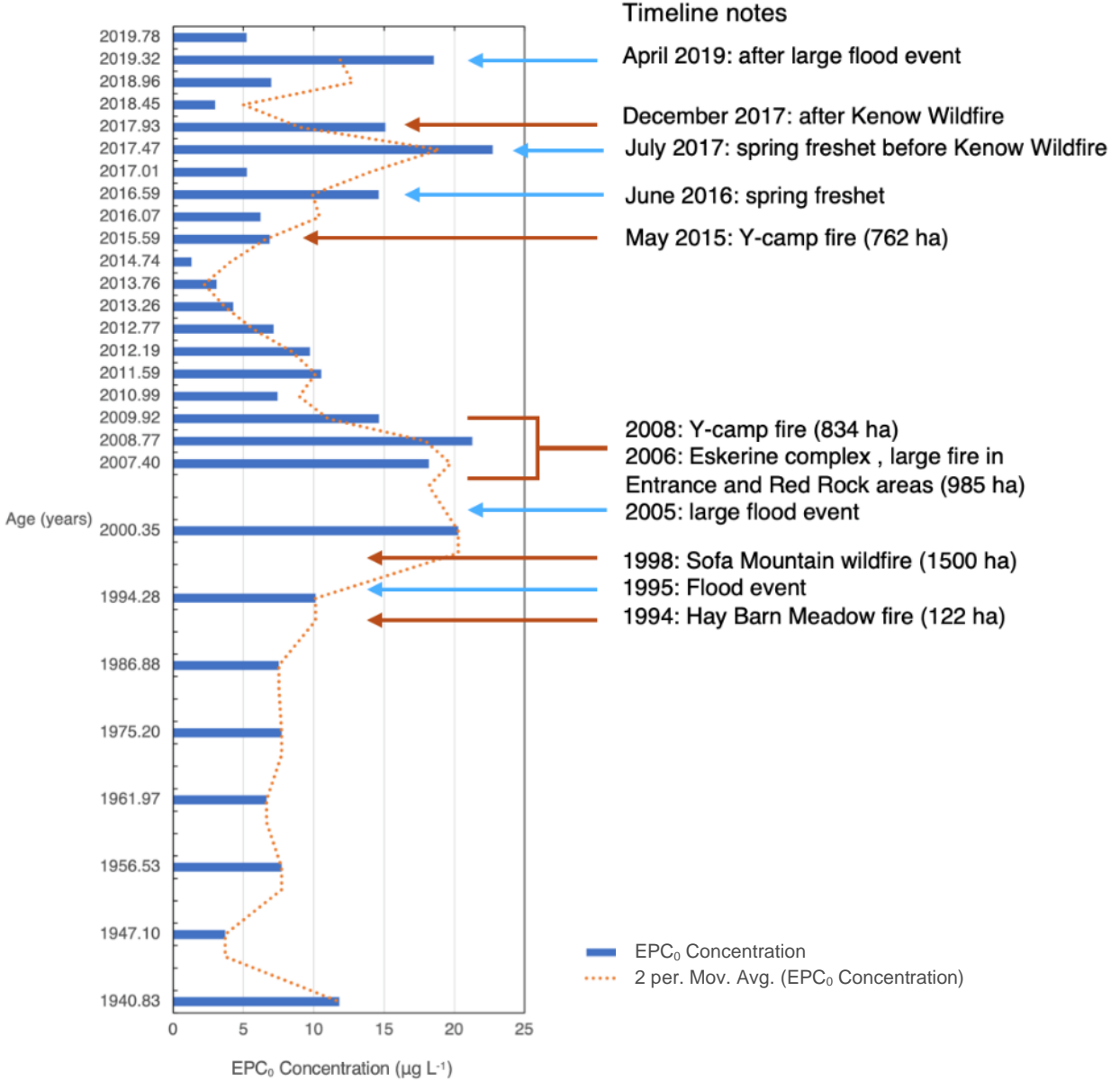


Figure 4.6 – Vertical profile of EPC₀ as a function of depth. Orange line represents the moving average, interpolated between analyzed values.

4.5 Estimates of P flux from benthic sediments

To estimate the potential release of phosphorus to the water column from the top 5 cm layer of benthic sediment it was necessary to know the dissolved P concentration in the lake. However,

since no soluble P measurements were made when sediment coring was conducted, monthly water chemistry data from the outflow of Lower Waterton Lake was used as a proxy (unpublished Dr. Uldis Silins, University of Alberta). Monthly water column SRP concentrations from 2018 to 2019 at the lake outflow and sections of the core were used to determine maximum SRP release potential (0-2.5 cm). Unpublished water chemistry data indicated that median SRP concentrations in WLNP rivers and surrounding areas were low, ranging from $0.5 \mu\text{g P L}^{-1}$ – $6.0 \mu\text{g P L}^{-1}$ over 24 months. The mean annual SRP value was used to determine maximum SRP release potential (Table 4.2). In 2018, SRP concentrations in the lake outflow ranged from $0.5 - 3 \mu\text{g P L}^{-1}$, with the median value of $1.6 \mu\text{g P L}^{-1}$. In 2019, SRP concentrations in the river ranged from $1 - 6 \mu\text{g P L}^{-1}$, with the median value of $2.6 \mu\text{g P L}^{-1}$. Equation 2.3.4 was used to estimate the SRP diffusive flux across the sediment-water interface. Estimated potential SRP desorption from sediment was generally low, ranging from $0.26 \mu\text{g P/g sediment}^{-1}$ in fall 2019 at 0-0.5 cm – $1.41 \mu\text{g P g/sediment}^{-1}$ in spring 2019 at 0.5-1.0 cm. Benthic sediment from sediment core intervals dated as the same SRP sampling years, all demonstrated potential to release SRP into the overlying water column (Table 4.2). The highest internal P loading of 293.6 g was estimated in spring 2019. The lowest potential of internal P loading was 29.2 g in the summer 2018.

Table 4.2 – Estimates of phosphorus flux from lake sediments as an indicator of internal P loading Soluble reactive phosphorus (SRP) data were measured in the Waterton River, immediately downstream of LWL core location. Negative values of potential SRP desorption ($\mu\text{g P/g sed}^{-1}$) represent the release of SRP from the sediment to the water column.

Year	Depth (cm)	Median yearly SRP ($\mu\text{g /L}$)	EPC_0 ($\mu\text{g P/L}$)	Potential SRP desorption ($\mu\text{g P/g sed}^{-1}$)	Estimates of internal P loading (g)
2019	0 – 0.5	2.6	5.22	-0.26	54.2
	0.5 – 1.0	2.6	18.54	-1.41	293.7
	1.0 – 1.5	2.6	6.97	-0.38	79.1
2018	1.5 – 2.0	1.6	2.98	-0.14	29.2
	2.0 – 2.5	1.6	15.08	-1.28	266.6

Chapter 5

Discussion

The goal of this thesis was to quantify the concentration and distribution of particulate P forms, major elements, and estimate the potential of wildfire derived benthic sediment as an internal source of P loading in Lower Waterton Lake. Watershed managers require multiple data sources to make science-based planning and management decisions. Knowledge of long-term data trends that enable background (or pre-impact) conditions to be compared with current conditions are useful to illustrate natural variability in environmental processes and to develop models that simulate potential future scenarios (Smol, 1992). Sediment cores can be used to provide key insights to water quality managers by measuring and interpreting physical, chemical, and biological indicators preserved in sedimentary records (Smol, 1992). The management of freshwater supplies are becoming increasingly difficult due to the uncertainty regarding shifts in land disturbance and hydroclimate due to climate change, which is of strategic and national importance for water supply (Emelko et al., 2016). Lake response to catchment disturbance depends on the resiliency of the system, combined with the nature and severity of the disturbance (Kpodonu et al., 2019). The temporal history of lake response following historical landscape disturbance (e.g., wildfire, flooding) is necessary to inform the future of ecosystem response based on historical observations (Smol, 2010).

5.1 Temporal changes in the distribution of particulate phosphorus forms

Geomorphic features and disturbance type in source watersheds are important factors influencing the delivery and storage of sediment and associated nutrients and contaminants in lakes. While the delivery of materials can be delayed or reduced to receiving lakes, these waters can still be altered by the addition of pyrogenic materials and sediment-associated nutrients, such as phosphorus (Moody and Martin, 2004; Silins et al., 2009; Wilkinson et al., 2009; Burd et al., 2018). Lower Waterton Lake is oligotrophic (CCME, 2004) and the addition of limiting nutrients (e.g., P) to this lake can alter nutrient dynamics, potentially causing increases in primary productivity (Schindler, 1977; Carpenter, 1998; Corell, 1998; Emelko et al., 2016). Several studies

examining the vertical distribution of sediment-associated P in lake sediment cores, report that TPP commonly increases with sediment depth (Boström et al., 1982; Carey and Rydin, 2011; Wilson et al., 2010) and that these lake sediments generally exhibit greater Al-bound P than eutrophic lakes (Kopáček et al., 2005). Notably, sediment cores collected in oligotrophic lakes often have elevated levels of TPP at the sediment-water interface (Wilson et al., 2010; Carey and Rydin, 2011). This was observed in the sediment core of LWL, where TPP increased in the top 2 cm of sediment (Figure 4.4a). This may be due to the release of P into the water column or indicate there is P incorporated into active pore water biota (Ahlgren et al., 2006). Additionally, vertical mixing of the water column from turbulent winds, can increase oxygen concentration and, desorbing loosely bound P (Boström et al., 1982; Smol, 2008). The shallow lake depth combined with the normal occurrence of strong winds in WLNP and LWL may promote the desorption of P from the sediment to the water column.

