Coagulant addition for managing sediment-associated phosphorus bioavailability to prevent cyanobacterial blooms in drinking water reservoirs

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 final revisions, as accepted by my examiners.

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Statement of Contributions

Nik Knezic was the sole author for Chapters 1 and 4 which were written under the co-supervision of Dr. Monica Emelko and Dr. Mike Stone and were not written for publication.

This thesis consists in part of two manuscripts prepared as refereed papers for publication that have not yet been submitted. Exceptions to sole authorship of material are as follows:

Research presented in Chapter 2:

This research was conducted at the University of Waterloo by Nik Knezic under the co-supervision of Dr. Monica Emelko and Dr. Mike Stone. Nik Knezic designed the literature review and completed the compilation of information with assistance from Dr. Monica Emelko and Dr. Mike Stone. Dr. William (Bill) Anderson contributed to designing the searches for information regarding cyanobacteria and cyanotoxins. Nik Knezic drafted the manuscript and each author provided intellectual property on manuscript drafts.

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Research presented in Chapter 3:

This research was conducted at the University of Waterloo by Nik Knezic under the co-supervision of Dr. Monica Emelko and Dr. Mike Stone. Nik Knezic designed the study with consultations from Dr. Monica Emelko and Dr. Mike Stone who contributed to the data analysis. Nik Knezic drafted the manuscript and each author provided intellectual input on manuscript drafts.

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Abstract

To ensure the uninterrupted supply of adequate amounts of drinking water, many utilities rely on reservoirs for raw (i.e., untreated) water storage prior to treatment. For example, reservoirs are integral to storing water originating as mountain snowpack that melts and slowly releases water to downstream rivers and lakes, serving ~75% of the western United States and Canada and approximately two billion people globally. Although raw water supply reservoirs have been historically managed for water quantity, not quality, reservoir management objectives are rapidly evolving. The importance of reservoir management for source quality is increasing as the relationships between source water quality, treatment costs, finished water quality, and public health protection are better understood, and climate change-exacerbated pressures on that relationship are better described.

Water supply reservoir management is increasingly recognized as an integral component of risk management in the water industry due to the inextricable connection of climate change to source water quality and treatment costs, finished water quality, and public health protection. Multipurpose reservoirs frequently provide seasonal flow equalization, storage during periods of high precipitation (i.e., rain, snow melt), hydroelectric power, and flood mitigation; they also ensure that demand can be met during low flow periods and droughts. Notably, reservoirs are not typically managed for influxes of fine sediment and associated nutrients, which are more frequent in many areas because of climate change-exacerbated landscape disturbances such as wildfires and extreme precipitation.

Algae, especially cyanobacteria, blooms are one of the biggest threats to water quality and the provision of safe drinking water globally. High densities of algal cells have the potential to lead to customer complaints, service disruptions, and even outages, especially in water treatment plants lacking advanced treatment options. Phosphorus (P) is the limiting nutrient for primary productivity in freshwater. Fine sediment is the primary vector of phosphorus transport in aquatic systems, thus fine sediment management to mitigate or prevent releases of bioavailable P to the water column could be integrated into water treatment operations, potentially as a climate change adaptation strategy. Drinking water reservoirs are not typically designed to manage internal loading of phosphorus; while this has been well studied in lakes, investigations of management strategies such as coagulant addition to prevent phosphorus release from bottom sediments (i.e., phosphorus inactivation) to mitigate the proliferation of cyanobacteria in raw water storage reservoirs are scant. Here, a series of lab- and field-scale analyses were conducted to (i) describe phosphorus release from fine sediment in a raw water reservoir, (ii) characterize its availability for biological uptake, (iii) evaluate phosphorus inactivation by application of common coagulants ($FeCl_3$, alum, PACl), and (iv) evaluate the combination of strategically-timed reservoir dredging and coagulant application on phosphorus inactivation and turbidity reduction. This study demonstrated that significant amounts of phosphorus were readily released from fine sediment in the study reservoir, suggesting the need for fine sediment management. Application of typical doses of common chemical coagulants, especially $FeCl_3$ effectively inactivated phosphorus to below target thresholds in the presence of fine sediment, as would be expected. Moreover, the combination of reservoir dredging and coagulant application during higher algae risk periods not only inactivated phosphorus, but also eliminated the potential for its re-release to the reservoir water column with the concurrent benefit of turbidity reduction. Thus, this study demonstrated that seasonal coagulant application coupled with strategically-timed reservoir dredging may offer utilities reliant on offline raw water storage reservoirs an effective P inactivation approach for risk management and climate change adaptation.

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List of Abbreviations

AP   apatite phosphorus
BMP   best management practice
DBP   disinfection by-product
DWR   drinking water reservoir
DWTP   drinking water treatment plant
Dx   specific diameter

EPC_0 equilibrium phosphate concentration
g   grams
HVR   Hidden Valley Reservoir
L   liter
MIB   2-methylisoborneol
ML   million liters
mL   milli liters
MLD   million liters per day
MWTP   Mannheim water treatment plant
NAIP   non-apatite inorganic phosphorus
ng   nano gram
NOM   Natural organic matter
OP   organic phosphorus
P   phosphorus
PI   phosphorus inactivation
ROW   Region of Waterloo
RPM   revolutions per minute
SWM   storm water management pond
TBP   total biologically available phosphorus
TDP   total dissolved phosphorus
TOC   total organic carbon
TP   total phosphorus
TPP   total particulate phosphorus
v   volume
μm   micro meter

Chapter 1
Introduction

1.1 Background

To ensure the uninterrupted supply of adequate amounts of drinking water, many utilities rely on reservoirs for raw (i.e., untreated) water storage prior to treatment. Reservoirs are integral components of many drinking water treatment plants that are used to ensure the adequate and continuous supply of water. They are used for flow equalization and storage; they also ensure that demands are met (Avakyan, 1967; Brunner et al., 2019). For example, reservoirs are integral to storing water originating as snowpack in watersheds that melts and slowly releases water to downstream rivers and lakes, serving approximately 75% of the western United States and Canada and approximately two billion people globally (Mankin et al., 2015; U.S. Geological Survey, 2021). Despite historical management focus on water quantity, source water supply reservoir management objectives are evolving to simultaneously focus on water quality. Water quality importance is further amplified as the relationships between source water quality and treatment costs, finished water quality, and public health protection are better understood. Furthermore, these relationships are becoming more important as their climate change-exacerbated pressures are better described.

With increasing recognition of the inextricable connection of climate change to source water quality and treatment costs, finished water quality, and public health protection, there is also increasing recognition of water supply reservoir management as an integral component of broader risk management in the water industry (Emelko et al., 2011). Multipurpose reservoirs frequently provide seasonal flow equalization, storage during periods of high precipitation (i.e., rain, and/or snow melt), hydroelectric power, and flood mitigation; they also ensure that demand can be met during low flow periods and droughts (University of Idaho, 2001; U.S. Geological Survey, 2021). Notably, however, reservoirs are not frequently managed for influxes of fine sediment and associated nutrients, which (i) are more frequent in many areas because of climate change-exacerbated landscape disturbances such as wildfires and extreme precipitation (Emelko et al. 2016) and (ii) promote the proliferation of algae (U.S. Environmental Protection Agency, 2001; 2021; Lehman, 2010; Silins et al. 2014; Orihel et al., 2015; Emelko et al. 2016).

Algae, especially cyanobacteria, blooms are one of the biggest threats to water quality and the provision of safe drinking water in Canada and globally. High densities of cyanobacteria can increase source water turbidity, thereby increasing chemical coagulant demand, settling time, sludge production, and disinfectant/oxidant demand; they can also clog filters and decrease filter run times (Emelko et al. 2011; Crittenden, 2012). Notably, some cyanobacteria have the potential to form unpleasant taste and odor compounds, and release toxins of human health and environmental concern, which may require advanced treatment beyond that typically found in most conventional treatment plants (Jung et al., 2004; Watson et al., 2008; Westrick, 2008; Ho et al., 2012a). These toxins include the neurotoxin anatoxin-a, the hepatotoxin microcystin, and cylindrospermopsin which is a hepatotoxin and may also impact other organs (Beasley, 2020); while all have implications for human health, only microcystin is currently regulated in Canada (Health Canada, 2012).

Cyanobacteria require specific environmental conditions (e.g., temperature, pH, light availability, and nutrient availability) to survive, grow, and proliferate (Paerl et al., 2001; Watson et al., 2008; U.S. Environmental Protection Agency, 2012a). Phosphorus (P) is considered the limiting nutrient for primary productivity in freshwater systems; thus, it is also recognized as a key factor in cyanobacteria growth and metabolism (Schindler, 1974; U.S. Environmental Protection Agency, 2012a; Metcalf & Codd, 2014). A total phosphorus concentration of 30 µg P L^{-1} is a commonly cited threshold for eutrophication (Barlow-Busch et al., 2006; Chambers et al., 2012). This threshold is a benchmark rather than a predictive tool; cyanobacteria can still proliferate in aquatic systems with total phosphorus concentrations less than 30 µg P L^{-1} (Winter et al., 2008; Vuorio et al., 2020), though it is considered less likely to occur. Although it is widely understood that phosphorus bioavailability alone does not guarantee algal proliferation and other factors such as temperature, micronutrient availability, light levels, etc. are also significant factors that affect it (Paerl et al., 2001; Watson et al., 2008; U.S. Environmental Protection Agency, 2012a), if drinking water reservoir managers can limit water column total phosphorus concentrations so that they are below this threshold, the potential for cyanobacteria proliferation and the associated production of toxins and unpleasant taste and odor compounds may be mitigated nonetheless (Watson et al., 2008; U.S. Environmental Protection Agency, 2012a).

Fine sediment (i.e., < 63 µm) is the primary vector for phosphorus transport in aquatic systems (Stone & English, 1993; Auer et al., 1998; Davies-Colley & Smith, 2001; Dodds, 2003). Due to its low

settling velocities and highly adsorptive surfaces, fine sediment and associated phosphorus can be propagated across the river continuum and transferred to downstream drinking water reservoirs (Emelko et al., 2016). Fine-grained sediments have the potential to release phosphorus to the water column via desorption and dissolution; when this occurs in lakes and reservoirs it is referred to as internal loading (Withers & Jarvie, 2008). Biological controls such as periphyton and phytoplankton, microorganisms, and macrophytes can further impact internal phosphorus flux (from sediments to the water column) to varying extents (Withers & Jarvie, 2008).

Increases in the frequency and severity of climate change-exacerbated landscape disturbances, changing hydro-climactic conditions, and temperature variation from climactic normal can impact water quality (Moore et al., 2008; Schindler et al., 2008; Blake et al., 2009; Stone et al., 2011; Seeboonruang, 2012; Silins et al., 2014). Both natural (i.e., wildfires, hurricanes, floods, and precipitation changes) and anthropogenic (e.g., urbanization, agriculture, industrial) landscape disturbances can alter hydrological processes and affect water quality and treatability. Wildfires, for example, are among the most severe landscape disturbances that can impact water globally (Robinne et al., 2016; 2019). They can lead to increases in the amount of precipitation that reaches the landscape; consequently, they also increase runoff of contaminants from the landscape (Moody & Martin, 2001; Williams et al., 2019), even in systems with already deteriorated source water quality (Emmerton et al., 2020). After fires, water temperature can increase (Wagner et al. 2014) and concentrations of nutrients (Kunze & Stednick, 2006; Silins et al., 2014), suspended sediments (Kunze & Stednick, 2006; Silins et al., 2009), metals (Wolf et al., 2008), and other contaminants (Kalabokidis, 2000; Crouch et al., 2006) are typically increased. Such changes in source water quality can can lead to increased drinking water treatment costs and sometimes necessitate investment in upgraded treatment infrastructure (Emelko et al., 2011; Emelko and Sham, 2014; Price et al., 2017).

While some aspects of source water quality change are unique to disturbance type (e.g., fire retardant releases, industrial spills) and most landscape disturbances have the potential to degrade source water quality and impact drinking water treatability to some extent, one of the most notable of such impacts is the increased mobility and delivery of fine (i.e., cohesive) sediments to receiving waters. This is because they can lead to increases in nutrient availability—especially bioavailable phosphorus—in the water column (DePinto et al., 1981; Carpenter et al., 1998; Mainstone & Parr, 2002; Withers & Jarvie, 2008). These impacts on water quality and treatability can persist for decades

and propagate for tens of kilometers or more downstream after severe disturbance in some physiographic settings (Stone et al., 2014; Emelko et al., 2016), increasing fine sediment-associated phosphorus loading rates to lakes and reservoirs. Increases in the delivery of nutrient-rich fine sediment increases the probability of cyanobacterial proliferation in lakes and reservoirs. It has been recently suggested that alarming continental-scale increases in lake and stream total phosphorus (TP) concentrations in oligotrophic rivers draining relatively undeveloped forest environments that were identified through surveys of thousands of water bodies in the conterminous U.S. can likely be attributed to climate change driven extremes in precipitation and high magnitude runoff events, alarmingly resulting in oligotrophic systems "disappearing" in the U.S. (Stoddard et al., 2016). Many such oligotrophic water bodies also serve as high quality source supplies of drinking water. At present, drinking water reservoirs are not typically designed to manage internal loading of phosphorus and while this process has been well studied in lakes, investigations of management strategies (e.g., coagulant addition) to prevent phosphorus release from bottom sediments (i.e., phosphorus inactivation) to the mitigate algal proliferation in raw water storage reservoirs are scant (Pütz & Benndorf, 1998). Thus, there is an opportunity to develop fine sediment management approaches that mitigate or prevent releases of bioavailable phosphorus to the water column and integrate them into drinking water source protection and supply management plans, as well as climate change adaptation strategies.

Increasing phosphorus loading from watersheds and its connectivity to promoting cyanobacterial growth highlights the need for water utilities to implement sediment management strategies that focus on drinking water source quality. While this work has been attempted in lakes and on-line reservoirs (Welch & Cooke, 1999; Wagner & Adrian, 2009), there is a paucity of research conducted in offline reservoirs, where sediment delivery to rivers is increasing due to climate and landscape change. Minimizing the effects of phosphorus release to the water column of drinking water reservoirs can be accomplished by (1) reducing the external loading of sediment upstream of the reservoir, and/or (2) suppressing the internal loading of phosphorus within the reservoir (Wagner & Adrian, 2009). While there is abundant scientific literature and guidance regarding small and large lake-scale best management practices (e.g., erosion control measures such as silt fences, enhanced agriculture practices such as reduced cultivated fallow, wastewater treatment plant upgrades focused on phosphorus removal) design to reduce the external loading of fine sediment, consideration of

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strategies for suppressing internal loading within drinking water reservoirs remains limited (U.S. Environmental Protection Agency, 2001; Canadian Council of Ministers of the Environment, 2016).

1.2 Research Objectives

The overall goal of this research was to identify and develop drinking water reservoir management strategies to prevent the proliferation of cyanobacteria in raw (i.e., untreated) water storage reservoirs. To address this goal, the specific objectives of the research were to:

1) characterize the physical and geochemical properties, including particulate phosphorus forms, of fine (i.e., cohesive) sediment deposited in a drinking water reservoir to evaluate sediment phosphorus buffering;

2) identify an aqueous phase total phosphorus threshold concentration above which cyanobacteria proliferation is more likely to occur in drinking water reservoirs;

3) evaluate the phosphorus release potential of fine sediment deposited in a drinking water reservoir;

4) evaluate chemical coagulant inactivation (i.e., sequestration) of phosphorus as a measure to reduce cyanobacteria proliferation in drinking water reservoirs; and

5) evaluate the combination of strategically-timed reservoir dredging and coagulant application for phosphorus inactivation in drinking water reservoirs.

1.3 Research Approach

Minimizing the potential for cyanobacterial bloom occurrence by managing nutrient availability in drinking water reservoirs is an effective risk management strategy for some water providers (Knopman et al., 2018). Although many water utilities have implemented best management strategies for reducing the amounts of bioavailable phosphorus in source watersheds, those strategies have largely focused on reducing external loads to source water or drinking water reservoirs. Internal loading of phosphorus can also be managed; several studies have focused on minimizing internal

loading of phosphorus in lakes (Harper et al., 1999; Welch & Cooke, 1999; Huser et al., 2011; Wagner et al., 2017) or inline drinking water reservoirs (Kennedy et al., 1987; Barko et al., 1990; National Academies of Sciences, Engineering, and Medicine, 2018) for improving water quality. Critically, however, they have been studied often using just one management strategy, and without a focus on directly implications for drinking water treatment.

There are currently no known standardized monitoring programs for identifying the impact of fine sediment and associated phosphorus releases in drinking water reservoirs on the potential for cyanobacteria growth. These gaps include sediment sampling protocols (e.g., location, replication of samples, time of year), incoming water quality parameter monitoring (i.e., dissolved and total phosphorus concentrations), phosphorous release potential design, and fine sediment management practices. In addition, few studies have examined the spatial variability of the phosphorus in bottom sediment within the drinking water reservoir. Thus, to address the research objectives identified in Section 1.2, this research was conducted in three phases.

The general outline of the study describing how each objective supports the overall research structure is shown in Fig.1-1. The first experimental phase of this thesis research was designed to evaluate fine sediment buffering of phosphorus availability in a drinking water reservoir that stores agriculturally and municipally impacted source water. Fine grained sediment samples were collected from the partnering water utility's Hidden Valley Reservoir (Waterloo, Ontario, Canada) and physical and geochemical properties of sediment including grain size distribution, major element composition, as well as particulate phosphorus speciation were evaluated (Objective #1). Next, a total aqueous phosphorus concentration threshold was identified based on a review of the literature. It was identified to serve as a performance target representing the aqueous phase total phosphorus concentration above which cyanobacteria proliferation is more likely to occur in drinking water reservoirs (Objective #2). Batch experiments were then conducted to quantify the potential for reservoir bottom sediments to release phosphorus to the water column (Objective #3).
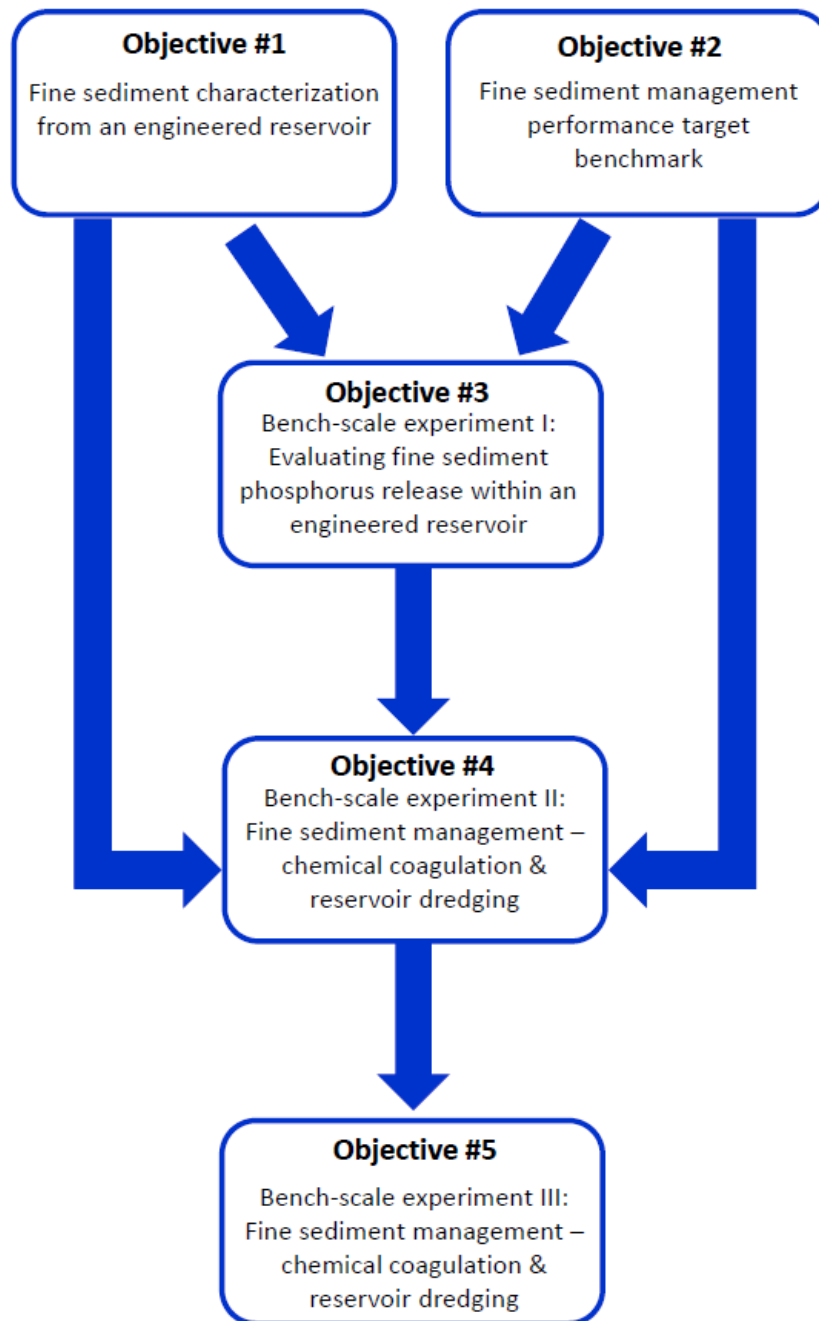
Figure 1-1: Conceptual diagram of the inter-connectivity between the major research objectives comprising this thesis research.

The second phase of the research was designed to investigate the efficacy of adding common coagulants used in the drinking water industry to reservoir inflows to minimize phosphorus release from deposited fine sediments to the reservoir water column (Objective #4). Phosphorus inactivation by three chemical coagulants ($FeCl_3$, alum, and poly-aluminum chloride [PACl]) commonly used in drinking water treatment was investigated. The aqueous phase total phosphorus threshold concentration identified in completing Objective #2 was used as a performance target for identifying applied coagulant doses that corresponded to successful phosphorus inactivation.

The third phase of bench-scale analysis was to investigate the potential treatment performance benefits of strategically-timed sediment removal (i.e., dredging) from engineered reservoirs in combination with coagulant addition for phosphorus inactivation in drinking water reservoir (Objective #5). Reservoir dredging after the start of coagulant application to the reservoir inflow ensures that both (i) dissolved phosphorus in the inflow and (ii) particulate phosphorus bound to sediment deposited in the reservoir have been inactivated. Three chemical coagulants ($FeCl_3$, alum, and PACl; the same coagulants investigated during the phase II experiments) were again investigated and the synergistic effects of chemical coagulation and strategically-time reservoir dredging for phosphorus inactivation in drinking water reservoirs were evaluated.

1.4 Thesis Organization

This thesis consists of four chapters, with Chapters 2 and 3 formatted for submission to refereed journals (i.e., a paper-based thesis); thus, there is some repetition of introductory material in Chapters 2 and 3.

Chapter 2 provides an overview of current knowledge and research regarding fine grained sediment impacts on phosphorus release to water supplies and associated risks of cyanobacteria proliferation on drinking water treatment. Chapter 2 concludes with a summary of the limitations of current knowledge regarding fine sediment impacts on drinking water reservoirs. Chapter 3 provides a brief literature background for the theory behind the management options of fine sediment, the experimental design, results of the management options, and recommendations for fine sediment management going forward along with future research opportunities. Finally, in Chapter 4 the

experimental data are integrated with the literature review to provide fine sediment protocols for the reduction of bioavailable phosphorus within drinking water reservoirs. The chapter concludes with a summary, a list of conclusions and description of the implications to drinking water source management.

Chapter 2

Management of cyanobacteria bloom risks in drinking water reservoirs requires the consideration of fine sediment

2.1 Reservoirs and drinking water supply

Surface water originates on landscapes primarily as a combination of runoff during precipitation and snowmelt events, and groundwater inputs to rivers—it is largely generated on forested landscapes in Canada and the United States. Globally, it provide drinking water for approximately two-thirds of Americans (Stein & Butler, 2004) and Canadians, and one-third of the world's largest cities (Dudley & Stolton, 2003). Reservoirs are used globally to manage source water for multiple uses that include crop irrigation, energy production, flow equalization, flood protection and domestic water supply (University of Idaho, 2001; Patterson et al., 2016;). Notably, many utilities rely on reservoirs for raw (i.e., untreated) water storage prior to treatment to ensure uninterrupted supply of drinking water.

Two general reservoir designs are used for the storage of water along the river continuum. They include (1) online designs in which rivers are impounded and water storage occurs in the river channel and floodplain, and (2) offline designs in which water is diverted from the river channel and stored in an engineered facility or depressions in the landscape (Ackers et al., 2010) (Fig. 2-1). Reservoir design is a function of the landscape and river characteristics, and retention time (ranging from a few hours to years). Drinking water reservoirs can serve as single, or multi-purpose facilities (Patterson et al., 2016). Although water supply reservoirs have been historically managed for water quantity, they have been managed to a much lesser degree for water quality (U.S. Environmental Protection Agency, 2001). Given the evolving pressures of landscape disturbance coupled with the increasing effects of climate warming on surface water quality, reservoir management objectives are rapidly evolving.
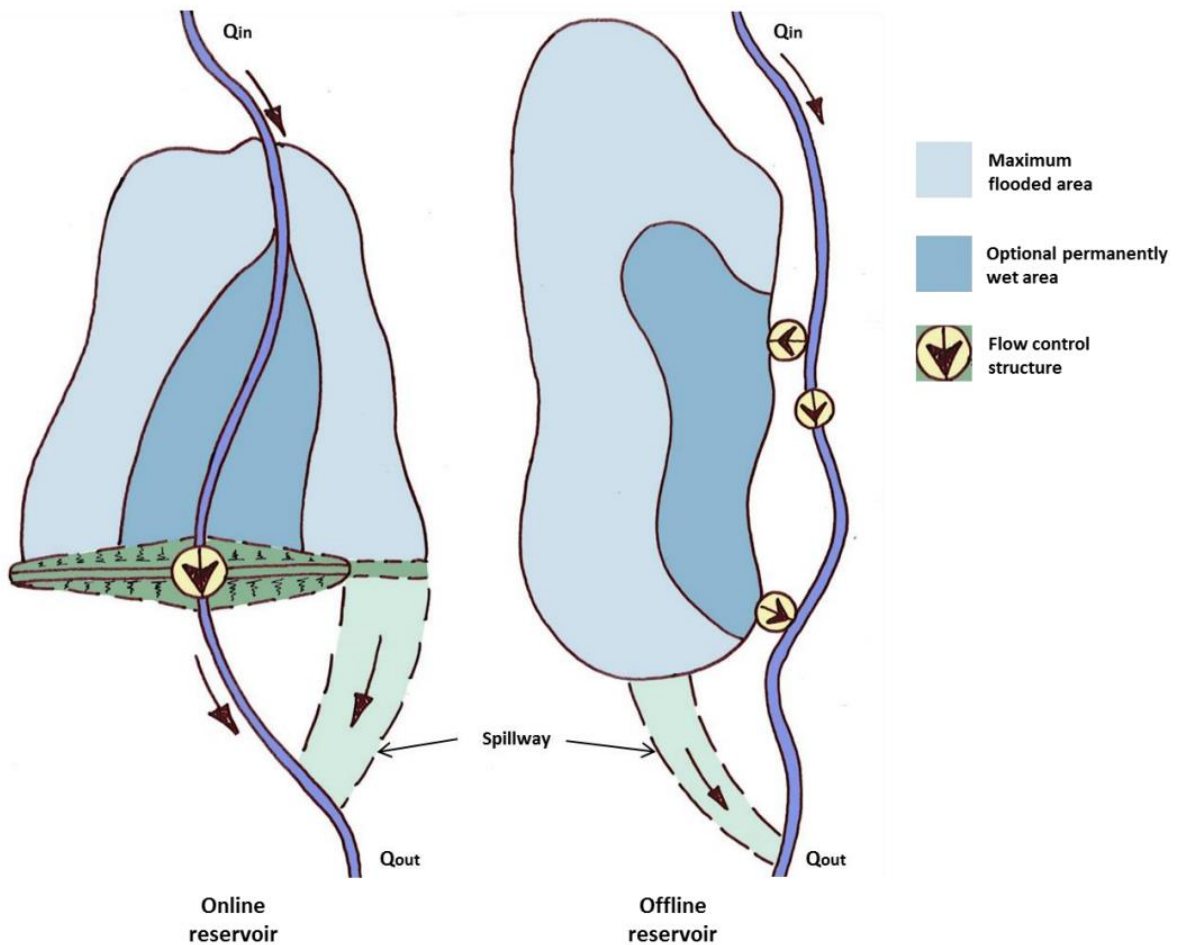
Figure 2-1: Online reservoirs temporarily store water within the river channel, while offline reservoirs have river water diverted away from the river channel into them. Online reservoirs are impounding structures typically made from earth (e.g., dirt, sand, and/or rocks), or concrete which impede the flow of the river and floodplain (i.e., a dam). Offline reservoirs consist of an intake structure to divert river flow with a storage area formed by either low ground levels or retaining structures (Patterson et al., 2016).