In most undisturbed forested environments, the losses of P in runoff are low (Burke et al., 2005). However, following wildfire, elevated concentrations of TPP have previously been reported in rivers (Allin et al., 2012; Emelko et al., 2016; Tullio, 2022). The enrichment ratio is a measure of the uniform contribution of TPP from catchment to the system. Previous studies have reported high enrichment ratios in the order of 3-6 of phosphorus on fine sediment following wildfire (Saá et al., 1994). However, Lane et al. (2011) reported enrichment ratios of 1.5 – 2 following wildfires. The discrepancy between enrichment ratios was attributed to short transport pathways from hillslope to stream (Lane et al., 2011). The enrichment ratios of sediment from LWL following high wildfire periods in WLNP were 25% in 2000, 30% in 2018 and 27% in 2007 (Appendix A3), while Tullio (2022) reported enrichment ratios of 53% - 63% in burned river sediment from Blakiston Creek following the Kenow Wildfire. The modest difference in enrichment ratios for the WLNP area demonstrates that there are other important controls which influence PP form and concentration in sediment such as regional geology and geomorphology, water chemistry, and disturbance. Additionally, the difference of site selection between river and lake means that spatial heterogeneity in hydraulic conductivity differences probably acted to minimize surface flow pathways in lake sediment (Lane et al., 2011).

In rivers with wildfire disturbance, concentrations of bioavailable PP forms (i.e., NAIP) have been reported in the water column ~5 times greater than in rivers draining unburned

landscapes (Emelko et al., 2016; Smith et al., 2011). During periods of high wildfire activity in WLNP (e.g., Kenow Wildfire in 2017, Y-camp prescribed burn in 2008, Eskerine complex prescribed burn in 2006, and Sofa Mountain wildfire in 1995), elevated NAIP concentrations were recorded in the sediment core of LWL. One year following the 2017 Kenow wildfire, mean NAIP concentrations increased from 112.7 ug P g⁻¹ dw to 164.3 ug P g⁻¹ dw from December 2017 to December 2018 . Following the large, prescribed burns in 2006 and 2007 (Erskine Complex and Hay Barn Prescribed Burn), mean NAIP concentrations increased from 163.6 ug P g⁻¹ dw to 206.0 ug P g⁻¹ dw. Following the 1995 Sofa Mountain wildfire, mean NAIP concentrations increased from 150.8 ug P g⁻¹ dw to 202.4 ug P g⁻¹ dw. While the concentrations are shown to have increased in LWL, they are not to the extent we expected. In the upstream Blakiston Creek, following the Kenow Wildfire, Tulio (2022) reported NAIP concentrations from 200 – 800 ug P g⁻¹ dw. The difference in NAIP concentration between Blakiston Creek and LWL may be the presence of landforms in the watershed that act as a sink to sediment-associated P forms. Although concentrations of NAIP were unexpectedly low, previous research on lakes affected by wildfire show comparable concentrations of NAIP compared to LWL (Emelko et al., 2016). The Oldman reservoir is oligotrophic and was impacted by recent wildfires and has very similar concentrations of NAIP fractions compared with LWL (<200 ug P g⁻¹ dw) (Figure 5.3). Both catchments received eroded materials from sources of sedimentary glacio-lacustrine materials rich in Ca and Mg. This highlights the importance of catchment geology in the role of PP forms. Indeed, the NAIP concentrations observed in a wildfire-impacted lake are unexpectedly lower than those observed in agriculture-impacted watersheds, non-stationarity of changing climate may present more frequent wildfire and flood events, increasing NAIP concentrations (Flannigan et al., 2005; Milly et al., 2008).

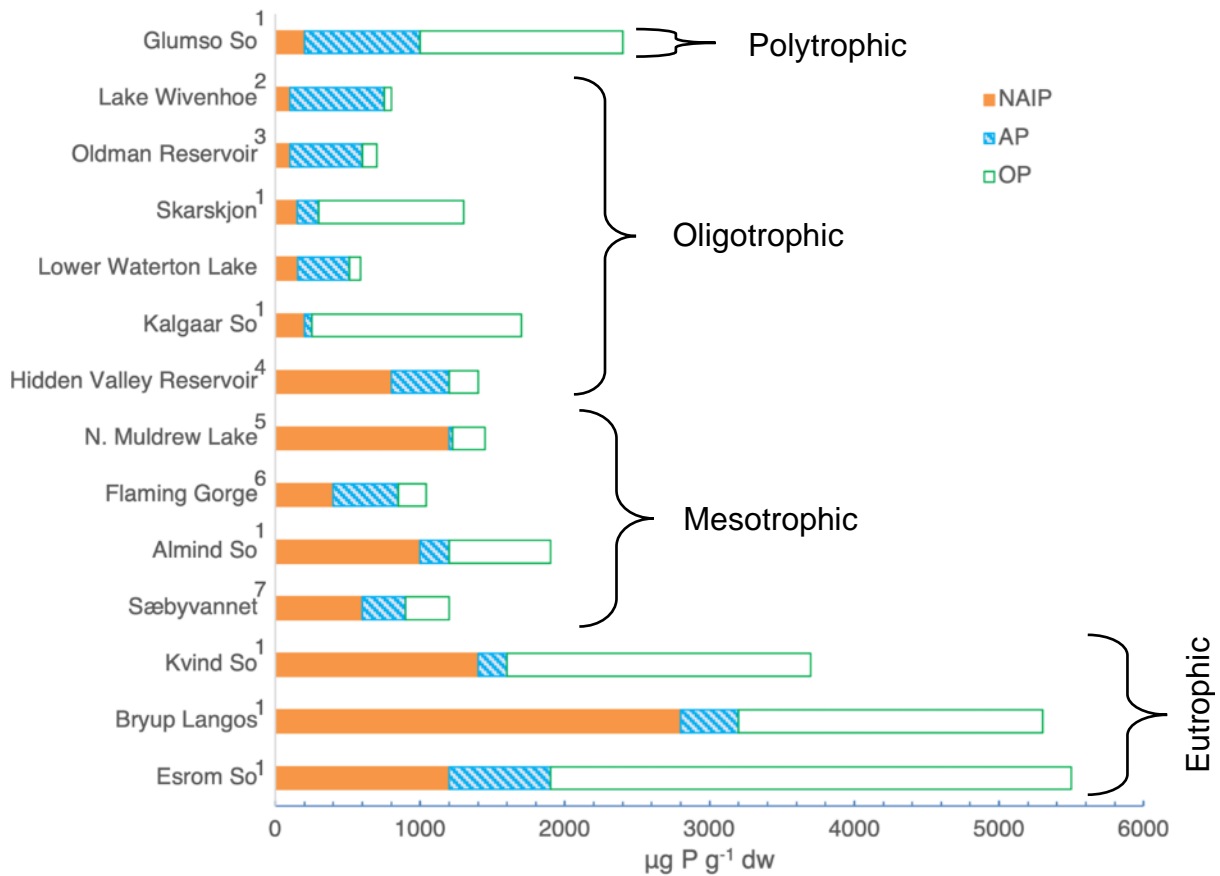


Figure 5.1 – Comparison of sediment associated particulate phosphorus forms (PP) from Lower Waterton Lake along with other lakes and reservoirs impacted by land disturbances. The data from the study lake is a composite mean value of PP forms. ⁶Messner et al., 1984; ⁵White and Stone, 1996; ¹Pettersson et al., 1988; ²Kerr et al., 2011; ⁷Łukawska-Matuszewska et al., 2013; ³Emelko et al., 2016; ⁴Knezic, 2021.

Other PP forms (e.g., AP, OP) in LWL were similar to oligotrophic lakes impacted by land disturbances (Figure 5.3) In LWL, AP was the most abundant particulate phosphorus fraction. This is analogous to Lake Wivenhoe (Kerr et al., 2011) and Oldman Reservoir (Emelko et al., 2016), which show large fractions of AP and low amounts of OP. The higher AP concentration is likely

due to erosion of surficial deposits glacial fluvial origin the watershed (Eyles et al., 2000). The concentration of AP is commonly elevated in lakes with a calcareous sediment bottom, since it is the only phosphorus form to be conserved as an allochthonous mineral (Pettersson et al., 1988). While AP and NAIP were found at higher concentration in LWL, OP remained relatively constant at a lower concentration in the sediment core and could be attributed to several reasons that include, the ingress of particulate organic matter in floodplains and gravel-beds reduces lake loading rates (Meyer et al., 1992; Moody and Martin, 2001) and the pristine nature of Waterton Lakes National Park, where there is limited anthropogenic inputs of P (Parks Canada, 2018b).

While physiographic characteristics of catchments, land use and disturbance type partially explain changes in the spatial and temporal distribution of PP forms in aquatic systems (Stone and Mudroch, 1989; Withers and Jarvie, 2008; Records et al., 2016), other factors such as temperature, redox conditions, and sediment geochemistry can influence the loading and subsequent distribution of PP forms in lake sediment (Owens and Walling, 2002; Stone and English 1993). The bioavailability of PP and potential of phosphorus to bind to sediment, can be influenced by the geochemical composition of sediment (Stone and English, 1993). This was observed in LWL, as the dominating NAIP species is reductant soluble reactive P (BD-RP) (Figure 4.4b) and may point to the presence of Mn and Fe-oxides within WLNP. The Red Rock Canyon is situated 17 km upstream in Blakiston Creek and is formed of Fe-rich argillite (Parks Canada, 2018b). Concentrations of Mn and Fe remained relatively stable throughout the core, except in the top 0.5 cm which had increasing Mn concentrations. This may be due to a redox gradient at the top of the sediment core (upper 2 cm), as overlying oxic water oxygenates the sediment at the sediment-water interface. Manganese may be increasing in concentration here due to intense mineralization processes close to the sediment-water interface (Granina et al., 2004). Following wildfire, significant concentrations of Mn are found at elevated concentrations in streams and rivers impacted by wildfire (Tulio, 2022; Emelko et al., 2016). The increase in Mn may also be due to the influx of pyrogenic materials, which typically have elevated levels of Mn (Bodí et al., 2014). Pyrogenic derived materials in wildfire affected systems, can be continually eroded from the terrestrial landscape after the fire (Bodí et al., 2014). Although Ca can be relatively stable in sediment, its concentration is highly variable in LWL. This may be due to the abundance of

calcium carbonate (CaCO_3) in ash (Bodí et al., 2017; Kozerski and Kleeberg, 1998), and long-term effects of the historical fire regimes in WLNP on LWL.