Typical online reservoirs allow for water quality equalization that improves drinking water treatability, while many offline reservoirs are designed to have the further option of closing intakes if upstream surface source water quality deteriorates (e.g., pollutant spill or high turbidity event). The multi-utility of online reservoirs is evidenced in the "World Register of Dams", which accounts for all registered, large (> 15m) dams from over 100 countries. In 2020, there were a total of 58,713 registered dams and 7,963 registered reservoirs used for drinking water supply; a further 4,587

reservoirs were classified as "multi-purpose" (ICOLD, 2011). These data underscore global dependency on reservoirs for the provision of potable water; this dependency is further realized when accounting for the smaller (< 15m) inline reservoirs and all sizes of offline reservoirs are considered.

For at least the last seven years, source water supply availability has continually been identified by drinking water professionals in North America as a critical long-term challenge facing the drinking water industry over the last seven years (AWWA, 2015 – 2021). Specifically, there are a growing number of threats to quantity, quality, and seasonal availability of source water in North America, which currently accounts for 60% of all drinking water within the U.S. and is facing increasing demand due to industrial and municipal development (U.S. Environmental Protection Agency, 2012a; Dieter et al., 2018). Water quantity threats to source water include increases in precipication frequency and severity, changes in flood frequency, while increases in global temperature result in increasing periods of severe drought (Schindler & Donahue, 2006; U.S. EPA, 2021). Threats to water quality include increased erosion and delivery of particulate matter in runoff generated by changes to the intensity, duration and frequncy of precipitation (Emelko et al., 2011). Particularily, increased sediment and associated nutrient delivery, coupled with increases in global temperatures can also lead to more common blooms of harmful algal blooms (U.S. EPA, 2021). Shifts in seasonal availability of water caused by events such as increased precipitation, earlier snowpact melt, and prolonged periods of drought can threaten the availability of source water for drinking water utilities across the globe. Concerningly, these threats to source water quality and treatability are expected to be exacerbated by climate change due to the increasing frequency and severity of natural disturbances such as wildfires and extreme precipitation events (Emelko et al., 2011).

2.2 Nutrient dynamics and algal blooms in reservoirs

The quality of surface waters globally is increasingly under threat from the excessive delivery of nutrients to receiving waters and their subsequent role in the proliferation of algae (Winter et al., 2011; (Hallegraeff, 1993) Drinking water reservoirs are particularly susceptible to bloom occurrence because they are not properly designed to optimize water quality. Shallow depths, warm water temperatures and quiescent conditions are ideal for algal occurrence (Crittenden et al., 2012). Critically, the presence of algae in raw water storage reservoirs and drinking water treatment plants

can pose significant threats to the provision of adequate amounts of safe drinking water by challenging conventional water treatment technologies such as coagulation, flocculation, clarification, granular media filtration, and disinfection (Westrick et al., 2010; Emelko et al., 2011).

Algal bloom occurrence can reduce water throughput in water treatment systems, which can ultimately lead inability to meet demands and water use restrictions. While most algal cells can be removed with effective coagulation and sedimentation, cells that remain suspended can clog filter beds or membranes, causing gradual or rapid head loss (Crittenden et al., 2012). The accumulation of nuisance algae can cause an increase in pressure resulting in reduced filter run times, extended filter backwash times, or increased risk of turbidity breakthrough (Crittenden et al., 2012; Qu et al., 2012). Algal blooms can also lead to customer complaints due to the release of unwanted metabolites, which impart taste and odors to water. While not harmful, these compounds (geosmin and 2-methylisoborneol - MIB) produced by algal blooms and cyano- and other bacteria can produce foul odors that result in customer complaints (Crittenden et al., 2012). Due to the low threshold for detection, particularly in geosmin and MIB, water treatment plants typical target maximum concentrations of ~5 ng L^{-1} of these of compounds in final treated water (McGuire et al., 1981; Simpson & MacLeod, 1991). Advanced treatment technologies (e.g., activated carbon, advanced oxidation) are often required to remove these compounds because they are not easily removed by conventional water treatment methods (Jung et al., 2004; Skjevrak et al., 2004; Jüttner & Watson, 2007; Ho et al., 2012a).

The presence of algal blooms in raw water storage reservoirs can necessitate "do not drink" advisories to utility customers when toxins may be produced by the algae. Notably the presence of cyanobacteria and even cyanobacterial blooms in water treatment plants can challenge final treated water quality by (1) exerting increased chemical oxidant demand, which can increase the potential for disinfection by-product (DBP) formation (Aktas et al., 2012; Crittenden et al., 2012; Antoniou et al., 2014), (2) clogging filters which can potentially lead to turbidity and/or pathogen breakthrough (Pirbazari et al., 1993; Emelko 2001; Emelko et al., 2005; Ho et al., 2006; Drikas et al., 2009; Zamyadi et al., 2012a; American Water Works Association, 2012), and/or (3) potentially releasing cyanotoxins during the treatment process (Jüttner & Watson, 2007; Westrick, 2008). While not all algal blooms contain cyanobacteria, and not all cyanobacteria can produce and release cyanotoxins, increases in cell counts are generally linked to increased potential for cyanotoxin production (Hitzfield et al., 2000).

Most cyanotoxins are retained in healthy cyanobacteria cells or bacteriophages. However, when those cells become stressed or lyse, such as during water treatment, toxins can be released to the water (Thornton et al., 1996; Lopez et al., 2008; Mankiewicz et al., 2003). These toxins include hepato-, neuro-, or dermatoxins, and respectively affect the liver, brain, or skin (Hitzfield et al., 2000; Haider et al., 2003). They are not easily removed during conventional water treatment (Jung et al., 2004; Skjevrak et al., 2004; Jüttner & Watson, 2007; Ho et al., 2012a; U.S. Environmental Protection Agency, 2012a; Ontario Ministry of Environment and Climate Change, 2012; Antoniou et al., 2014).

The global occurrence of cyanobacteria blooms and the associated challenges posed to drinking water treatment are expected to deteriorate as a result of a changing climate (Hallegraeff, 1992; Thornton et al., 1996; Haider et al., 2003; Winter et al., 2011; Loza et al., 2014. These threats will likely increase because of climate change-driven increases in frequency and severity of natural landscape disturbances such as fires and extreme precipitation (Moore et al., 2008; Wagner & Adrian, 2009). While measures to mitigate algal blooms in reservoirs have been proposed (U.S. Environmental Protection Agency, 2001), only a few are designed to address nutrient availability and fine sediment (U.S. Environmental Protection Agency, 2001). These options include (1) hypolimnic/sediment oxygenation, (2) dredging, and (3) phosphorus inactivation.

2.2.1 Algal blooms and drinking water treatment risks

Several examples of cyanobacterial proliferation risks to the provision of safe drinking water have been reported in the literature (Alliance for the Great Lakes, 2019). One of the most extreme cases of this occurred when unsafe levels of the cyanotoxin microcystin-LR were measured in the final treated effluent of the Toledo drinking water treatment plant in August 2014 (U.S. Environmental Protection Agency , 2012a). A "Do Not Drink" advisory was promptly announced, leaving nearly half a million residents without access to safe water for over three days. The toxins were attributed to the reoccurring cyanobacterial blooms that occurred in the western basin of Lake Erie (Fig. 2-2). A few weeks after the Toledo incident a similar "do not drink" order was issued for the residents of Pelee Island, Ontario (Alliance for the Great Lakes, 2019).

Figure 2-2: Reoccurring algal bloom on the Western Lake Erie Basin near Toledo, Ohio, U.S.A. (Alliance for the Great Lakes, 2019). A 2014 bloom in this area released microcystin-LR that lead to a "do not drink" advisory leaving almost 500,000 people without access to clean drinking water.

2.2.2 Sediment-associated phosphorus and algal blooms

While the morphological and limnological characteristics of some drinking water reservoirs (i.e., shallow depths and quiescent conditions) may be ideal for cyanobacteria growth, bioavailable phosphorus (in both dissolved and particulate forms) is one of the key drivers of cyanobacterial proliferation (Håkanson et al., 2007; Metcalf & Codd, 2014). Specifically, phosphorus is the key limiting nutrient for primary productivity and proliferation of algae more specifically, in freshwater systems (Schindler, 1977; Barlow-Busch et al., 2006). In a recent study conducted across a range of ecozones in Canada, Chambers et al. (2012) reported a range of total phosphorus thresholds (10-30 µg TP L^{-1}) that resulted in a greater potential of good ecological condition for benthic algae abundance. It is

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important to note that this threshold is a benchmark rather than a predictive tool; source waters with an aqueous total phosphorus concentration below 30 µg P L^{-1} can still have cyanobacteria and bloom formation, however, it is considered less likely to occur. While the dynamics and drivers of cyanobacteria blooms remain poorly understood (Ma et al., 2014; Holland & Kinnear, 2013), a reduction of phosphorus availability has been shown to decrease the likelihood of bloom occurrence in drinking water reservoirs (Ma et al., 2015; Schindler et al., 2016; Paerl et al., 2016).

The total phosphorus concentration in aquatic systems is the sum of total dissolved phosphorus (TDP) and total particulate phosphorus (TPP) forms. These forms are operationally defined. Dissolved phosphorus is that fraction that passes through a 0.45 µm filter, while the larger fractions retained on the filter comprise total particulate phosphorus forms (American Water Works Association, 2017). TDP is made up of either orthophosphate or colloidal bound orthophosphate, both of which are readily bioavailable (Reynolds & Davies, 2001). Particulate phosphorus (PP) forms are fractionated into three PP forms: (1) non apatite inorganic phosphorus (NAIP), (2) apatite phosphorus (AP), and (3) organic phosphorus (OP). NAIP is typically bound to iron, aluminum, and manganese on particle surfaces, and they can be released into the water column as dissolved P which is the most bioavailable P form. AP is bound to calcium carbonate and is stable and not bioavailable. While OP is not directly bioavailable, some components of this fraction can be bioavailable after undergoing hydrolysis or mineralization. Humic acids comprise the largest fraction of OP in aquatic systems; this fraction is not rapidly bioavailable (Reynolds & Davies, 2001; Emelko et al., 2016).

2.3 Phosphorus loading in reservoirs: External vs internal loading

Phosphorus can enter an aquatic system either via external loading or internal loading pathways (Auer et al., 1998). External loading pathways for phosphorus can include wet and dry deposition, ground water inputs, surface runoff and tributary inflows (Marsden, 1989). Tributary inflows contain both particulate and dissolved fractions, but sediment-associated particulate phophorus is the predominant form. The relative proportion of disolved and particulate forms in tributary infows depends on landscape and hydrological conditions and the time of the year.

Fine sediment and associated phosphorus can enter surface waters via either point or non-point sources (Fig. 2-3), which include continuous flows from municipal or industrial waste streams

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such as wastewater treatment plants. In contrast, non-point sources of phosphorus are often discontinuous and diffuse, typically from runoff, ground water inputs or atmospheric deposition. Approximately 84% of external phosphorus loading to aquatic surface waters has been attributed to non-point sources such as croplands, pastures, rangelands, and forests (Carpenter et al., 1998). The challenge of external loading is expected to become more severe, due to continued natural (i.e., wildfires, hurricanes, floods, and precipitation changes) and anthropogenic (e.g., urbanization, agriculture, industrial) landscape disturbances that alter hydrological processes and in turn increase sediment erosion (Shakesby & Doerr, 2006; Smith et al., 2011; Intergovernmental Panel on Climate Change, 2013; Emelko et al., 2016). It follows that as the frequency and severity of these disturbances increase, the rates of external phosphorus loading to drinking water reservoirs will also increase unless best management practices to mitigate phosphorus transfer are effectively implemented.
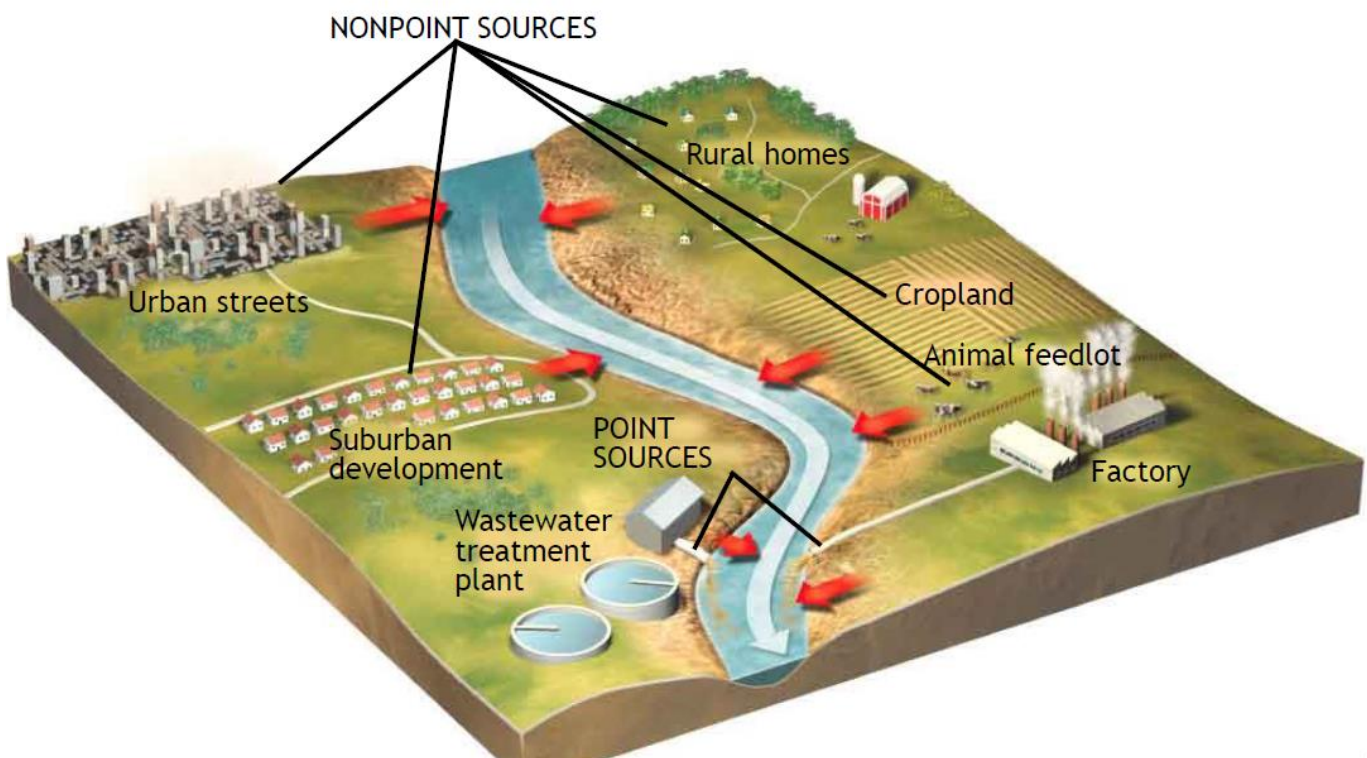
Figure 2-3: Point and non-point sources of fine sediment and associated phosphorus that impact surface water (Whitehead, 2020) emphasize the importance of reservoir management for mitigating diffuse source water pollution relative to direct discharges.