While phosphorus is primarily associated with fine sediment, including clays with Mn, Al, Fe, and organic coatings (Stone and Mudroch, 1989), its form and concentration are dependent on geochemistry (Pettersson et al., 1988). Generally, PP and elements such as Al, Fe, and Mn are positively correlated, while other elements such as Mg, Si, and Ca are negatively correlated with PP (Stone and Mudroch, 1989; Evans et al., 2004). In the current study, significant positive correlations were present between TPP and Fe ($p < 0.01$), and Mn ($p < 0.05$) concentrations, and between OP and Ca ($p < 0.01$) and Na ($p < 0.05$). Significant negative correlations between TPP and Al ($p < 0.05$), Ca ($p < 0.05$), and K ($p < 0.05$) were observed (Table 4.1). These correlations are likely a function of landscape physiography and chemistry of pyrogenic materials. In a study of the composition of suspended solids in Blakiston Creek and Cameron Creek, similar correlations were present between TPP and Mn, Ca, and Fe concentrations (Tullio, 2022). Following wildfire disturbance, previous studies have observed mixed results for concentrations of K, Cl, Ca, and Mg in lakes. Carignan et al. (2000) found 2 to 6-fold increases in concentrations of K, Cl, Ca in severely burned watersheds which were rapidly flushed out of the system. Conversely, Allen et al. (2003) and McEachern et al. (2000) reported increases in concentrations of Ca, Mg, Ba, K, and Cl to lakes, with slow recovery over decades. While these studies point to the input of major elements to surface water of lakes following wildfire, benthic sediment was not examined. Indeed, the cycling of these elements in lakes results in the flux of elements from the water column to the sediment (Dean, 1999). While environmental factors such as pH, anoxia, and temperature control lake biogeochemical dynamics, sediment concentrations are linked to water column concentrations and sediment inputs from the watershed (Dean, 1999). While ions associated with pyrogenic materials should increase in benthic sediment of lakes following wildfire (McCollough et al., 2019), the change in elemental composition strongly varies spatially from landscapes and anthropogenic activities (e.g., SO_4 deposition, erosion).

5.2 Landscape features control on lake sedimentation

While changes in the concentrations of PP forms were observed in the sediment core, they were much lower than hypothesized and were much smaller than the concentrations of PP

measured in the tributary inflow (e.g., Blakiston Creek). Several factors, such as landscape forms and hydraulic patterns in the watershed, may account for the differences in accumulation and concentrations. Coincidental controls such as geomorphic landscape features and hydroclimatic variability can cause elevated river flow to increase the flux of sediment to receiving waters. Comparing records of past river discharge and associated sediment deposition patterns in lakes due to alluvial flooding can improve our understanding of the forcing of high discharge events in Waterton Lakes National Park. Four distinctive peaks in sedimentation rate observed in the LWL sediment core suggest that episodes of high discharge have previously occurred which increased the flux of sediment to the lake (Figure 5.1). Dry mass accumulation rates spiked in *ca.* 2017, 2013 (± 1.2 years), 1995 (± 2.6 years), 1961 (± 7 years), and 1948 (± 10 years). The organic matter content in the Lower Waterton Lake sediment core was extremely low compared to the content of inorganic matter. This is common in mountain catchments, where Rocky Mountain, glaciated catchments have poor soil development, limited vegetation cover, and low autochthonous production in lakes (Lamoureux and Gilbert, 2004).

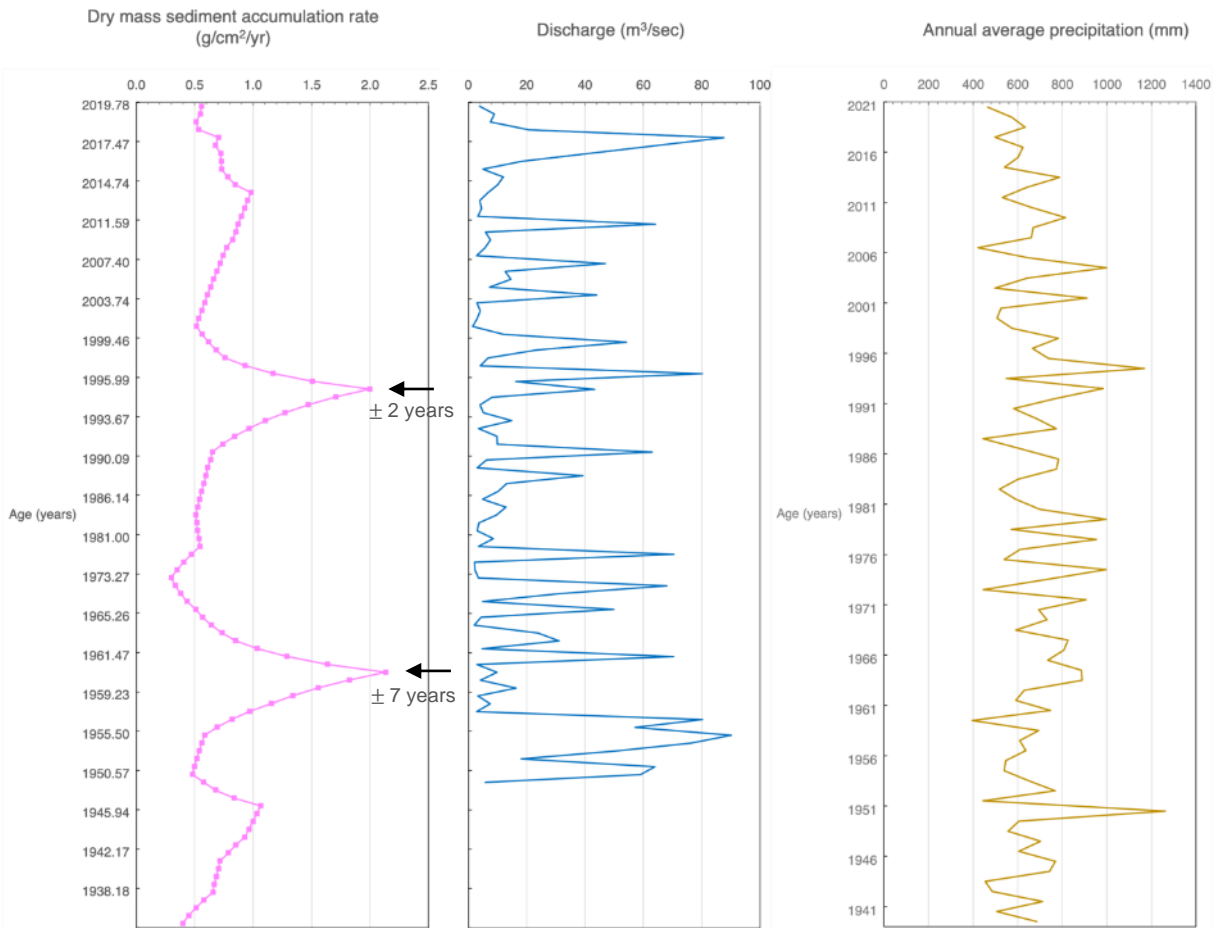


Figure 5.2 – Graph showing relations between the estimated sedimentation rate in the core from Lower Waterton Lake (1935 to 2019), the discharge records at the lake’s outflow (1935 to 2019), and average annual precipitation in Waterton Lake National Park (1940 – 2021) .

Variability in annual and seasonal precipitation patterns can drive changes in sediment erosion, transport, and accumulation (Perron, 2017; Hirschberg et al., 2021). Generally, periods with increased sedimentation rates correspond with high discharge events, which create flow conditions in rivers that exceed the critical shear stress for erosion of deposited sediment required to remobilize and transport these materials to receiving rivers and lakes (Wolman, 1989; Wren and Davidson, 2011; Myhre et al., 2019). Estimated sedimentation rate from Lower Waterton Lake and reported lake outflow discharge and precipitation are presented in Figure 5.2. The peaks in

sedimentation rate are likely due to the observed peaks in discharge rates that were measured at gauging station (station 05AD003) below Lower Waterton Lake. In the absence of historical lake inflow discharge data from Blakiston Creek, the Waterton River gauging station is used here as a proxy to identify potential high flow periods. Sedimentation rate peaks are observed generally in spring and summer seasons (Figure 5.2). This may be due to seasonal patterns in hydrological processes on the eastern slopes of the Rocky Mountains. Within areas surrounding the Rocky Mountains, spring freshet from snowmelt runoff typically occurs from March-April, with annual peak discharge occurring between May and June, before returning to baseflow conditions in July or August (Hauer et al., 1997). During the spring freshet, rivers discharge over 70% of their annual water budget and instantaneous discharges 10 to 100 times greater than mean low flow can occur (Hauer et al., 1997). The regimes of highly dynamic hydroclimatic conditions in large catchments can result in increased runoff leading to seasonal floods (Kundzewicz et al., 2008). Precipitation regimes from WLNP can inform us of wet vs dry years in the area. A possible explanation for the large increases in sedimentation in 1995 and 2013 could be the severe flood events and high annual precipitation in WLNP (Parks Canada, 2018a). Floods can create large-scale geomorphic impacts such as riverbed and bank erosion (Chambers et al., 2004), overbank sediment deposition (Asselman and Middlekoop, 1995), and channel widening (Krapesch et al., 2011). During flood events, fine sediments are prone to downstream transport, accumulating in lake basins (Hoffman et al., 2009). Previous research suggests that there is strong evidence of a link between climate change, wildfires, and projected changes in runoff, which are large enough to push peak flow beyond the range of historical behaviours (Milly et al., 2008). Additionally, changing climate in fire-prone terrain has clear implications for future landscape effects such as increased erosion, and negative implications for downstream water supplies (Meyer et al., 2001; Benda et al., 2003; Van Dijk et al., 2012).

While there are large discharge events each year in WLNP, the present work suggests that only a few are large enough to mobilize and transport sediment across the river continuum. Studies have shown that wildfires can strongly influence sedimentation rates in lakes through altered hydrologic connectivity and transport from uplands to channel network (Pompeani et al., 2020; Rhoades et al., 2019). Wildfire disturbances in LWL watersheds may be the driver that mobilizes sediment, as the reduced vegetation and hydrophobicity of ash layers can increase overland flow,

erosion, and discharge in a watershed (Smith et al., 2011). In the present work, both historical wildfire disturbance and inferred increased discharge rate, coincided with increased sedimentation rate (Figure 5.1). However, the recent data from the 2017 Kenow wildfire showed increases in sedimentation rates were somewhat delayed compared to previous hydrologic or wildfire events in drainage areas of LWL. During the Kenow wildfire, WLNP received no precipitation, followed by ~20 cm of snow at the beginning of October 2017, which restricted erosion of pyrogenic materials and sediment (Parks Canada, 2019). By the following spring (2018), WLNP had a snowpack ~75% above normal. The combination of a large snowpack, heavy June rains, and abrupt warming, resulted in rapid melting and unseasonably high spring freshet (Parks Canada, 2019). However, evidence of this high potential erosion period was not reflected in sedimentation rates measured in LWL. Additionally, in Blakiston Creek, peak particulate concentrations in the water column weren't recorded during spring of 2018, but instead two years following the Kenow wildfire, in 2019. Sediment delivery in catchments can be both delayed and reduced, by several geomorphic and hydrological processes including landscape features, climate, soil type, erodibility, and vegetation (Walling, 1983; De Vente et al., 2002). Variability in landscape attributes such as the presence of alluvial fans, lake depth, land use, and response time can have a significant influence on the magnitude of sedimentation rate (Smol, 2008).

In addition to the delayed sedimentation rate, another possible explanation for the timing of events may also be the gauging station data, as it may not be a very sensitive measure of inflow to the lake. The Waterton Lakes chain is composed of very large basins (e.g., Middle and Upper Waterton Lake, surface area of 10.1 km² and max depth of 150m) and can act as a buffer that moderates inflow to Lower Waterton Lake. The delay of delivery of sediment to LWL may point to the importance of site-specific landscape forms and their controls on transport and fate of materials in catchments. The connectivity between the tributary inflow and the main channel, will dictate whether downstream waters will be impacted following landscape disturbance (Thomas, 2001). While the other Waterton Lakes are very large basins, Lower Waterton Lake is much smaller and resembles the widening of a river. Lower Waterton Lake has an average renewal rate of 161 times per year (Anderson and Green, 1976), whereas Middle and Upper Lake have average renewal rates of 0.8 and 7.1 times per year, respectively (Anderson and Green, 1976). This fast turnover time may influence sedimentation rate, geochemical cycling, and P availability. As fine

sediment and pyrogenic materials remain suspended in flowing waters (Stone and English, 1993), the short residence time of LWL may result in the materials to quickly depart from the lake. This may provide an explanation for the differences in TPP concentrations between Blakiston Creek and LWL, as fine sediment may not all be settling in LWL when there are high discharge events (Jin and Ji, 2004). Hydrodynamics and sediment transport in shallow lakes are driven by wind and flow velocity, inducing tubulation near the benthic sediment (Jin and Ji, 2004; Smol, 2008).

Geomorphic landscape features are inherently important in the delivery and timing of sediment across the river continuum. Many sediment erosion studies at the watershed scale have highlighted the lack of continuity between upstream sediment sources and the amount of sediment delivered at the basin outlet (Walling, 1983; Phillips, 1995; Dedkov and Mozzherin, 1996) and/or the fraction stored within the basin (Kelsey et al., 1987; Wasson et al., 1996; Wilkes et al., 2019). Alluvial fans and floodplains can attenuate sediment delivery, and therefore act as large sediment sinks in watershed channel networks (Meyers and Wells, 1997; Fryirs et al., 2007; Riley and Meyer, 2015). The geomorphology of the Blakiston valley may play a significant role in sediment delivery to LWL. The large alluvial fan situated between the mouth of Blakiston Creek and LWL inflow is likely acting as a sediment trap, which increases the ingress of fine sediment into the bifurcating gravel-bed above LWL. Previous studies have shown that alluvial fans reduce sediment delivery to lakes by buffering slopes and valley floors (Phillips, 1995; Fryirs et al., 2007). Streams flowing across alluvial fans bifurcate and flow velocity is reduced due to infiltration which can limit sediment delivery to lakes (Meyer et al., 1995). It has been reported that that fire-related debris flow, pyrogenic materials, and associated sediments are deposited on alluvial fans and comprise ~30% of the volume of alluvial fan material (Meyer et al., 1992; Riley and Meyer, 2015). Low concentrations of sediment accumulation in LWL may be due to the build-up of ingressed material on the alluvial fan due to reduced flow velocities leading to increased deposition of fine sediment illustrated in Figure 5.2. Notably, these materials will be stored in alluvial fans for

decades or possibly centuries until a sufficiently high magnitude freshet or storm event can occur to mobilize, and transport deposited sediment into Lower Waterton Lake.

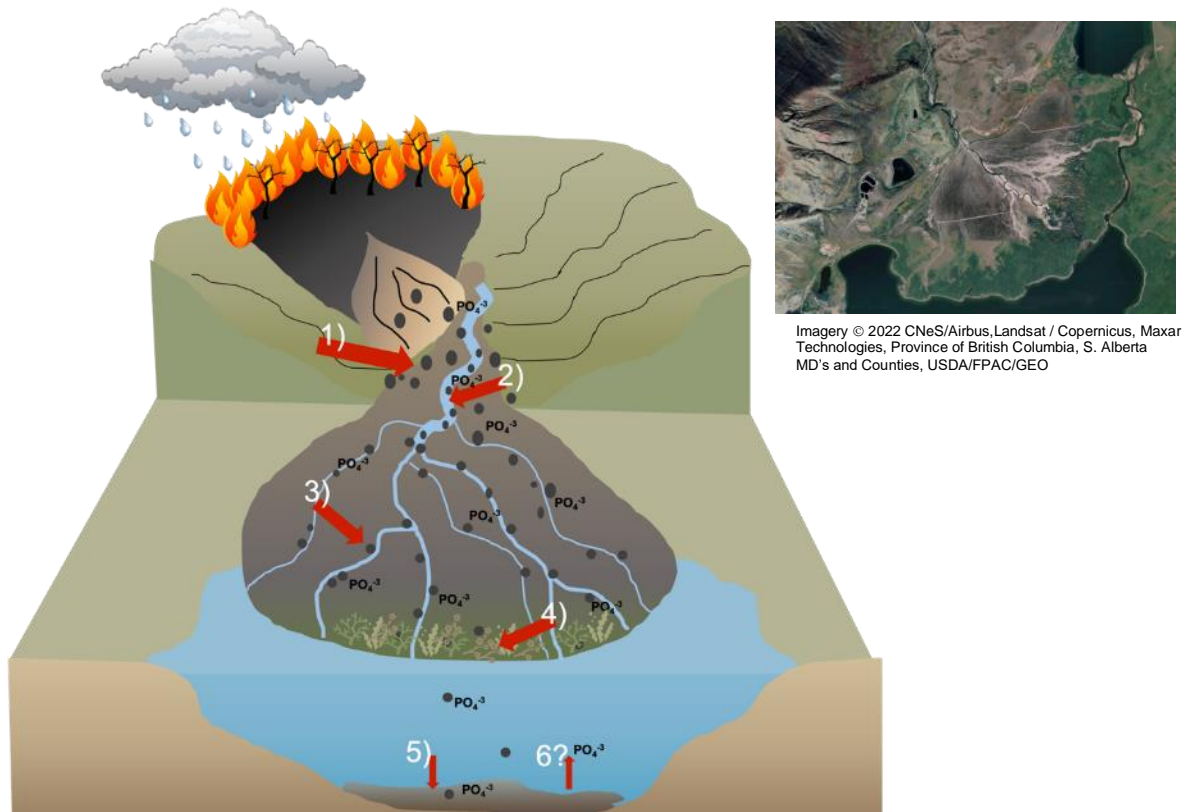


Figure 5.3 – Landsat image and conceptual diagram of post-fire sediment attenuation in the Blakiston watershed. 1) Sediment ingress into floodplain. 2) Sediment ingress into gravel-bed river. 3) Sediment attenuation on alluvial fan. 4) Attenuation of sediment by vegetation. 5) Sediment deposition into lake. 6) PO_4^{3-} potential release back into water column.

Blakiston Creek flows within a glaciated “U”-shaped valley, with an extensive floodplain that frequently floods. The presence of wide flood plains in the mid and lower sections of Blakiston Creek are important for sediment storage during flooding (Benda et al., 2003). Accordingly, the storage of sediment represents a critical source of sediment to LWL but periodicity of sediment supply is related to the occurrence of exceedingly high discharge events. Sedimentation rates in

LWL appear to substantially increase when discharge events at the lake outflow are $> 60 \text{ m}^3/\text{s}$ (Figure 5.1). Sediment storage on floodplains is a common geomorphic process and increased sedimentation rates in these alluvial features typically occur during precipitation when river discharge is elevated (Gomi et al., 2007; Brooks et al., 2010; Van Dijk et al., 2012). The compounding effects of wildfire disturbance (e.g., reduced vegetation, hydrophobic soils, and reduced infiltration rates) and large precipitation events following wildfires can cause severe flooding (Moody and Martin, 2001; Smith et al., 2011). These discharge related responses in sedimentation have been referred to as “fire-flood sequences” (Meyer and Wells, 1997; Moody and Martin, 2001; Smith et al., 2011). In 1995, there was a prescribed fire near LWL (Hay Barn Meadow, 122 ha) and a large wildfire occurred in a tributary to LWL (Sofa Mountain Wildfire, 1500 ha). In 1995, a large flood event occurred which caused extensive damage to roads and infrastructure in WLNP (Parks Canada, 2018a). The 1995 flooding and prescribed-fire disturbances are reflected in results of this study, as a large increase in sedimentation rate that followed in 1996. Evidence for similar processes in wildfire disturbed watersheds were observed following the 1996 Buffalo Creek wildfire (4690 ha, Denver, Colorado; Moody and Martin, 2001). These authors reported that sediment and pyrogenic materials were transported to and subsequently deposited on an alluvial fan. Sediment was not exported from the watershed until a subsequent extreme erosional event (Moody and Martin, 2001). Notably, fire-flood sequences have been observed predominantly in glaciated valleys (such as the Rocky Mountains) where there are observed increases in sedimentation rates following a fire-flood sequence (Meyer and Wells, 1997).