In response to risks due to external phosphorus loading to surface waters, BMPs have been implemented to reduce sediment and associated nutrient loading. Examples of BMPs include tertiary wastewater treatment plants, riparian buffers, wetland construction, and enhanced agricultural measures such as optimized fertilizer application, cover crop use, or reduced tillage practices (Carpenter et al., 1998; Rahman & Bakri, 2010; Jiao et al., 2015; Hao et al., 2016). When properly designed and maintained BMP's can reduce phosphorus loading to surface waters, however some of the agricultural measures, despite their best intentions, lead to increases in bioavailable phosphorus in the surface waters (Jarvie et al., 2017). Furthermore, the expected water quality improvements to BMPs in many freshwater systems have been delayed due to "legacy phosphorus" stored in rivers and lakes due to decades of historical nutrient loading (Kleinman et al., 2011; Sharpley et al., 2013).

In contrast to external loading, internal loading describes the role of deposited sediment in an aquatic system to release phosphorus to the water column through adsorption/desorption (i.e., sorption) reactions (Nürnberg et al., 2013). Phosphate release from fine sediment has been identified as a main driver for algal growth in many freshwater systems and can result in partial or delayed responses to external loading reduction efforts (Lehman, 2010; Nürnberg et al., 2013; Orihel et al., 2015). Due the impact of internal loading in drinking water reservoirs, water utilities must consider fine sediment management strategies for suppressing the impact of internal loading. Phosphate release from fine sediment into the water column is complex and depends on several factors such as: sediment grain size and geochemistry, redox conditions, pH, competitor ions, stream order and sediment phosphate concentration (Boström & Pettersson, 1982).

Phosphorus sediment sorption behavior is a two-step process that involves (1) slow solid-state diffusion and (2) rapid surface sorption (Froelich, 1988). The direction of phosphorus sorption process can be determined by plotting the solid phase phosphorus as a function of aqueous phosphorus concentration (Fig. 2-4). These plots are typically determined using phosphorus isotherm experiments and sorption models (e.g., Langmuir, Freudlich, Tempkin). The ability of sediments to absorb or desorb phosphorus can be quantified by estimating the maximum absorption capacity of sediment or the solution concentration at which neither absorption or desorption occurs. This condition is known as the 'zero equilibrium phosphorus concentration' (EPC_0) (Froelich, 1988). EPC_0 is determined using batch experiments in the laboratory using a series of SRP concentrations mixed with a known mass of sediment for sufficient time that equilibrium between the sediment and solution can

be assumed (Golterman, 2004). The data are then fitted with initial phosphorus concentration (μg P L^{-1}) on the x-axis and P absorption/desorption (μg P g$_{sediment}^{-1}$) on the y-axis (Eqn. 2.1). Where $P_{initial}$ is the inlet aqueous phosphorus concentration (μg P L^{-1}), P_{final} is the final aqueous phosphorus concentration (μg P L^{-1}) after equilibrium has been established between the sediment and water, V is the volume of water used (L), and wt_{sed}^{-1} is the mass of sediment used in the experiment (g).

$$P_{Ads} = \left(P_{initial} - P_{final}\right) * V * (wt)_{sed}^{-1} \qquad (2.1)$$
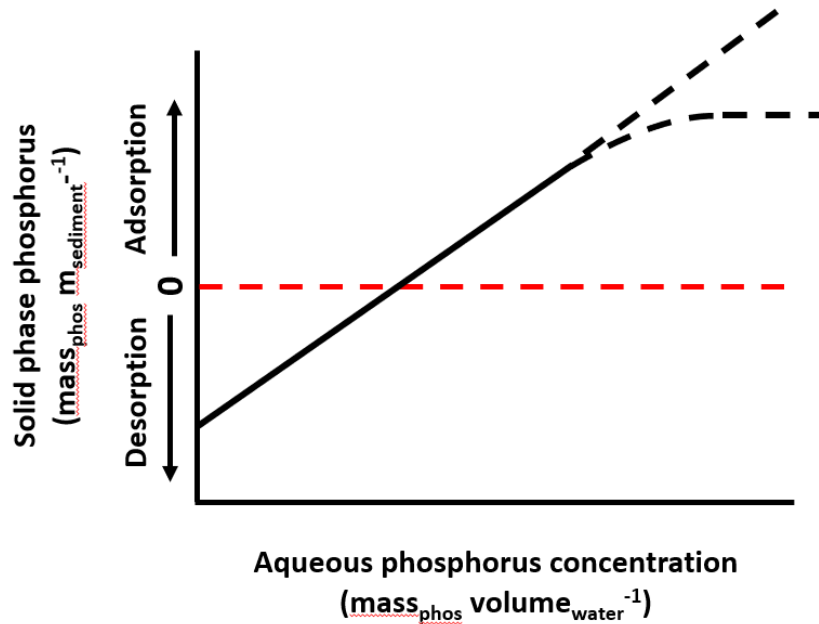
Figure 2-4: Solid phase phosphorus concentration as a function of initial concentration (modified from Dunne et al., 2005). The aqueous phase concentration at which the rates of adsorption and desorption are equal is the equilibrium phosphate concentration (EPC_0). Phosphorus desorbs from sediment to the water column at aqueous phase contrations below the EPC_0; it adsorbs to sediment at concetrations above the EPC_0. Higher EPC_0 values increase the range of aqueous phase phosphorous concentrations 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. Dashed black lines represent the potential for various sorption models (e.g., Langmuir, Freudlich, Tempkin).

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The slope of the fitted line in Fig. 2-4 represents the absorption coefficient; steep slopes represent greater ability to buffer phosphorus (Froelich, 1988). Buffer diagrams are used to examine mechanistic behavior of sediments (Froelich, 1988). However, solution pH, ionic strength, temperature, ratio of sediment to solution and solution chemistry can all influence sediment sorption characteristics (Klotz, 1988; Lucci et al., 2010; Bhadha et al., 2012), therefore results from sorption studies (phosphorus isotherms) cannot be directly translated to ecosystems, though they can nonetheless provide a relative proxy for this process (Barrow, 1983).

The equilibrium phosphate concentration (EPC_0) is defined as dissolved phosphorus concentration at which the rates of adsorption and desorption are equal. If the aqueous concentration is greater than the EPC_0, sediment will adsorb excess aqueous phosphorus to reestablish the aqueous concentration at the equilibrium value; while if the aqueous concentration is less than the EPC_0, sediment will release phosphate to the water column to re-establish the EPC_0. Higher EPC_0 values increase the range of dissolved phosphorus concentrations at which release of phosphorus from the sediment to the water column occurs, thereby increasing the availability of this limiting nutrient for algal proliferation. Under this condition, phosphate release from the sediment can be particularly problematic and can be quickly utilized by primary producers, further increasing the phosphorus concentration gradient, and creating a condition that promote more phosphorus release from the sediment thus creating a feedback cycle of phosphate release (Emelko et al., 2016). Due to the inherent risk associated with cyanobacterial growth in drinking water reservoirs, it follows that one potential management option for water utilities would be to develop strategies for managing fine sediment to reduce the risk of cyanobacterial growth.

2.4 Fine sediment and water quality management in reservoirs

Several BMPs have been proposed to mitigate the external and internal loading of P from sediment to aquatic systems. Current approaches to mitigate cyanobacterial blooms are largely used to reduce external loading (Binkley & Brown, 1993; Hao et al., 2016) using erosion control measures, enhanced agricultural methods, advanced wastewater treatment plant upgrades, establishment of riparian buffers, construction of wetlands, and reductions in fertilizer application (Marsden, 1989; Rahman &

Bakri, 2010; Jiao et al., 2015). Notably, these BMPs have had varied success and require refinement in their design and application. It should be further underscored that while such practices are predominantly focused on reducing phosphorus transport and delivery to receiving streams, relatively fewer strategies are available for limiting phosphorus availability and mobility within the water column (i.e., internal loading).

The suppression of fine sediment-associated phosphorus release to the water column may be achieved through several individual or combined treatment strategies. The frequent removal of deposited fine sediment from the reservoir (i.e., reservoir dredging) is one option for limiting phosphorus release to the water column (US EPA, 2014). This strategy is an effective measure for reducing the mass of bioavailable phosphorus in a reservoir, however this benefit is temporary due to subsequent sediment accumulation inside the reservoir after dredging. In practice, reservoir dredging may be financial and/or operationally challenging depending on reservoir design and operation constraints as well as accessibility.

Alternative reservoir management strategies would be to oxidize fine sediment using either (1) aeration by adding dissolved oxygen to the water column, or (2) injecting $Ca(NO_3)_2$ to bottom sediments to stimulate denitrification (Ripl, 1976). It has been shown that oxidized sediment in shallow freshwater systems can prevent cyanobacterial blooms across a trophic range (Molot et al., 2021). This may be operationally difficult depending on the reservoir flowrate and depth, incoming sediment redox state, mass of sediment entering the reservoir, and oxygen demand of the reservoir.

For reservoirs where internal phosphorus loading has prolonged eutrophication despite significant phosphorus reductions from external loading sources, the potential use of coagulants for the suppression of phosphorus release has shown promise as a management practice (Cooke et al., 2005). This practice has been used in lakes impacted by stormwater runoff and in one in-line reservoir with the intent of minimizing turbidity for managing drinking water source quality (Table 2-1).

Table 2-1: Historical examples of lakes, storm water management ponds (SWM's), and drinking water reservoirs (DWR's) using chemical coagulation for improving water quality. Quality improvement included **(1)** phosphorus inactivation (PI), or **(2)** turbidity rection. NR indicates data that was not reported in the literature. (A) Harper et al., 1999; (B) Wagner et al., 2017; (C) Welch & Cooke, 1999; (D) (National Academies of Sciences, Engineering, and Medicine, 2018); (E) Huser et al., 2011.

Lake	Class	Treatment goal	Coagulant type	Coagulant dose (mg L^{-1})	Initial [TP] (ug L^{-1})	Final [TP] (ug L^{-1})
Ella[A]	SWM	PI	Alum	5-10	232	26
Dot[A]	SWM	PI	Alum	5-10	351	24
Osceola[A]	SWM	PI	Alum	5-10	37	26
Lucerne[A]	SWM	PI	Alum	5-10	100	30
Hamblin (1995)[B]	SWM	PI	Alum	9	454	46
Hamblin (2015)[B]	SWM	PI	Alum	4.5	310	13
Ashumet (2001)[B]	SWM	PI	Alum	4.3	290	100
Ashumet (2010)[B]	SWM	PI	Alum	4	300	60
Long[B]	SWM	PI	Alum	3	163	62
Mystic[B]	SWM	PI	Alum	3.3	555	65
Lovers[B]	SWM	PI	Alum	5	116	24
Stilwater[B]	SWM	PI	Alum	5	290	38
Herring[B]	SWM	PI	Alum	5	357	21
Great[B]	SWM	PI	Alum	5	57	32
Lovell's[B]	SWM	PI	Alum	2.5	167	35
Cliff[B]	SWM	PI	Alum	3.8	87	12

Table 2-2 continued:

Lake	Class	Treatment goal	Coagulant type	Coagulant dose (mg L^{-1})	Initial [TP] (ug L^{-1})	Final [TP] (ug L^{-1})
Erie[C]	Lake	PI	Alum	NR	115	86
Campbell[C]	Lake	PI	Alum	NR	49	23
Long (North)[C]	Lake	PI	Alum	NR	42	24
Long (South)[C]	Lake	PI	Alum	NR	31	16
Pattison (North)[C]	Lake	PI	Alum	NR	28	8
Pattison (South)[C]	Lake	PI	Alum	NR	30	32
Long(2)[C]	Lake	PI	Alum	NR	63	19
Wapato[C]	Lake	PI	Alum	NR	46	61
Pickerel[C]	Lake	PI	Alum	NR	35	47
Kenisco Reservoir[D]	DWR	TR	Alum	NR	/	/
Cedar[E]	Lake	PI	Alum	NR	43	24
Isles[E]	Lake	PI	Alum	NR	66	46
Calhoun[E]	Lake	PI	Alum	NR	36	15
Harriet[E]	Lake	PI	Alum	NR	32	18

While the use of coagulants to inactivate phosphorus in lakes impacted by agricultural and urban runoff, it has not been widely used by drinking water reservoir managers (Cooke et al., 2005). Coagulant is sometimes added to the in-line Kenisco Reservoir in New York City to reduce turbidity in the source water (National Academies of Sciences, Engineering, and Medicine, 2018). However, the historical precedence of only applying single alum doses to lakes may have limited utility for phosphorus inactivation in reservoirs with high sediment loads. Cooke et al (2005) reported that reservoirs with relatively high sediment loading would likely require frequent, or continuous coagulant dosing thus be financially prohibitive for water utilities. This practice may provide co-benefits of coagulant and sludge reduction and reduce the total organic carbon (TOC) content entering drinking water treatment plants (Harper et al., 1999).