5.3 Role of benthic sediment as an internal loading source of phosphorus

While the hydrology community has made significant advances in understanding the effects of wildfire on river systems and surface water, minimal research has been conducted on lake sediment. To our knowledge, this study is the first of its kind to provide new insights regarding the dynamics of phosphorus sorption in a lake sediment core collected within large-scale wildfire landscape disturbance. Laboratory determination of EPC_0 is a useful indicator of the abiotic controls of sediment on the uptake/release of P in aquatic systems (Froelich, 1988). Larger EPC_0 values indicate an increase in the release potential of SRP to the water column due to desorption

from fine sediment, thereby increasing the availability of P for algal proliferation in the water column (Froelich, 1988; Søndergaard et al., 2003). In the present study, larger EPC_0 values coincide with historical wildfire activity and spring freshet within WLNP which is likely attributed to the fire-flood sequence. Atmospheric deposition of wet and dry pyrogenic materials may have also been deposited in Lower Waterton Lake, thus contributing to the additional P input (Tulio, 2022; Spencer and Hauer, 1991; Bodí et al., 2014). High severity burns from wildfires produce pyrogenic materials rich in phosphorus (Bodí et al., 2014). Additionally, surface runoff during flood events from grazing land adjacent to LWL, may also have contributed to increased PP in the lake. The combination of higher EPC_0 (i.e., P release potential due to desorption from fine sediment) and elevated bioavailable NAIP post-wildfire may lead to algal proliferation, decreasing lake water quality (Emelko et al., 2016; Miller and Yates, 2006; Silins et al., 2014; Smith et al., 2011). The increase in P mobility in oligotrophic lakes following a wildfire can have cascading, long-term legacy impacts on internal bioavailable P loading (Emelko et al., 2016; Stone et al., 2011).

To date, most studies of phosphorus dynamics in lakes have focused primarily on examining the effects of agriculture on lake sediment composition and its impact on the nutrient status, whereas the impact of wildfire is beginning to receive more recent attention in the literature (McCullough et al., 2019). Phosphorus sorption dynamics have previously been reported in lakes where anthropogenic disturbances (agriculture and WWTPs) are present (Table 2.1). EPC_0 values reported in the literature are highly variable and differ across landscape disturbances and aquatic systems. Eutrophic lakes affected by agriculture typically have max EPC_0 values $>120 \mu\text{g P L}^{-1}$ (Belmont et al., 2009; Krogerus and Ekholm, 2003), and solids in WWTP effluent have higher EPC_0 max values ($>240 \mu\text{g P L}^{-1}$; Yin et al., 2016; Zhu and Yang, 2018). In rivers affected by wildfire, max EPC_0 values were reported to range from $207 \mu\text{g P L}^{-1}$ (Tulio, 2022) and $1200 \mu\text{g P L}^{-1}$ (Son et al., 2015), and bed sediment in rivers impacted by multiple land disturbances, such as wildfire and harvesting, have a maximum EPC_0 of $579 \mu\text{g P L}^{-1}$ (Watt et al., 2021). Indeed, these EPC_0 values are within the same boundaries of systems that are typically eutrophic and experience reduced water quality (Belmont et al., 2009; Zhu and Yang, 2018; Son et al., 2015). Following the Kenow Wildfire, suspended solids in Blakiston Creek had EPC_0 max values of $\sim 1856 \mu\text{g P L}^{-1}$ (Tulio, 2022). Indeed, this is extremely high and may point to the detrimental nature of wildfire

and pyrogenic materials on river systems. However, max EPC_0 in benthic sediment of LWL was $23 \mu\text{g P L}^{-1}$ following the Kenow Wildfire. The large discrepancy between EPC_0 values of suspended solids from Blakiston Creek and benthic sediment from LWL strongly point to the potential of some landscape features to attenuate sediment delivery to lake systems. The cascading effects of geomorphic landscape features on sediment storage in the river continuum which include storage in flood plains, gravel bed rivers and alluvial fans is illustrated in Figure 5.3. While the connectivity between hillslope and rivers in the headwater tributaries of the Blakiston Creek watershed is high, in lower reaches of the river the connectivity between hillslope and stream is much lower due to the presence of wide floodplains which attenuate sediment delivery from hillslopes to receiving streams. Sediment transport from Blakiston Creek to the Lower Waterton Lake is further reduced by sediment ingress in gravel beds of the river and subsequent deposition as the river bifurcates and loses energy on the alluvial fan. Following the Kenow Wildfire, the increased potential of sediment delivery was not realized because post fire rainfall was negligible. Additionally, EPC_0 differences between river and lakes may point to the complexity of EPC_0 in aquatic systems, as it is a function of physical, chemical, and environmental process. This includes particle size (Stone and English, 1993), mineralogy (Whithers and Jarvie, 2008), pH, temperature, residence time of sediment, ionic strength, sediment to solution ratios and solution chemistry (Bhadha et al., 2012; Lucci et al., 2010). The addition of pyrogenic materials also has the potential to alter the geochemistry of lakes, as ash can increase pH when in solution (Bodí et al., 2014). Increases in pH can increase EPC_0 , pointing towards the potential of ash to alter lake biogeochemical P cycling (Koski-Vähälä and Harikainen, 2001). Wildfire is increasing in extent and severity worldwide, which could have long-term implications for lakes. The addition of limiting nutrients and pyrogenic materials to the water column may potentially change geochemical cycling processes in lakes resulting in eutrophication (Emelko et al., 2016; Mast et al., 2008).

Within a watershed, P dynamics may undergo change due to differing sediment supply from hillslope, to receiving stream, to river, and into lake (Bowes et al., 2003). The potential for sediment-associated P to desorb into the water column varies greatly throughout the sediment core. The main drivers of the uptake and desorption of P in lake sediment include co-precipitation with Al, and Fe hydroxides and Ca compounds, and assimilation by aquatic plants, bacteria, and biofilms (Mainstone and Parr, 2002). This study did not measure environmental factors such as

redox conditions, alkalinity, temperature, and pH, which can have strong influence on P mobility (Nurnberg 1984; Chen et al., 2019). However, it has been reported that EPC_0 increases due to high pH and low redox potential and may result in changes in sorption potential (Koski-Vähälä and Harikainen, 2001). To characterize changes more fully in lake nutrient dynamics in response to the loading of post-wildfire materials, it is recommended that other parameters such as pH, redox potential, Ca, Fe and Mn should be measured directly in the lake. These parameters would help to provide a better understanding of mechanisms that control P dynamics between sediment and overlying water (House and Dennison, 2000).

5.4 Implications for drinking water source protection and nutrient storage

The effects of varying landscape disturbance on lake surface water quality has been widely reported (Carpenter et al., 1998; Nurnberg 2009), however much less is known about the effects of wildfire on lake P dynamics (McCullough et al., 2019). Landscape disturbance can degrade the quality of surface water and challenge the provision of drinking water unless new treatment infrastructure is incorporated into the water treatment plant. The combined effects of elevated NAIP and higher EPC_0 from fine sediments after a wildfire may result in deteriorated water quality and changes in the trophic state of receiving waters such as reservoirs (Emelko et al., 2011). However, it is difficult to disentangle the effects of wildfire on lake systems, due to the heterogeneous nature of watersheds, hydro-climatic variability, and both natural and anthropogenic disturbances that occur at both spatial and temporal scales. Understanding the variability in distribution of PP forms in lake sediment is crucial for long-term water quality in wildfire-threatened catchments.

Wildfires can have a long-lasting effect on lake physical and chemical properties, with direct responses from water quality and lake biota due to legacy P loading. The increase in fine sediment associated P loading to lakes following a wildfire has a potential to increase the internal cycling of nutrients (McCullough et al., 2019, Søndergaard et al., 2003; Silins et al., 2009). As Lower Waterton Lake has a short residence time and relatively high sediment surface contact with overlying water, it is likely that there may be a more intense, but brief, response to increased PP following the Kenow Wildfire. As smaller lakes with short residence times have a larger probability of having the polluted water discharged, larger lakes may experience the effects of the

pyrogenic biogeochemistry on the trophic structure. Legacy P loading following a wildfire is a key concern for water quality manager (Emelko et al., 2016), as phosphorus can be released from sediment depths as low as 20cm, and can last for decades (Søndergaard et al., 2003). This demonstrates the importance of studying sediment cores, as it is seen that there is high potential for larger amounts of P to be released from below the sediment-water interface.

Climate change is increasing fire severity and extent in forested watersheds, diverging from historical fire regimes globally. The combined effects of climate change and landscape disturbance has been shown to increasingly alter fire regimes in watersheds. This is also true for watersheds which have experienced a “fire deficit”, as they will be more likely to experience higher extensive burns due to fire-exclusion practices (Parks et al., 2015, Halofsky et al., 2020). Climate change also alters the precipitation trends in watersheds, with less frequent, but more severe rainfall events in some regions, allowing for fuel drying during the summer months and higher erosion potential during storm events (Halofsky et al., 2020). These large storm events can reach the critical shear stress necessary to mobilize and transport particulate matter which has been deposited on alluvial fan and ingressed into gravel-bed rivers and be deposited into lakes. Severe floods due to climate change can alter flow dynamics in tributaries to become non-stationary (Winkler, 2002). The fire-flood sequence can result in large increases of sediment and pyrogenic materials to downstream lakes (Meyers and Wells, 1997). Additionally, lakes can act as a natural fuel break for fire, influencing fire behaviour and extent, consequently becoming subject to direct wildfire effects.