In a trial to reduce algal biomass in the Eau Galle Reservoir in Spring Valley, Wisconsin, alum was used to control sediment-associated phosphorus release and minimize excessive algal growth (Barko et al., 1990). The reservoir was dosed to control sediment phosphorus release for five years,

and while phosphorus reduction was initially observed in the summer; there was no further effect in subsequent years for the reduction of epilimnetic algal biomass (Kennedy et al., 1987; Barko et al., 1990). Three possible reasons for the failure to reduce algal biomass with alum dosing were (1) high external sediment loading (Barko et al., 1990), (2) previously unaccounted groundwater recharge (Barko et al., 1990), and (3) inadequate dosing based on more current reservoir volume estimates (Cooke et al., 2005). Another study investigated the use of alum-based water treatment residual augmented with powdered activated carbon to sequester biologically available phosphorus in the source water (Carleton & Cutright, 2020). Preliminary results show a ~65 % phosphorus reduction, however this was over a 30-day trial, which may be practical for large in-line reservoirs but is less practical for smaller off-line reservoirs with low residence times (hours to days). Accordingly, while the potential for coagulant addition to mitigate bioavailable phosphorus in reservoirs has been demonstrated (Table 2-1), further study and verification of this practice for use by water utilities with off-line reservoirs is warranted.

Chapter 3

Coagulant addition for managing sediment-associated phosphorus bioavailability to prevent cyanobacterial blooms in drinking water reservoirs

3.1 Introduction

Surface water is a critically important resource, accounting for approximately 61% of all public use water within the U.S. (Dieter et al., 2018). It is facing increasing threats within North America (Winter et al., 2011) and around the world (Hallegraeff, 1993), however, due to increases in cyanobacterial bloom occurrence. Algae blooms, especially cyanobacteria, are one of the biggest threats to water quality and the provision of safe drinking water in Canada and globally. High densities of cyanobacteria can increase source water turbidity, thereby increasing chemical coagulant demand, settling time, and disinfectant/oxidant demand; they can also clog filters and decrease filter run times (Crittenden, 2012). Notably, some cyanobacteria have the potential to form unpleasant taste and odor compounds, such as geosmin and methyl-isoborneol (MIB), which although not toxic, result in a foul scent and can lead to customer complaints (McGuire et al., 1981; Simpson & MacLeod, 1991). Cyanobacteria can also release metabolites that are toxins of human health and environmental concern, which may require advanced treatment beyond that typically found in most conventional treatment plants. These toxins include the hepatotoxins microcystin and cylindrospermopsin, and the neurotoxin anatoxin-a; while all have implications for human health, only microcystin is currently regulated in Canada (Health Canada, 2012).

Environmental conditions including temperature, pH, light availability, and nutrient availability affect the survival, growth, and proliferation of cyanobacteria in freshwater systems (Paerl et al., 2001; Watson et al., 2008; U.S. Environmental Protection Agency, 2012a). The availability of key nutrients is especially critical to cyanobacteria and more broadly algae proliferation in lakes, reservoirs, and even drinking water treatment plants (Håkanson et al., 2007; Metcalf & Codd, 2014). Phosphorus is considered the limiting nutrient for primary productivity in freshwater systems (Schindler, 1977; Barlow-Busch et al., 2006). Recently, a study across a range of ecozones in Canada

reported that total phosphorus (TP) concentrations of 10 to 30 µg TP L^{-1} associated with good ecological condition with respect to benthic algal abundance, diatom composition, and macroinvertebrate composition, while higher nutrient concentrations in aquatic systems are associated with increased algal abundance, loss of sensitive benthic macroinvertebrate taxa, and an increase in benthic diatom taxa indicative of eutrophication (Chambers et al., 2012). Thus, a total phosphorus concentration of 30 µg P L^{-1} is a commonly cited threshold for eutrophication (Schindler, 1977; Barlow-Busch et al., 2006; Chambers et al., 2012). This threshold is a benchmark rather than a predictive tool; cyanobacteria can still proliferate in aquatic systems with total phosphorus concentrations less than 30 µg P L^{-1} (Winter et al., 2008; Vuorio et al., 2020), though it is considered less likely to occur. While the dynamics of cyanobacteria bloom formation are not well defined (Holland & Kinnear, 2013; Ma et al., 2014), reducing phosphorus availability in the drinking water reservoirs should decrease the potential for bloom occurrence (Paerl et al., 2016; Schindler et al., 2016). Thus, if drinking water reservoir managers can limit water column total phosphorus concentrations so that they are below a threshold of approximately 30 µg P L^{-1}, the potential for cyanobacteria proliferation and the associated production of toxins and unpleasant taste and odor compounds may be mitigated (Watson et al., 2008; U.S. Environmental Protection Agency, 2012a).

The total phosphorus concentration is comprised of total dissolved phosphorus (TDP) and total particulate phosphorus (TPP). These forms are operationally defined by passing water through a 0.45 µm nominal porosity filter, which will retain the TPP and allow the TDP to pass through (American Water Works Association, 2017). TDP is readily bioavailable and made up of orthophosphate and colloidal bound orthophosphate (Reynolds & Davies, 2001). Sequential extraction procedures are used to fractionate TPP to forms: (1) non apatite inorganic phosphorus (NAIP), (2) apatite phosphorus (AP), and (3) organic phosphorus (OP). Bioavailable phosphorus in the NAIP fraction is typically bound to iron, aluminum, and manganese; the redox sensitive compounds comprising NAIP can desorb into the water column at certain environmental conditions, making them bioavailable. AP is a particulate phosphorus form that is bound to calcium carbonate and is generally considered unavailable for biological uptake. While the OP fraction is not directly bioavailable, it can become bioavailable after hydrolysis or mineralization (Emelko et al., 2016).

Fine sediment (< 63 µm in size) is widely recognized as the primary vector for phosphorus transport in aquatic systems (Stone & English, 1993; Auer et al., 1998; Davies-Colley & Smith, 2001;

Dodds, 2003). Land disturbances such as agriculture and urbanization in watersheds accelerate the transfer of sediment-associated phosphorus from terrestrial to aquatic systems and downstream propagation to drinking water reservoirs (Froelich, 1988; Stone & English, 1993; Jarvie et al., 2005). The deposition of sediment and associated phosphorus can influence nutrient dynamics in reservoirs through the release of phosphorus from bottom sediment to the water column, a process known as internal loading (Marsden, 1989). Fine sediment can enter upstream receiving surface waters by either (1) point source loading, or (2) non-point source loading. Point source discharges often are continuous flows from municipal or industrial waste streams such as an industrial wastewater runoff pipe. In contrast, non-point source discharges tend to be discontinuous, connected to agriculture, forests, or unusual precipitation. In North America, the vast majority (84%) of phosphorus entering the aquatic surface waters is attributed to non-point loading from such sources as croplands, pastures, rangelands, and forests (Carpenter et al., 1998). The challenge of external phosphorus loading to water supplies is expected to be increasingly severe, due to continued natural (i.e., wildfires, hurricanes, floods, and precipitation changes) and anthropogenic (e.g., urbanization, agriculture, industrial) landscape disturbances which alter hydrological processes that can increase sediment erosion (Smith et al., 2011; Intergovernmental Panel on Climate Change (IPCC), 2013; Emelko et al., 2016).

Phosphate release from fine sediment (i.e., internal loading) has been identified as a main driver for algal growth in many freshwater systems; it can lead to partial or delayed responses to external loading reduction efforts (Nürnberg, 1988; Lehman, 2010; Orihel et al., 2015). Phosphate release from fine sediment to the water column is influenced by: sediment grain size and geochemistry, redox conditions, pH, competitor ions, and sediment phosphate concentration (Boström & Pettersson, 1982). Phosphate adsorption/desorption to/from sediment is a two-step process. Phosphate release from fine sediment to the water consists of (1) slow solid-state diffusion and (2) rapid surface sorption (Froelich, 1988). The direction of the adsorption/desorption process can be determined by plotting the solid phase phosphorus (i.e., mass of phosphorus adsorbed per mass of sediment) as a function of aqueous phosphorus concentration. The aqueous phase concentration at which the rates of adsorption and desorption are equal is defined as the equilibrium phosphate concentration (EPC_0). If the dissolved phosphorus concentration is higher than the EPC_0, sediment will adsorb dissolved phosphorus. However, when the aqueous phosphorus concentration

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is below the EPC_0, sediment will desorb phosphate to the water column, thereby potentially stimulating algae proliferation.

Increasing phosphorus loading from watersheds and its connectivity to potential cyanobacterial growth highlights the need for water utilities to implement sediment management strategies that focus on drinking water source quality. At present, drinking water reservoirs are not typically designed to manage internal loading of phosphorus and while this process has been well studied in lakes and on-line reservoirs (Welch & Cooke, 1999; Wagner & Adrian, 2009), investigations of management strategies (e.g., coagulant addition) to prevent phosphorus release from bottom sediments (i.e., phosphorus inactivation) to the mitigate the proliferation of cyanobacteria in raw water storage reservoirs are scant (Pütz & Benndorf, 1998). Thus, there is an opportunity to develop fine sediment management approaches that mitigate or prevent releases of bioavailable phosphorus to the water column and integrate them into drinking water source protection and supply management plans, as well as climate change adaptation strategies. Minimizing the effects of phosphorus release to the water column of drinking water reservoirs can be accomplished by (1) reducing the external loading of sediment to the source water, and/or (2) reducing the potential for internal loading of phosphorus to the reservoir water column (Wagner & Adrian, 2009). While there is abundant literature on small and large lake-scale BMPs designed to reduce the external loading of fine sediment (e.g., erosion control measures, enhanced agriculture practices, wastewater treatment plant upgrades), consideration of strategies for suppressing internal loading within drinking water reservoirs remains limited (U.S. Environmental Protection Agency, 2001; Canadian Council of Ministers of the Environment, 2016). Accordingly, the overall goal of this investigation was to develop fine sediment management strategies to prevent the proliferation of cyanobacteria in raw (i.e., untreated) water storage reservoirs.

3.2 Materials and methods

To investigate strategies for fine sediment management, a series of lab- and field-scale analyses were conducted to (i) describe P release from fine sediment in a drinking water reservoir, (ii) characterize its availability for biological uptake, (iii) evaluate P inactivation by application of coagulants ($FeCl_3$, alum, poly-aluminum chloride) commonly used in drinking water treatment, and (iv) evaluate the combination of strategically-timed reservoir dredging and coagulant application on P inactivation and turbidity reduction.

3.2.1 Hidden Valley Reservoir

Located in southwestern Ontario, Canada, the Region of Waterloo (ROW) provides drinking water to approximately 600,000 people. Its current drinking water demand is approximately 320 million liters per day (MLD). Approximately 20 to 25% of that demand is from surface water and the remainder is ground water (Region of Waterloo, 2021). To meet the surface water demand, ROW constructed the Hidden Valley Reservoir (HVR), an offline reservoir located on the bank of the urban and agriculture impacted Grand River (Fig. 3-1). The HVR is a linear flow engineered reservoir with a storage capacity of approximately 150 million liters (ML). Water enters HVR via the low-lift pump at the riverbank, before flowing through four cells in the HVR in an over-under pattern with a retention time of approximately two days, before being pumped from the high-lift pump approximately 10 kilometers to the Mannheim Water Treatment Plant (MWTP). This reservoir is designed to store source water during periods of water quality deterioration in the Grand River that are typically associated with high flow periods and sometimes spills. Some solids also settle out of the water column before the water is pumped to the MWTP. Notably, more proliferation of algae is evident at points in the reservoir that are farther from the intake (i.e., Cells #3 and #4, especially Cell #4).

Figure 3-1: Aerial view of the Hidden Valley Reservoir (HVR) in Waterloo, Ontario, Canada from which the sediment and source water investigated herein were obtained. Source water is pumped from the Grand River to the first of four cells (i.e., Cell #1) located on the right-hand side of this image. Water flows through the cells in an over-and-under configuration. Residence time in the HVR is approximately 72 hours (Region of Waterloo, 2021). Notably, more proliferation of algae is evident at points in the reservoir that are farther from the intake (i.e., Cells #3 and #4, especially Cell #4).