Large wildfires have made media headlines in recent year, pointing to their destruction and endangerment of communities (Radeloff et al., 2018; Schoennagel et al., 2017), yet these headlines overlook the importance of sustaining critical ecosystem services (e.g., drinking water, recreation, erosion control), provided by these endangered watersheds. Recently, there has been more research focused on the implications for ecological communities (Silins et al., 2014), and downstream water supplies (Emelko et al., 2014). Decision from land-use managers directly influence downstream water quality, demonstrating that land, water, and fire should be managed and studied jointly in fire-prone watersheds (McCullough et al., 2014). Typical management treatments such as fuels reduction and prescribed fire in critical forested watersheds can reduce the potential for wildfires in the future, however the transport and fate of sediment in watersheds should not be overlooked. Understanding the unique geomorphology of landscape forms can provide confidence in critical

watersheds for greater resilience of communities. For example, the alluvial fan present at the foot of Blakiston Creek is likely acting as a sediment trap for pyrogenic material and PP, reducing the impact wildfire disturbance has on P dynamic in LWL. Studying freshwater systems, such as those of Waterton Lakes National Park, is critical in enhancing our understanding of wildfire and landscape form, on the influence of critical source water supplies (Hauer et al., 1997; Watt et al., 2021).

5.5 Future Research

The heterogenous nature of watersheds, makes it difficult to detangle the effects of large-scale landscape disturbances. Hydro-climatic variability, natural and anthropogenic disturbances, and geomorphic landscape features, play into watersheds response following a wildfire at both spatial and temporal scales. While this research provided knowledge to fill some critical gaps in wildfire disturbance on lakes (McCollough et al., 2019), it also demonstrated the need for further research. The present thesis examined the vertical distribution of P forms and mobility in a sediment core, as well as the analysis of historical landscape disturbances and lake response. However, sedimentation rates greatly fluctuated with historical wildfire disturbance and flood events, which may be a role of specific landscape forms and function, such as wide flood plains and alluvial fans. As a result, future research should be cautious that point sampling could lead to inaccurate assessments of landscape disturbances. Future research should explore the role of alluvial fans in source watersheds, and their role in attenuating sediment and pyrogenic materials following wildfire disturbance. A series of piezometers should be installed along the downstream continuum of the alluvial fan to measure the hydraulic head, water chemistry, and to measure how the amount of water which ends up in the connecting channel. In addition, in oligotrophic systems, characterizing SRP alone may not be sufficient to describe disturbance impacts on aquatic systems, as SRP released from aquatic sediment can be quickly consumed by primary producers (Emelko et al., 2016). To understand P solubility, sediment geochemistry should be related to surrounding aqueous chemistry. This study did not measure environmental factors such as redox conditions, alkalinity, temperature, and pH, which can have strong influence on P mobility (Nurnberg 1984; Chen et al., 2019). It is recommended that EPC₀ experiments are supported by supplementary data

of pH, redox potential, and concentrations of dissolved Ca, Fe and Mn, and that these parameters would be measured directly in the system (House and Dennison, 2000).

5.6 Conclusions

The primary goal of this work was to examine the vertical distribution of particulate phosphorus forms (NAIP, AP, OP) and P mobility (EPC_0) in benthic sediment of an oligotrophic lake affected by severe wildfire in Waterton Lakes National Park, AB. The conclusions of this study are:

1. Sedimentation rates in Lower Waterton Lake were highly variable and attributed to the interplay between historical flood events and landscape features that alter the storage, mobility, and delivery of sediment to the lake.
2. Geomorphic landscape features, such as Blakiston alluvial fan and large floodplain, most likely attenuate delivery of sediment and pyrogenic materials Lower Waterton Lake, causing a lag in sediment delivery which may last over decades or centuries.
3. Materials deposited on the alluvial fan may remain ingressed until a large-scale flood event propagates sediments downstream, indicating a fire-flood sequence.
4. The geochemical composition of sediment in the core varied slightly as a function of depth and suggests that inputs of sediment to the lake have a relatively uniform composition.
5. Apatite phosphorus was the predominant form of PP in the core which corresponds to sediment geochemistry of surface materials consisting of this previously glaciated landscape.
6. TPP was positively correlated with Fe and Mn, and negatively correlated with Al, Ca, and K, which could be a function of pyrogenic material chemistry.
7. EPC_0 in sections of the lake sediment core were variable and corresponded with historical landscape disturbances (e.g., wildfire, flooding), demonstrating the increase in potential of SRP to the overlying water column during periods of disturbance.

Chapter 6

Synthesis of Conclusions

In forested landscapes, wildfire is the dominating natural landscape disturbance, such as those of the eastern slopes of the Rocky Mountains (Weber and Flannigan, 1997). Wildfires have been increasing in extent and severity due to the current trend of climate warming (Flannigan et al., 2005), leading to more catastrophic “mega-fires” (Williams, 2013). The effect of severe wildfire on terrestrial and aquatic ecosystems can threaten global water security through biological, chemical, and physical stressors (Moody et al., 2013;). Wildfire disturbance can increase erosion rates due to vegetation loss and has been shown to increase the rates of sediment erosion from hillslopes to receiving streams (Emelko et al., 2016; Wondzell and King, 2003). The increase of erosion of sediment, vegetative materials, and nutrients is raising concern for the degradation of water quality and implications to fresh drinking water (Emelko et al., 2016). The downstream propagation of fine sediment to lakes or reservoirs is a global problem, which can result in eutrophication, pollution, and biodiversity loss (Owens et al., 2005; Vörösmarty et al., 2003). Recent attention has been directed towards better understanding the short term and legacy effects of wildfire and sediment pressures in freshwater systems (Tulio, 2022; Emelko et al., 2016; Silins et al., 2014) and their effects on water supply (Emelko and Sham, 2014).

Critical forested source water regions in Alberta located on the Eastern slopes of the Rocky Mountains are under increasing pressure from largescale-wildfire disturbance. In 2017, the high severity Kenow Wildfire contiguously burned 35,000 ha (>95%) of Waterton Lakes National Park. Wildfire increases erosion potential of fine sediment, pyrogenic materials, and nutrients. The input of limiting nutrients, such as phosphorus, in oligotrophic systems, can increase primary productivity and is the main driver of eutrophication (Correll, 1998). Eutrophication is a primary concern for water quality managers, as it can result in taste and odor problems (Wnorowski, 1992) and impair treatment processes (Bernhardt, 1984), resulting in incremental cost increases for water treatment operations (e.g., increased chemical consumption and improved infrastructure). Due to the magnitude and speed of global climate change, strategies to protect critical source water areas are necessary to adapt to the water challenges induced by climate change (Emelko et al., 2016).

Fine sediments are the primary vector for P transport in aquatic systems (Kronvang et al., 1997). Particulate phosphorus (PP) forms dominate the majority of P entering aquatic systems from overland runoff. Physical, chemical, and biological factors play into the forms (NAIP, AP, OP) and the mobility (EPC_0) of PP in aquatic systems (Reddy et al., 1999; House, 2003). The particle size of sediment is an important factor in freshwater lakes, as fine sediment ($< 63 \mu\text{m}$) preferentially binds to P, due to its large surface area and metal-oxide coatings (Stone and English, 1993). Notably, fine sediment can influence P cycling and productivity in lakes, adsorbing and releasing SRP into the water column (Whithers and Jarvie, 2008). Within the current literature on P fractionation in freshwater lakes (Pettersson et al., 1988; Kerr et al., 2011; Emelko et al., 2016; Knezic, 2021; White and Stone, 1996; Messner et al., 1984; Łukawska-Matuszewska et al., 2013), and P mobility (Spears et al., 2007; Cyr et al., 2009; Yin et al., 2016; Belmont et al., 2009; Krogerus and Ekholm, 2003; Krogerus and Ekholm, 2003; James, 2017; Khanal et al., 2021; Shoja et al., 2017; Łukawska-Matuszewska and Xie, 2013; Zhu and Yang, 2018), studies have focused primarily on anthropogenic landscape disturbances, such as forestry, agriculture, and WWTPs. Results from these studies have varied but are clear that sedimentation contamination in freshwater lakes can result in a reduction in water quality, lasting for decades. There is limited knowledge about natural large-scale landscape disturbances, such as wildfire, on freshwater lake P dynamics. This study aimed to provide insight into the knowledge gaps surrounding wildfire disturbance on freshwater lake P-dynamics, and the legacy effect wildfires may cause in lake P mobility. Lower Waterton Lake sedimentation rate varied greatly from 2019 to ~1930. Spikes in sedimentation peaked around ~2013, ~1995, ~1961, and ~1948, likely correspond to large flood events with periods of high discharge in WLNP during unusually high spring flood events (Parks Canada, 2019). We hypothesized that sediment transport is attenuated by the Blakiston alluvial fan, until a large flood event remobilizes and deposits sediment into Lower Waterton Lake. Additionally, the fire-flood sequence is demonstrated in this study pointing to the importance of wildfire in generating runoff and sediment transport in mountain drainage basins (Meyer and Wells, 1997).

Sediment-associated P forms in Lower Waterton Lake remained relatively consistent, with AP dominating TPP fractions concentrations, notably 61% ($360.2 \mu\text{g P g}^{-1}$) on average, followed by NAIP (26%; $153.5 \mu\text{g P g}^{-1}$), and OP at 12% ($75.5 \mu\text{g P g}^{-1}$). The bioavailable fractions of TPP in Lower Waterton Lake are minimal, and do not contain a substantial amount of potentially

bioavailable phosphorus which could be released into the water column. This may be due to the sediment-trapping, as fine-grained sediment is more likely to ingress into the riverbed during low velocity flow over the large Blakiston alluvial fan, reducing the amount of fine sediment input into the lake (Stone and English, 1993). Further research should be conducted in a system where “sediment-trapping” would not be present. This would allow sediment cores to potentially detect fine sediment and contaminate input to the lake shortly following wildfire disturbance. While the results of TPP concentrations did not have significant increases following wildfire, the concentrations remained stable and demonstrates the ability of P to have a legacy effect in lake systems where alluvial fans are present.

Geology of watersheds are inherently important on influencing P form and mobility in the water column (Froelich, 1988). For example, the dominating NAIP species in Lower Waterton Lake is BD-RP and may point to the ubiquity of Mn and Fe-oxides within WLNP such as the presence of the Fe-rich argillite, in Red Rock Canyon. Although it was expected that Mn concentrations would be elevated post-wildfire (due to ash and pyrogenic contamination of surface water), no significant trends were observed in Lower Waterton Lake (except for a flux at the top 0.5cm of the sediment core). The absence of these relationships may point to the long-term effect of pyrogenic-derived materials within wildfire affected systems (Bodí et al., 2014). Unlike Mn, Ca has strong temporal variability in the sediment core. Ca is generally considered to be a relatively stable in sediment, however calcium carbonate (CaCO_3) is the dominating constituent of ash, pointing to evidence of the historical fire regimes in the area (Bodí et al., 2017; Kozerski & Kleeberg, 1998). Future studies should choose their sample sites carefully, paying particular attention to the surrounding landscape in attempt to prevent “sediment-trapping” above the collection point.

To our knowledge this study is the first of its kind to make progress in understanding the dynamics of phosphorus sorption in a lake sediment core collected within large-scale wildfire landscape disturbance. Phosphorus isotherm experiments, such as those conducted by Froelich (1988), are commonly used to determine P sorption dynamics. EPC_0 is a useful tool to quantify abiotic controls on the uptake or release of PP in freshwater sediment (Froelich, 1988). Larger EPC_0 values increase the range at which internal loading of phosphorus from the sediment to the water column occurs, thereby increasing the availability of this limiting nutrient for algal

proliferation in freshwater (Froelich, 1988; Søndergaard et al., 2003). In this study, the sorption characteristic of sediment from Lower Waterton Lake, were analyzed in a temporal perspective. In the present study, EPC_0 peaks generally coincided (within ~2 years) of wildfire disturbance and flood events. These increases in EPC_0 were likely a function of atmospheric deposition of wet and dry pyrogenic materials (e.g., ash) which may be deposited into Lower Waterton Lake. Wildfires burned with high severity (such as the Kenow wildfire), produce ash of high combustion completeness which is P-rich (Bodí et al., 2014). Historical wildfire activity also coincides with increases in EPC_0 , pointing to the cascading, long-term legacy impacts wildfire has on internal bioavailable P loading (Emelko et al., 2016; Stone et al., 2011). The peak concentrations in EPC_0 also corresponded to elevated SRP concentrations following the Kenow wildfire. All analyzed sediment intervals demonstrated the ability to release SRP to the overlying water column, pointing to the importance in understanding long-term implications wildfires have on critical source water. Future work should measure all biological and geochemical factors in the study site which can influence P sorption dynamics, to support more accurate sorption experiments.

The transport and fate of sediment and pyrogenic materials is very important in determining potential effects of wildfire in watersheds. This study was after the work of Tullio (2022), analyzing the burned rivers (Blakiston Creek and Cameron Creek) draining into Lower Waterton Lake. While this study observed temporal changes in P form and mobility from ~1930 to 2020, Tullio analyzed P form and mobility from 2018 – 2020 in the gravel bed rivers. In 2019, TPP in Blakiston Creek had a max concentration of $950 \mu\text{g P g}^{-1} \text{ dw}$ following the Kenow wildfire (Tullio, 2020), whereas TPP in Lower Waterton Lake reached a max of $725 \mu\text{g P g}^{-1} \text{ dw}$. While many factors can alter the concentration of TPP in sediment, we observed that geomorphic landscape effects in the watershed have dominated the delivery of sediment to LWL. The Blakiston alluvial fan is located at the mouth of Blakiston Creek and separates Blakiston Creek from LWL. Our analysis of sedimentation rate in LWL show that large discharge events are required to mobilize sediment attenuated on the alluvial fan, to be deposited into LWL. While fine sediment typically has a low settling velocity, the large flood plain and alluvial fan can reduce flow rate, allowing infiltration of water and the settling of materials. The role of the Blakiston alluvial fan in this system is further demonstrated by the difference in EPC_0 values from Blakiston Creek and LWL. While Blakiston Creek experienced max EPC_0 of $1792 \mu\text{g P L}^{-1}$ following the Kenow wildfire

(Tullio, 2022), LWL only had max EPC_0 of $18.5 \mu\text{g P L}^{-1}$ after the fire. Many conditions are required to load lakes in watersheds such as hillslope connectivity, spatial and temporal variability, and non-stationarity. The difference in P concentration and mobility between the upstream reached and downstream lake separated by an alluvial fan, demonstrate the geomorphic effects of forested watersheds in the Rocky Mountains, and the potential resilience alluvial fan provide from large scale landscape disturbance.

The present thesis examined the vertical distribution of PP forms and P mobility in an oligotrophic lake affected by severe wildfire disturbance. While this research was the first of its kind and provided knowledge to fill critical research gaps, further research is required. For instance, the present work demonstrated the influence and legacy effect geomorphic landscape features can have on lake systems, however further research should be cautious in their sampling location, as site selection is imperative in accurate assessment of landscape disturbance. Further work should include lakes over a variety of landscapes with differing degrees of historical wildfire activity to detangle the complex nature of watersheds, including sampling of large geomorphic forms. Additionally, studies examining the potential for pyrogenic materials to proliferate the growth of cyanobacteria in lakes, would inform post-fire water quality and water treatability impacts and associated management decisions.

Wildfires can have a long-lasting effect on lake physical and chemical properties, with direct responses from water quality and lake biota mainly legacy phosphorus loading. Lakes in Waterton Lakes National Park are nearly pristine, and the use of sediment cores allow their response from large-scale landscape disturbances to be studied. The management of freshwater supplies are becoming increasingly difficult with the pressure of climate change, which is of strategic and national importance (Emelko et al., 2016). Findings from this work will enable and inform policy, technology, and innovation to communities reliant on critical forested watersheds as drinking water sources (Emelko and Sham, 2014). The assessment of legacy wildfire effects and bioavailable phosphorus dynamics in freshwater lake, is critical to informing source water protection (SWP) strategies, reservoir management strategies, and public health risks.

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Appendices

Appendix A: Supplementary Information and Data

Supplementary Data

Table A1 - Major elements for benthic sediment intervals collected from Lower Waterton Lake.

Date	Depth	Al	Fe	Mn	Mg	Ca	Na	K	P ₂ O ₅
Years	cm	% dry mass							
2019.79	00.0-0.5	2.03	2.79	0.157	2.49	2.27	0.025	0.41	0.17
2019.36	00.5-1.0	1.96	2.33	0.0495	2.17	1.89	0.023	0.41	0.17
2019.01	01.0-1.5	2.19	2.66	0.0594	2.75	2.44	0.026	0.5	0.16
2018.52	01.5-2.0	2.25	2.59	0.057	2.85	2.71	0.025	0.52	0.15
2018.03	02.0-2.5	2.22	2.63	0.0624	2.82	2.88	0.029	0.57	0.16
2017.59	02.5-3.0	2.06	2.31	0.0585	2.83	2.91	0.025	0.51	0.14
2017.15	03.0-3.5	2.35	2.2	0.0677	2.9	3.6	0.022	0.55	0.15
2016.72	03.5-4.0	2.24	2.26	0.0681	3.25	3.92	0.025	0.57	0.13
2016.2	04.0-4.5	2.16	2.2	0.0677	3.26	4.21	0.024	0.5	0.13
2015.67	04.5-5.0	2.34	2.26	0.0727	3.2	4.41	0.026	0.56	0.14
2014.53	05.0-6.0	2.24	2.05	0.0714	3.1	4.48	0.024	0.56	0.13
2013.34	06.0-6.5	2.11	2.17	0.0683	3.14	4.56	0.025	0.56	0.12
2012.84	06.5-7.0	2.37	2.26	0.0682	2.92	3.84	0.027	0.61	0.13
2012.33	07.0-7.5	2.38	2.29	0.0722	3.19	3.94	0.027	0.62	0.14
2011.71	07.5-8.0	2.57	2.39	0.073	3.16	3.55	0.026	0.63	0.12
2011.05	08.0-8.5	2.64	2.37	0.0729	2.98	3.36	0.025	0.67	0.14
2010.36	08.5-9.0	2.54	2.61	0.0777	2.98	3.8	0.024	0.67	0.15
2009.18	09.0-9.5	2.44	2.46	0.0867	2.78	4.43	0.023	0.6	0.14
2007.21	09.5-10.0	2.28	2.42	0.0858	2.95	4.47	0.027	0.63	0.15
2006.54	10.0-10.5	2.38	2.33	0.0806	2.64	3.89	0.024	0.58	0.15
2000.35	15.0-15.5	1.9	2.23	0.0632	2.56	3.91	0.027	0.5	0.16

1993.15	20.0-20.5	2.14	2.22	0.0613	2.57	3.01	0.025	0.51	0.14
1985.73	25.0-25.5	2.2	2.42	0.0889	3.24	4.61	0.026	0.5	0.15
1973.2	30.0-30.5	2.1	2.34	0.0639	2.53	2.87	0.028	0.47	0.14
1961.99	35.0-35.5	1.99	2.35	0.0742	2.39	3.26	0.026	0.53	0.15
1956.38	40.0-40.5	2.18	2.34	0.0669	2.91	3.96	0.029	0.51	0.13
1947.99	45.0-45.5	1.92	2.21	0.0509	2.46	2.79	0.028	0.49	0.14
1940.67	50.0-50.5	2.03	2.79	0.157	2.49	2.27	0.025	0.41	0.17

Table A2 – Temporal distribution of mean (\pm standard deviation) of PP forms ($\mu\text{g/g}_{\text{sediment}}$) in suspended and bed sediment fractions from Lower Waterton Lake.