Historical HVR intake total and dissolved phosphorus are presented as a time series in Fig. 3-2. These data show that phosphorus levels vary both seasonally and annually. Elevated total and dissolved levels of phosphorus in the HVR intake correspond to fluctuations in Grand River discharge (i.e., flow). The mean and standard deviation of total and dissolved phosphorus concentrations are (43.3 µg P L^{-1}, SD = 44.2, n = 463) and (15.3 µg P L^{-1}, SD = 20.2, n = 486), respectively. These data are essential to interpreting the data collected during the reservoir sediment phosphorus adsorption/desorption experiments.
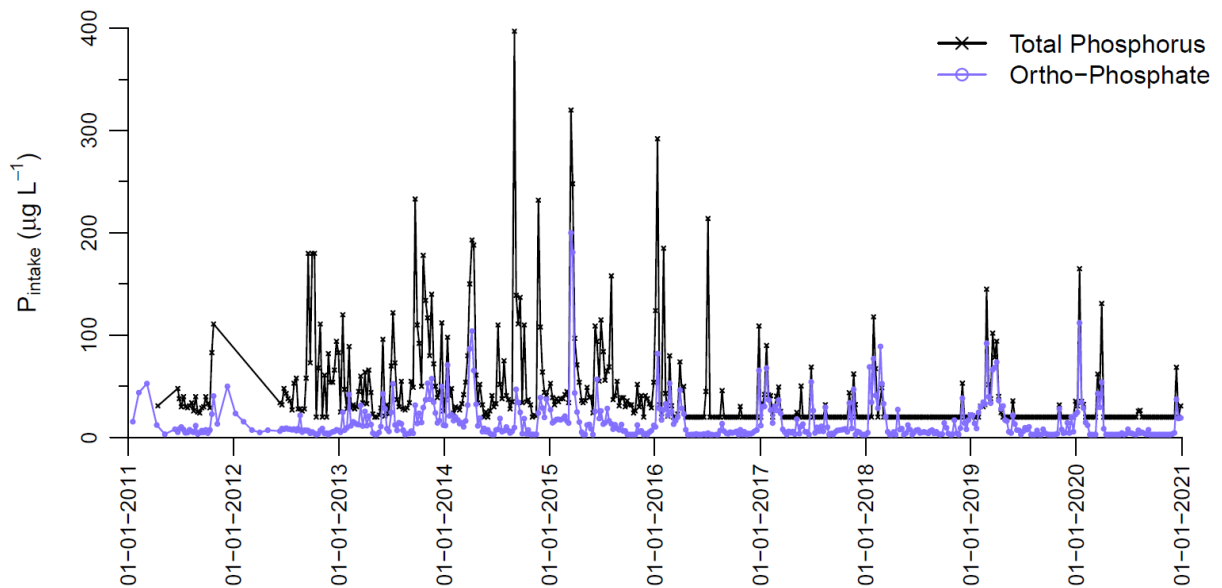
Figure 3-2: Total and dissolved phosphorus concentrations measured at the HVR intake from 2011 to 2021. The commonly accepted threshold for "good" water quality, i.e., the concentration at which algal proliferation is considered more likely, is 30 µg P L^{-1}. All values reported as below detection limit (TP: 20 µg P L^{-1} and ortho-P: 3 µg P L^{-1}) were plotted at the detection limit.

3.2.2 Sediment collection

Reservoir sediment was collected in Cell #1 of the HVR in September 2020 immediately after the cell was drained and prior to dredging (see photos in Appendix G). Samples were collected at three equidistant locations in the Cell to ensure representative materials were collected. Only the surface layer (0 to 5 cm) of deposited sediment was collected because it is the layer most important to the release of phosphorus at the sediment water interface (Fig. 3-3). This layer of loosely bound fine sediment (sometimes referred to as surficial fine-grained laminae (Droppo & Stone, 1994) deposits at low flow and can release phosphorus to the water column (and potentially promote algae proliferation) when the aqueous phase phosphorus concentration is below the EPC$_0$. During sediment collection, an oxidation-reduction potential (ORP) probe was used to measure the redox conditions in the top 10 cm of the sediment deposit. The measurements were read in triplicate at each sample location and reported in the results section.

Figure 3-3: Deposited sediment collected from HVR Cell#1 in September 2020. Two distinct layers are evident: (1) a thin top layer of loosely bound fine sediment that deposits at low flow (sometimes referred to as surficial fine-grained laminae (Droppo & Stone, 1994) and (2) a more consolidated bottom layer. The cell was dredged in October 2019. Approximately 3 to 4 inches of sediment deposited in the cell during the subsequent year.

3.2.3 Sediment composition

The major element composition of sediment (Si_2O, Al_2O_3, CaO, MgO, Na_2O, K_2O, Fe_2O_3, MnO and P_2O_5) was measured with X-ray fluorescence at a commercial laboratory (Act Labs, Burlington, ON, Canada) and the results are reported as percent dry weight (Mudroch & Duncan, 1986). Analytical accuracy was confirmed using Canadian Reference Standards AGV-1, MRG-1, NCM-N, GSP-1, and SY-3; the results were accurate to 1%. Particulate phosphorus forms were evaluated using a sequential extraction method to fractionate particulate phosphorus into five operationally defined forms

(Petterson et al., 1988). Non-apatite inorganic phosphorus (NAIP) which is considered the most bioavailable form (DePinto et al., 1981) is defined as the sum of three reactive phosphate fractions: loosely sorbed P (1.0 M NH_4Cl–P extractable phosphorus), reductant soluble phosphorus (0.11 M $NaHCO_3.Na_2S_2O_4$ extractable phosphorus), and metal oxide bound phosphorus (1.0 M NaOH extractable phosphorus) (Boström & Pettersson, 1982). Apatite phosphorus (AP) is the 0.5 M HCl extractable phosphorus fraction bound primarily to Ca and Mg carbonates and represents phosphorus forms that are sensitive to low pH. This fraction is assumed to consist mainly of apatite phosphorus (natural and detrital), carbonate bound phosphorus, and traces of hydrolysable organic phosphorus. Thus, the AP fraction is considered a relatively stable, permanent sink of phosphorus in sediments (Kaiserli et al., 2002). The organic fraction (OP) is extracted using hot 1 M NaOH (85°C) (Emelko et al., 2016).

3.2.4 Evaluation of phosphorus release potential of fine sediment

Sediment samples collected from HVR were used to conduct batch experiments to determine the equilibrium phosphorus concentration (EPC_0). The EPC_0 is a measure of the potential of sediments to adsorb or release SRP depending on the ambient SRP concentrations in aquatic systems (House & Denison, 1998; 2000). A range of water matrices was formed using HVR raw water samples collected on February 9, 2021 (Raw, 100, 200, 400, and 600 µg P L^{-1}). A 25 mL aliquot of each phosphorus concentration (Raw, 100, 200, 400, and 600 µg P L^{-1}) was added in triplicate to 0.25 g of sediment in 50-mL centrifuge tubes. Centrifuge tubes were placed on the shaker table and shaken at 50 rpm at room temperature for 18-hours. An aliquot of 15-mL was filtered (0.45 µm) into scintillation vials and refrigerated. Concentrations of SRP were analyzed using a Technicon Autoanalyzer using the ammonium molybdate/stannous chloride method in the Biogeochemistry Lab in Environment 2 at the University of Waterloo for 2021 (Environment Canada, 1979). While the ratio of 0.25 g of sediment to 25 mL (i.e., 10:1 g L^{-1}) is the most accepted ratio for conducting bench scale EPC_0 experiments (Taylor & Kunishi, 1971; Froelich, 1988), scalability may require further analysis. For example, the HVR Cell #1 had approximately four inches of deposited sediment, yielding a sediment to water ratio of approximately 6.7 grams/liter, which suggests that the buffering capacity of the HVR may be overestimated (Appendix I). However, the loosely sorbed, readily bioavailable fine sediment depth is

likely much less than the four inches of deposited sediment. Further analysis is required to investigate the impact of sediment depth and ratio of sediment to water.

For the experiments involving coagulant addition, ferric chloride ($FeCl_3$), aluminum sulphate ($Al_2(SO_4)_3 \cdot 14H_2O$), or poly-aluminum chloride ($Al_a(OH)_b(Cl)_c(SO_4)_d$) coagulant was added to centrifuge tubes at a range of doses described below. Stock solutions of 5000 µg L^{-1} of $FeCl_3$ and aluminum sulfate were prepared to minimize variability and dosing volume into the centrifuge tubes to ensure the ratio of 0.25 grams of sediment to 25 mL of water was maintained. Sorption experiments were conducted using doses of 0, 10, 20, 25, 30 mg L^{-1} of $FeCl_3$ and poly-aluminum chloride, and 0, 20, 30, 40, 50 mg L^{-1} of alum. After coagulant addition, the centrifuge tubes were agitated for approximately ten seconds and then left in a test tube rack for 10 minutes so that any suspended sediment could settle. All samples were then filtered through a 0.45 µm filter (Whatman Puradisk nylon syringe filter, GE Health Sciences) into acid-washed, triple rinsed glass scintillation vials. Vials were capped and stored into a fridge at approximately 10°C prior to SRP analysis. Samples were removed from the refrigerator approximately one hour before SRP analysis was conducted on a calibrated Technicon Auto-analyzer II (Technicon Instruments Corp., Tarrytown, N.Y.). SRP concentrations were measured colormetrically using the stannous chloride ammonium molybdate procedure (Environment Canada - Water Quality Branch, 1979). All SRP concentrations are reported as PO_4^{3-}.

The phosphorus sorption capacity was calculated according to the following equation:

$$ q = \frac{(C_0 - C_E)}{M * V} \tag{3.1} $$

where q is the mass of phosphorus sorbed per mass of sediment (µg g^{-1}), C_0 and C_E are the initial and equilibrium aqueous phosphorus concentrations (µg P L^{-1}), M is sediment mass in the centrifuge tube (g), and V is the volume (L) of aliquot in the centrifuge tube.

3.2.5 Sediment removal to evaluate impact of reservoir dredging

To evaluate the impact of reservoir dredging, bench scale experiments were conducted to evaluate the phosphorus reduction (i.e., final aqueous phosphorus concentration – initial phosphorus concentration) achieved by adding chemical coagulants $FeCl_3$, alum ($Al_2(SO_4)_3 \cdot 14H_2O$), and poly-

aluminum chloride $(Al_a(OH)_b(Cl)_c(SO_4)_d)$ to the water matrix in the absence of fine sediment. The process that was followed was the same as described above.

A range of water matrices were formed using HVR raw water samples collected on February 9, 2021 (Raw, 100, 200, 400, and 600 µg P L^{-1}). A 25 ml aliquot of each phosphorus concentration (Raw, 100, 200, 400, and 600 µg P L^{-1}) was added in triplicate. Centrifuge tubes were placed on the shaker table and shaken at 50 rpm at room temperature for 18-hours. An aliquot of 15-mL was filtered (0.45 µm) into scintillation vials and refrigerated. Concentrations of SRP were analyzed using a Technicon Autoanalyzer using the ammonium molybdate/stannous chloride method (Environment Canada, 1979).

For the experiments involving coagulant addition, solutions of FeCl$_3$ and aluminum sulfate, and one industrial sample of poly-aluminum chloride were added to the centrifuge tubes at a range of doses described below. Stock solutions of 5000 µg L^{-1} of ferric chloride and aluminum sulfate were created to minimize variability and dosing volume into the centrifuge tubes to ensure 25 mL of water was maintained. Sorption experiments were conducted using doses of 0, 10, 20, 25, 30 mg L^{-1} of FeCl$_3$ and poly-aluminum chloride, and 0, 20, 30, 40, 50 mg L^{-1} of aluminum sulfate were used. After coagulant addition, the centrifuge tubes were agitated for approximately 10 seconds and then left in a test tube rack for 10 minutes so any suspended sediment could settle. All samples were then filtered through a 0.45 µm filter (Whatman Puradisk nylon syringe filter, GE Health Sciences) into acid-washed, triple rinsed glass scintillation vials. Vials were capped and stored in a refrigerator at approximately 4°C prior to SRP analysis. Samples were removed from the refrigerator approximately one hour before SRP analysis was then conducted on a calibrated Technicon Auto-analyzer II (Technicon Instruments Corp., Tarrytown, N.Y.). SRP concentrations were measured colormetrically using the stannous chloride ammonium molybdate procedure (Environment Canada - Water Quality Branch, 1979). All SRP concentrations are reported as PO_4^{3-}.

3.2.6 Coagulant dose selection

Simple or polymerized mineral salts and polymers are comprised of positively charged cations and/or polymer chains, and are typically added during drinking water treatment to destabilize and aggregate particles by one or more of four possible mechanisms: (i) double layer compression (which is not

relevant here because increasing ionic strength is not practiced in water treatment), (ii) adsorption and charge neutralization, (iii) enmeshment in precipitate (often referred to as "sweep flocculation"), and (iv) adsorption and inter-particle bridging, which is relevant to polymers only because they are very long chain macromolecules whose size is greater than the distances between particles in suspension during coagulation (Amirtharajah & Mills, 1982; Duan et al., 2003; Sharp et al., 2004; Edzwald & Haarhoff, 2011; Davis & Edwards, 2014). Thus, coagulation is commonly described as involving two primary mechanisms: (1) charge neutralization of negatively charged particles by adsorption of positively charged ions and polymers and (2) enmeshment of particles in precipitated metal hydroxide solids (Amirtharajah & Mills 1982; Bache et al., 1999; Pernitsky & Edzwald 2006). In the case of coagulation with metal salts, these coagulation mechanisms depend on pH and metal (e.g., aluminum, iron) concentration (Pernitsky & Edzwald 2006). $FeCl_3$, aluminum sulfate, and poly-aluminum chloride are ideal coagulants for surficial raw water as they rapidly hydrolyze when mixed with water (<10 seconds), forming insoluble participates that destabilize solids suspended in the raw water and neutralize their charge, enabling them to aggregate (Crittenden et al., 2012). As the destabilized particles aggregate and precipitate (i.e., flocculate) they also typically settle. The solubility diagrams for iron and aluminum are presented in Appendix H and provide some insight regarding likely optimal coagulant doses that depend on source water pH. Given that Grand River typically ranges from 7.9 to 8.3, optimal coagulant doses (i.e., those achieving sweep flocculation) is between 5 and 30 mg L^{-1} were expected for $FeCl_3$ and between 30 and 50 mg L^{-1} were expected for alum. Poly-aluminum chloride is manufactured and sold as a patented chemical with no published solubility curves, however water treatment industry applications typically range between 10 and 30 mg L^{-1} (Crittenden et al., 2012).

3.2.7 Quality control

A set of phosphorus standards (0, 25, 50, 100, 200, 400, 800 µg P L^{-1}) were used to create a standard calibration curve. Samples were analyzed when the coefficient of determination (R^2) for the calibration curve was greater than 0.995. To ensure there was no analytical drift during SRP analysis, standard solutions were evaluated after every fifteenth sample. If there was a greater than 5% deviation from the known concentration, that section of samples would be re-run (Appendix F). All

samples were run in triplicate (or n >3) and were plotted as the mean value with error bars of ± 1 standard deviation.

3.3 Results and discussion

3.3.1 Characterizing reservoir sediments and phosphorus sorption

To evaluate the phosphorus sorption behavior (release potential) of reservoir sediment, samples were collected from all four reservoir cells in spring 2013. Grain size characteristics of deposited sediment in each of the reservoir cells are summarized in Table 3-1. A decrease in grain size of deposited sediment along the length of the reservoir might be expected as a result of sediment fining, which is a process by which particles settle differentially based on their size and density (Froelich, 1988). This phenomenon is frequently observed as water flows from rivers to lakes and reservoirs as detailed in Yang, 2018. A clear decrease in grain size of deposited sediment along the length of the reservoir was not observed (Table 3-1). This observation was not surprising because the sediment entering the reservoir is already largely fine sediment (i.e., < 63 µm in size), as evident in Table 3-1, which indicates that approximately 80% of the sediment deposited in each of the four reservoir cells is fine grained. Moreover, it is important to note that some flocculation of the suspended sediment occurs naturally and may impact conventional grain size analysis, which would need to involve sonication to evaluate the particle size distribution of primary particles in the system. Such an analysis was beyond the scope of the present investigation. Regardless, the presence of a significant fraction of fine sediment within all four cells of the HVR indicates that there is potential for internal loading of phosphorus to the water column in each of the cells.

Table 3-1: Average grain size distribution of fine sediment collected from reservoir cells in the HVR May 2013 (mean ± 1 SD, n=9; adapted from Crumb, 2014). Fine grain sediment is operationally defined as sediment less than 64 µm in size. The majority of sediment collected in the HVR is fine grained, which indicates a high likelihood for phosphorus external loading, and the potential for internal sediment-associated release.

HVR Cell Location	Percentage of grains at or below a specific diameter (µm) = Dx							
	10%		50%		80%		90%	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Cell1	5.03	0.81	24.81	3.45	65.31	12.48	122.56	35.51
Cell2	4.73	0.22	21.14	1.13	52.32	2.23	92.85	7.53
Cell3	5.14	0.54	23.12	1.80	53.96	4.40	87.71	9.63
Cell4	5.16	0.22	25.74	0.43	63.52	0.58	111.42	1.17

The geochemical composition of sediment collected from the four HVR cells was analyzed to determine if the minerology indicated any differences between the type of sediment deposited within each cell. The key element and mineral composition of the deposited sediment in each of the reservoir cells are summarized in Table 3-2. Here, the distribution of reservoir bottom sediments containing metal oxide (i.e., Al_2O_3, Fe_2O_3, and MnO) fractions that are known adsorptive surfaces that bind phosphorus was generally higher in downstream reservoir cells and consistent with higher levels of bioavailable phosphorus contributing to the higher levels of algae proliferation in Cells #3 and #4. Bioavailable particulate phosphorus forms (i.e., NAIP) are widely understood to preferentially bind to these metal oxide surfaces. Thus, these geochemically analyses are consistent with higher levels of bioavailable phosphorus contributing to the higher levels of algae proliferation that have been anecdotally observed in downstream reservoir cells (i.e., Cells #3 and #4 in this study). Furthermore, there was a higher level of chlorite clay observed in Cell #4. In the presence of metals such as Fe, Mn, and Al, chlorites have phosphorus sorbing metal oxy-hydroxides on their surfaces (Britannica, 2018; Froelich, 1988; Reynolds & Davies, 2001; Withers & Jarvie, 2008). These are closely associated with the redox sensitive NAIP, which is the most biologically available type of particulate phosphorus. This observation suggests that as the potential for phosphorus release increases as water flows through the reservoir from Cell #1 to Cell #4, there can be a concurrent increase in the ambient dissolved phosphorus concentrations that can promote the growth of cyanobacteria and other algae, as indicated in Fig. 3-1.

Table 3-2: Mean metal and mineral concentration of fine sediment collected from reservoir cells in the HVR May 2013 (mean ± 1 SD, n=9) (Crumb, 2014). Chlorites contain metals such as Al, Mn, and Fe in their lattice. As the chlorite % increases through the cells, so too should the individual metal concentrations. Metal elements Al, Mn, and Fe form oxy-hydroxides which increase the potential for phosphorus release.

	Cell	1		2		3		4	
Metals	**Unit**	**Avg**	**±SD**	**Avg**	**±SD**	**Avg**	**±SD**	**Avg**	**±SD**
Al	%	1.33	0.2	0.5	0.1	2.47	0	2.63	0.1
Mn	ppm	1142	42.7	1233	49.3	1740	144.2	1697	35.1
Fe	%	2.75	0.1	2.76	0.1	3.26	0.1	3.34	0
Minerals	**Unit**	**Avg**	**±SD**	**Avg**	**±SD**	**Avg**	**±SD**	**Avg**	**±SD**
Chlorite	%	1.7	0.3	1.7	0.1	1.7	0.1	2.3	0.2

To evaluate the phosphorus sorption behavior (release potential) of reservoir sediment, samples were collected from all four reservoir cells in spring 2013. The sediment was equilibrated in ultrapure, Type 1 MilliQ[TM] water (specific resistance of 18.2 MΩ.cm) with various added dissolved phosphorus concentrations, and the solid phase phosphorus concentrations were analyzed; the results of these analyses are presented in Fig. 3-4. The lowest EPC_0 values were associated with sediments collected in Cells #1 and #2 (47 and 35 µg P L^{-1}, respectively); EPC_0 increased to 108 µg P L^{-1} and 407 µg P L^{-1}, in Cells #3 and #4, respectively.
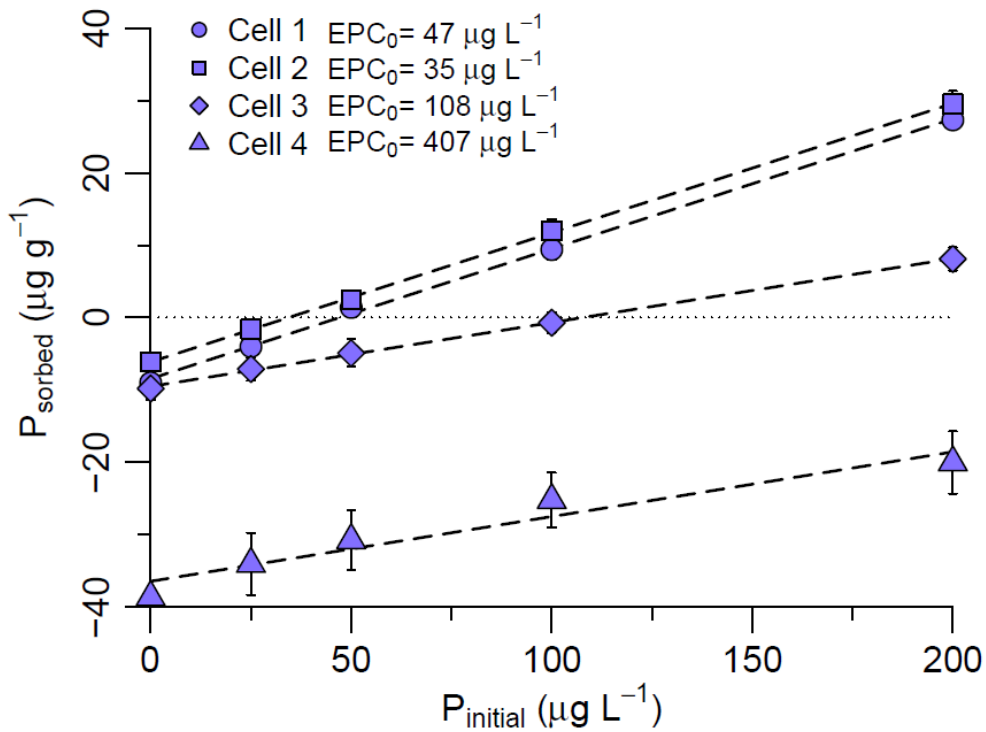
Figure 3-4: Phosphorus adsorption/desorption to ultrapure Type 1 MilliQ™ water by sediment collected from the four HVR cells in May 2013 (mean ± 1 SD, n=9 at each aqueous phase concentration; adapted from Crumb, 2014). EPC_0 for each Cell is indicated in the legend. Based on these values (i.e., higher EPC_0 values), sediments from cells #3 and 4 are more likely to release more phosphorus to the water column.

The sorption data show that the release potential of reservoir sediment (indicated by EPC_0) increases with distance across the reservoir from Cell #1 to Cell #4. The increase in release potential is likely related to the phenomenon of sediment fining, which is a process by which particles settle differentially based on their size and density, as discussed above. The accumulation of finer sediment in Cell #4 compared to Cell #1 suggests that as water flows across the reservoir, finer sediment fractions that have lower settling rates relative to coarser materials (e.g., sand) will preferentially settle in the reservoir locations/cells farthest from the inlet, thus creating zones of fine sediment deposition with high phosphorus release potential that can promote more algae proliferation such as that shown in Fig. 3-1. As discussed above, this phenomenon can occur even if it's not immediately

observed in conventional sediment grain size distribution analysis without sonication to ensure evaluation of only primary particles.

In the absence of matrix effects attributable to competitor ions, the equilibrium phosphate concentration (EPC$_0$) increased from 35 to 47 µg P L^{-1} in the first two cells of the reservoir to 108 µg P L^{-1} and 407 µg P L^{-1} in Cells #3 and #4, respectively. Fine-grained sediments can release phosphorus to the water column when aqueous phase phosphorus concentrations are below the EPC$_0$. Thus, these differences in EPC$_0$ confirm that, consistent with the geochemical analyses described above, sediments deposited further along the flowpath in the study reservoir (i.e., in Cells #3 and #4) are both (i) enriched in bioavailable particulate phosphorus (i.e., NAIP) and (ii) have a higher phosphorus release potential than sediments in Cells #1 and #2. Accordingly, this analysis demonstrates that all of the sediments deposited in the reservoir have the potential to release significant amounts of phosphorus to the water column, and sediments deposited in Cells #3 and #4 in particular are management priorities.

Particulate phosphorus forms (i.e., NAIP, AP, and OP concentrations) in the sediment collected from HVR Cell #1 in 2020 were evaluated. Overall, NAIP comprised 57% (817 µg P g^{-1}) of the total particulate phosphorus (1,413 µg P g^{-1}) for Cell 1 at the HVR, followed by AP at 26% (374 µg P g^{-1}) and OP at 16% (222 µg P g^{-1}). Of the three NAIP fractions, the reductant soluble concentration was the highest of the fractions at 58% (472 µg P g^{-1}), followed by the metal oxide bound fraction at 39% (322 µg P g^{-1}), while the loosely sorbed fraction comprised just 3% (23 µg P g^{-1}) of the NAIP.

The HVR particulate phosphorus composition and concentrations were then compared to those reported for lakes and reservoirs of various trophic states (Fig. 3-4). It has been well documented that land use change can influence the forms of total particulate phosphorus fractions in bed sediment (McCallister & Logan, 1978; Stone & Mudroch, 1989; Stone & English, 1993; Fogal, 1995) as well as suspended sediment (Logan et al., 1979; DePinto et al., 1981). The total particulate phosphorus concentration of over 1,400 µg P g^{-1} coupled with a relatively high NAIP concentration of over 800 µg P g^{-1} reflects NAIP and PP concentrations that are among the highest of all reported reservoirs in this study; it is more consistent with reported mesotrophic lakes than raw (untreated) drinking water reservoirs. These values are also consistent with some of the world's most deteriorated river watersheds such as the rivers: Dart, Exe, and Avon (Emelko et al., 2016).

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In the HVR, the majority of the particulate phosphorus fractions were NAIP, specifically the reductant soluble and metal oxide bound forms. These redox sensitive forms are readily bioavailable and are the most likely form of particulate phosphorus to be released into the water column. Given the findings presented in Fig. 3-1, Table 3-1, and Table 3-2, there is evidence to indicate that, of the HVR reservoir cells, Cell #1 is the least likely to promote phosphorus release to the water column. These observations may suggest that the NAIP, AP, and OP concentrations found in the latter cells, particularly Cells #3 and #4, may have even higher particulate phosphorus concentrations. These findings further support substantial potential for phosphorus release from the fine sediment into the water column in the HVR cells, particularly in Cells #3 and #4.
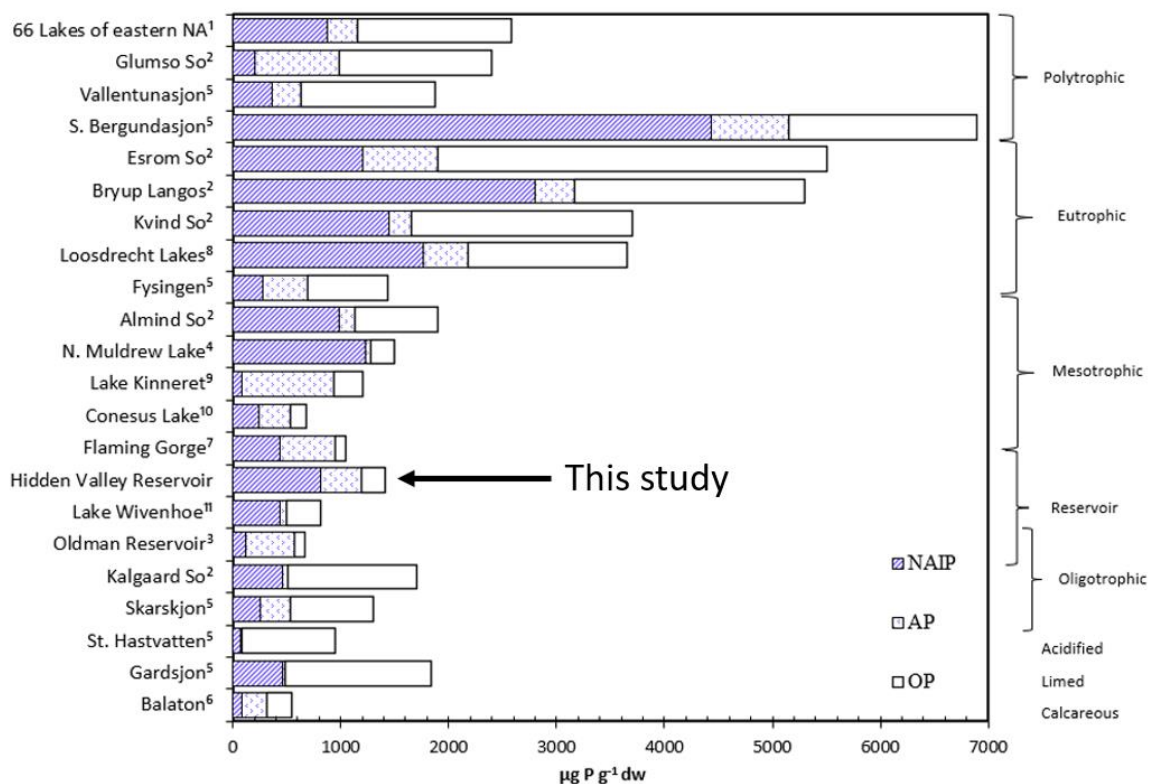
Figure 3-5: Comparison of sediment-associated particulate phosphorus (PP) forms in the HVR along with other agricultural and urbanized lakes and reservoirs across the world. The data from the studied reservoir is a composite mean value that includes the 2020 reservoir sample. 1) Ostrofsky, 1987; 2) Pettersson et al., 1988; 3) Emelko et al., 2016; 4) White & Stone, 1996; 5) Pettersson, 1986; 6) Pettersson & Istvanovics, 1988; 7) Messner et al., 1984; 8) Boers et al., 1984; 9) Eckert et al., 2003; 10) Noll et al., 2009; 11) Kerr et al., 2011.