Date	Depth	NH ₄ Cl-RP	BD-RP	NaOH-RP	Total NAIP	AP	OP	TPP
Years	cm	$\mu\text{g/g}_{\text{sediment}}$						
2019.79	00.0-0.5	< 10	110	94.0	214.0	414	97	725
2019.36	00.5-1.0	< 10	102	83.3	195.3	415	82.7	693
2019.01	01.0-1.5	< 10	79.3	74.6	163.9	412	90.6	666.5
2018.52	01.5-2.0	< 10	85.5	68.8	164.3	419	88.4	671.7
2018.03	02.0-2.5	< 10	44.2	58.5	112.7	416	96	624.7
2017.59	02.5-3.0	< 10	59.6	52.7	122.3	399	92.3	613.6
2017.15	03.0-3.5	< 10	47.3	44.8	102.1	392	92.4	586.5
2016.72	03.5-4.0	< 10	32.7	41.1	83.8	418	88.3	590.1
2016.2	04.0-4.5	< 10	35.8	41.6	87.4	403	96.3	586.7
2015.67	04.5-5.0	< 10	32.9	38.0	80.9	403	87.7	571.6
2014.53	05.0-6.0	< 10	31.1	35.5	76.6	403	85.7	565.3
2013.34	06.0-6.5	< 10	34.4	38.3	82.7	396	85.4	564.1

2012.84	06.5-7.0	< 10	110	42.5	162.5	305	60.9	528.4
2012.33	07.0-7.5	< 10	110	43.2	163.2	317	61.1	541.3
2011.71	07.5-8.0	< 10	109	46.2	165.2	284	64.7	513.9
2011.05	08.0-8.5	< 10	94.1	45.5	149.6	273	70.6	493.2
2010.36	08.5-9.0	< 10	108	49.2	167.2	279	70.3	516.5
2009.18	09.0-9.5	< 10	118	57.3	185.3	308	84	577.3
2007.21	09.5-10.0	< 10	127	69.0	206.0	319	80.9	605.9
2006.54	10.0-10.5	< 10	116	66.3	192.3	301	82.8	576.1
2000.35	15.0-15.5	< 10	107	46.6	163.6	384	53.4	601
1993.15	20.0-20.5	< 10	120	72.4	202.4	330	59.6	592
1985.73	25.0-25.5	< 10	97.1	43.7	150.8	368	52.1	570.9
1973.2	30.0-30.5	< 10	114	64.5	188.5	331	72.9	592.4
1961.99	35.0-35.5	< 10	105	52.0	167.0	361	48.6	576.6
1956.38	40.0-40.5	< 10	120	76.5	206.5	351	73.3	630.8
1947.99	45.0-45.5	< 10	113	53.6	176.6	352	48.7	577.3
1940.67	50.0-50.5	< 10	110	44.7	164.7	333	47.6	545.3

Table A3 – NAIP enrichment factors from sediment intervals in Lower Waterton Lake.

Date	Depth	NAIP/TPP
Years	cm	$\mu\text{g}/\text{g}_{\text{sediment}}$
2019.79	00.0-0.5	0.295
2019.36	00.5-1.0	0.282
2019.01	01.0-1.5	0.246
2018.52	01.5-2.0	0.245
2018.03	02.0-2.5	0.180
2017.59	02.5-3.0	0.199
2017.15	03.0-3.5	0.174
2016.72	03.5-4.0	0.142

2016.2	04.0-4.5	0.149
2015.67	04.5-5.0	0.142
2014.53	05.0-6.0	0.136
2013.34	06.0-6.5	0.147
2012.84	06.5-7.0	0.308
2012.33	07.0-7.5	0.301
2011.71	07.5-8.0	0.321
2011.05	08.0-8.5	0.303
2010.36	08.5-9.0	0.324
2009.18	09.0-9.5	0.321
2007.21	09.5-10.0	0.340
2006.54	10.0-10.5	0.334
2000.35	15.0-15.5	0.272
1993.15	20.0-20.5	0.342
1985.73	25.0-25.5	0.264
1973.2	30.0-30.5	0.318
1961.99	35.0-35.5	0.290
1956.38	40.0-40.5	0.327
1947.99	45.0-45.5	0.306
1940.67	50.0-50.5	0.302

Table A4 - Sorption isotherm linear equations and EPC_0 concentrations ($\mu\text{g L}^{-1}$) for suspended and bed sediment from Lower Waterton Lake

Date (years)	Depth (cm)	y-Intercept	EPC_0 ($\mu\text{g P L}^{-1}$)	R^2	Slope
2019.79	00.0-0.5	-0.52	5.2	0.99	$y = 0.0987x - 0.5152$
2019.36	00.5-1.0	-1.64	18.5	0.99	$y = 0.0887x - 1.6422$
2019.01	01.0-1.5	-0.6	7.0	0.99	$y = 0.0867x - 0.6043$
2018.52	01.5-2.0	-0.31	3.0	1.00	$y = 0.1038x - 0.3094$

2018.03	02.0-2.5	-1.43	15.1	0.99	$y = 0.095x - 1.433$
2017.59	02.5-3.0	-2.68	22.8	0.99	$y = 0.0967x - 2.6835$
2017.15	03.0-3.5	0.18	5.2	1.00	$y = 0.1027x + 0.1815$
2016.72	03.5-4.0	-1.44	14.6	0.99	$y = 0.0984x - 1.4378$
2016.2	04.0-4.5	-0.61	6.2	0.99	$y = 0.0978x - 0.6074$
2015.67	04.5-5.0	-0.64	6.9	0.99	$y = 0.0926x - 0.6353$
2014.53	05.0-6.0	-0.13	1.3	0.99	$y = 0.1001x - 0.1309$
2013.34	06.0-6.5	-0.31	3.1	0.99	$y = 0.1x - 0.3073$
2012.84	06.5-7.0	-0.41	4.3	0.99	$y = 0.0974x - 0.4144$
2012.33	07.0-7.5	-0.68	7.1	0.99	$y = 0.0955x - 0.682$
2011.71	07.5-8.0	-0.92	9.7	0.99	$y = 0.0975x - 0.9228$
2011.05	08.0-8.5	-1.00	10.5	1.00	$y = 0.0953x - 1.0023$
2010.36	08.5-9.0	-0.71	7.4	0.99	$y = 0.0958x - 0.7105$
2009.18	09.0-9.5	-1.54	14.6	0.99	$y = 0.105x - 1.536$
2007.21	09.5-10.0	-2.06	21.3	0.99	$y = 0.097x - 2.0631$
2006.54	10.0-10.5	-1.73	18.2	0.99	$y = 0.0953x - 1.7329$
2000.35	15.0-15.5	-2.00	20.3	0.99	$y = 0.098x - 1.9862$
1994.28	20.0-20.5	-0.95	10.1	0.99	$y = 0.0938x - 0.9489$
1986.88	25.0-25.5	-0.71	7.5	0.99	$y = 0.0948x - 0.7127$
1975.20	30.0-30.5	-2.00	7.7	0.99	$y = 0.095x - 1.9993$
1961.97	35.0-35.5	-0.65	6.6	0.99	$y = 0.0977x - 0.6475$
1956.53	40.0-40.5	-0.75	7.7	1.00	$y = 0.097x - 0.7479$
1947.10	45.0-45.5	-0.35	3.7	1.00	$y = 0.0959x - 0.3549$
1940.83	50.0-50.5	-1.07	11.8	0.99	$y = 0.0904x - 1.0671$

Appendix B: Quality Control/Quality Assurance

Phosphorus Speciation QA/QC

Table B1 – Particulate phosphorus speciation duplicate analyses from Act Labs.

Analyte Symbol	AP	OP	NAIP	NAIP	NAIP
Unit Symbol	ppm	ppm	ppm	ppm	ppm
Detection Limit	0.02	0.02	0.02	0.02	0.02
Analysis Method	HCL-RP	Refractory P	BD-RP	NH4CL-RP	NaOH-RP
S15 Orig	28.4	64	109	< 10	45.4
S15 Dup	28.4	65.5	108	< 10	46.9

Table B2 – Major element sediment geochemistry duplicate analyses from Act Labs.

Analyte Symbol	Al	Fe	Mg	Mn	Ca	Na	K	P ₂ O ₅
Unit Symbol	%	%	%	ppm	%	%	%	%
Detection Limit	0.01	0.01	0.01	1	0.01	0.001	0.01	0.001
Analysis Method	AR-MS	AR-MS	AR-MS	AR-MS	AR-MS	AR-MS	AR-MS	AR-MS
S15 Orig	2.53	2.38	3.27	735	3.49	0.027	0.6	0.12
S15 Dup	2.61	2.39	3.05	725	3.61	0.026	0.66	0.13
S28 Orig	1.97	2.17	2.47	507	2.74	0.029	0.47	0.14
S28 Dup	1.87	2.25	2.45	512	2.84	0.028	0.52	0.14

Phosphorus Sorption QA/QC

Control Cups and Drifts – Quality control cups with null samples were placed in triplicate evenly throughout runs to ensure cleanliness of glassware, with all samples indicating lack of phosphorus contaminations and all standard deviations falling below 5 µgP/L. Quality Cups and Drifts for P concentrations of 10, 25, 50, 100 and 200 µg/L were run in at least triplicate, longer runs had more quality cups. Drifts (200 µg L⁻¹) were placed evenly throughout each run. For each run all standard deviations fell below 5 µg L⁻¹.