The Cell #1 sediment redox potential in several locations was analyzed using an ORP probe. Overall, there were nine total readings with an average ORP in Cell #1 of 233 mV a $\pm$ 11.8mV. Given this confirmation of anoxia, it is likely that the redox sensitive deposited sediment in the HVR can be readily released into the water column (Molot et al., 2021). As confirmed in Fig. 3-5, there was a substantial concentration of the redox sensitive, NAIP fraction of the deposited sediment within the HVR (817 µg P g^{-1}). The anoxic sediment findings indicate that the sediment in the bottom of the HVR contains a substantial source of potentially bioavailable phosphorus which could be released into the water column.

Overall, several lines of evidence were presented to demonstrate substantial internal loading of bioavailable phosphorus in the study reservoir that stores raw (untreated) drinking water originating in an agriculturally- and municipally-impacted watershed. These include:

(1) identification of metal oxide (i.e., Al_2O_3, Fe_2O_3, and MnO) fractions that are known adsorptive surfaces that bind phosphorus to the reservoir bottom sediments,

(2) measurement of a higher level of chlorite clay in Cell #4, which in the presence of metals such as Fe, Mn, and Al, includes phosphorus-sorbing metal oxy-hydroxides on its surface,

(3) demonstration of an equilibrium phosphate concentration (EPC_0) that is higher than the dissolved phosphorus concentration typically found in the source water, thereby indicating that phosphorus will rapidly desorb from the sediment to the reservoir water column where it will be immediately available for biological uptake,

(4) quantification of a high concentration of the redox sensitive, rapidly desorbed NAIP particulate phosphorous form in the reservoir bottom sediments, consistent with a source water supply impacted by substantial agricultural runoff and more consistent with levels globally observed in mesotrophic lakes than untreated drinking water reservoirs, and

(5) demonstration of a zone of anoxia in reservoir bottom sediment indicates geochemical conditions that promote the desorption of phosphorus from bottom sediments to the reservoir water column and serve as confirmation of internal loading of dissolved phosphorus in the reservoir.

Collectively and consistently, these data indicate that the development of fine sediment management strategies to prevent the proliferation of cyanobacteria in raw (i.e., untreated) water storage study reservoir is warranted. Increases in the frequency and severity of climate change-associated disturbances such as extreme precipitation events further underscore the potential for increased delivery of phosphorus-rich fine sediment to the reservoir. This potential, coupled with higher average temperatures and longer periods of drought between precipitation events, indicates that the development of strategies to manage reservoir fine sediment-associated risks to drinking water quality and treatability would be meaningful for risk management and climate change adaptation.

3.3.2 Chemical coagulant addition for phosphorus inactivation

Bench-scale batch experiments were conducted using sediment HVR water and sediment from Cell #1 to which common drinking water treatment coagulants were added. Phosphorus adsorption/desorption to the HVR water column from sediment inactivated with a range of coagulant doses is shown in Fig. 3-6 for $FeCl_3$ (A), alum (B), and PACl (C) coagulants. The experiment conducted without coagulant addition (i.e., 0 mg L^{-1} coagulant) served as the benchmark for evaluating phosphorus inactivation by coagulant application. The EPC_0 of 271 µg L^{-1} that was calculated for the Cell #1 sediment collected in 2020 (Fig. 3-6) was substantially higher than that (47 µg L^{-1}) from the experiment conducted with sediment collected in 2013 and MilliQ^{TM} water (Fig. 3-4). This difference may be associated with shifts in the nature of the deposited sediment and/or the absence of matrix effects attributable to competitor ions when the ultrapure water was used. Evaluating the factors that contributed to these differences was not an objective of the present investigation, however.
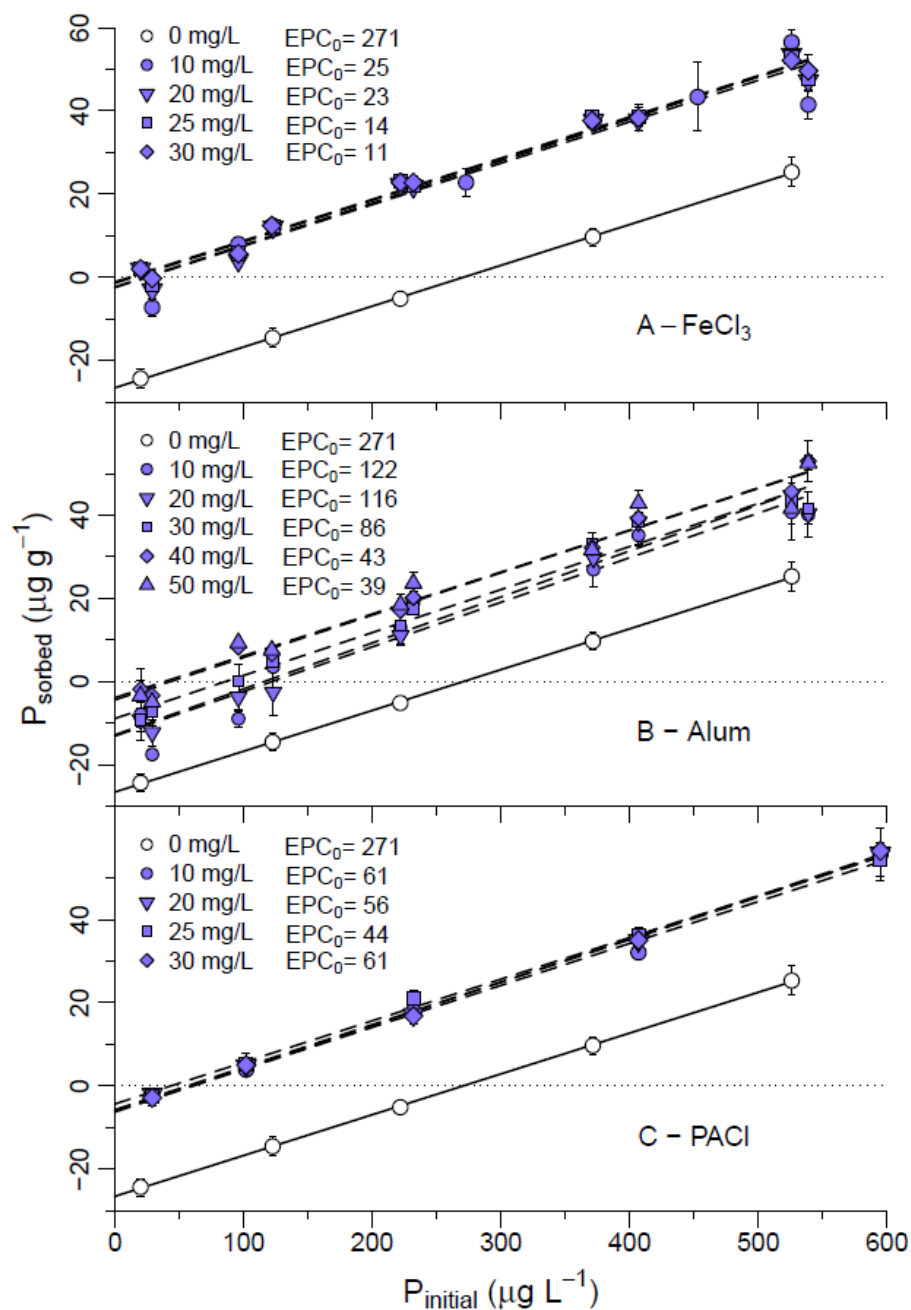
Figure 3-6: Phosphorus adsorption/desorption to the HVR water column from sediment collected from Cell#1 in September 2020, inactivated with a range of coagulant doses of **(A)** $FeCl_3$, **(B)** alum, and **(C)** PACl (mean ± 1 SD, n=3 at each aqueous phase concentration, EPC_0 values have units of µg L^{-1}). The EPC_0 for each coagulant-sediment-water matrix combination is indicated in the legend. Based on these values (i.e., lower EPC_0 values), sediments from cells with inactivated phosphorus are less likely to release more phosphorus to the water column.

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It is important to note that typical dissolved phosphorus (i.e., ortho-phosphate) concentrations in the HVR intake water have ranged from approximately 20 to 200 µg P L^{-1} with a 95% confidence interval of 20 to 200 µg P L^{-1}, over the past ten years (Fig. 3-2). Given that dissolved phosphorus concentrations in the HVR intake water are almost always less than the EPC$_0$ of 271 µg P L^{-1}, Fig. 3-6 indicates that sediment deposited in the HVR will be a net source of dissolved phosphorus to the water column (i.e., phosphorus will regularly desorb from the sediment), likely releasing 25 to 5 µg P g sediment^{-1}.

FeCl$_3$ addition resulted in the rapid reduction of the EPC$_0$ to below 30 µg P L^{-1} using all investigated coagulant doses (Fig.3-6 A). Significantly, these coagulant doses of FeCl$_3$ are typical within drinking water treatment practice and were all able to reduce the EPC$_0$ value below the target threshold of 30 µg P L^{-1}. In contrast, alum addition resulted in a steady decline of the EPC$_0$, but none of the tested doses decreased the EPC$_0$ to below the performance target of 30 µg P L^{-1}. Despite failing to reach the accepted threshold, alum addition (at a dose of 50 mg L^{-1}) achieved 86% reduction in the EPC$_0$, which would substantially reduce the amount of phosphorus released from the fine sediment to the water column. Finally, poly-aluminum chloride addition to the batch experiments resulted in mixed performance with respect to EPC$_0$ reduction when compared to FeCl$_3$ and alum. While PACl was not especially effective, a FeCl$_3$ dose of only 10 mg L^{-1} reduced the EPC$_0$ to 25 µg L^{-1} and thus met the performance target of 30 µg L^{-1}. In contrast, an alum dose of 50 mg L^{-1} only reduced the EPC$_0$ to 39 µg L^{-1}. Thus, the application of FeCl$_3$ and alum to the reservoir inflow would reduce the potential for cyanobacteria and other algae bloom formation. A concurrent reduction in treatment plant influent turbidity would be expected; this co-benefit of was not explicitly evaluated, however.

Overall, these experiments indicated that the addition of common chemical coagulants to raw (untreated) water storage reservoir inflows can inactivate phosphorus and reduce the EPC$_0$ (Fig. 3-6). All three coagulants were able to at least somewhat reduce the EPC$_0$ in the system. Despite the presence of fine sediment, coagulant addition for minimizing phosphorus release from fine sediment to the water column appears to be a promising management option. Chemical coagulation is a common step in the drinking water treatment process, so while coagulant addition to drinking water reservoirs for improving water quality is not widely practiced, the principles of coagulation still apply. These findings align with fundamental understanding of iron and aluminum solubility in water.

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The approximate sweep flocculation (i.e., optimal flocculation) dose would be expected in the mid-20s for $FeCl_3$ and poly-aluminum chloride, and in the mid-40s for aluminum sulfate at the pH of the Grand River. The superiority of $FeCl_3$ in reducing the EPC_0 was not surprising given the differences between iron and aluminum solubility in the pH range (i.e., 7 to 8) of most natural surface waters.

It is important to note that coagulant addition during the batch tests was expected to reduce the EPC_0 to a value below 271 μg L^{-1}, with a performance target of 30 μg P L^{-1}or lower, to substantially reduce the amount of phosphorus released from the fine sediment to the water column, thereby reducing the potential for proliferation of cyanobacteria and other algae. While the target of 30 μg P L^{-1}or lower is applied to dissolved phosphorus here, it is important to recall that it originates from a more general total phosphorus (TP) guideline that was developed to reflect both dissolved phosphorus and the potential for internal loading of phosphorus from sediment in lakes, as well as phosphorus forms that are not bioavailable (Chambers et al., 2012). Here, only the potential for internal loading of bioavailable phosphorus from sediment is being evaluated; thus, a lower performance target (e.g., ~5 μg L^{-1} of dissolved phosphorus) may be required in practice. This performance target is applied herein for preliminary evaluation of the various coagulant options and potential selection of approaches that would be considered for further evaluation at pilot-scale.

3.3.3 Combining reservoir dredging with chemical coagulation

To evaluate the potential efficacy of reservoir dredging combined with chemical coagulant addition, bench scale experiments were conducted to compare the initial and final aqueous phosphorus concentrations in the absence of fine sediment with a range of added coagulant types and doses. The final aqueous phase phosphorus concentration as a function of the initial concentration of dissolved phosphorus in the HVR source water that was inactivated with a range of chemical coagulant doses of (A) $FeCl_3$, (B) Alum, and (C) PACl is shown in Fig. 3-7. These data indicate that coagulant addition in the absence of fine sediment reduces the bioavailable phosphorus to near or below the performance target, depending on the inflow concentration of dissolved phosphorus while concurrently eliminating the potential for phosphorus re-release to the reservoir water column. These results are generally consistent with those reported in Fig. 3-6, when sediment was present during coagulant application.
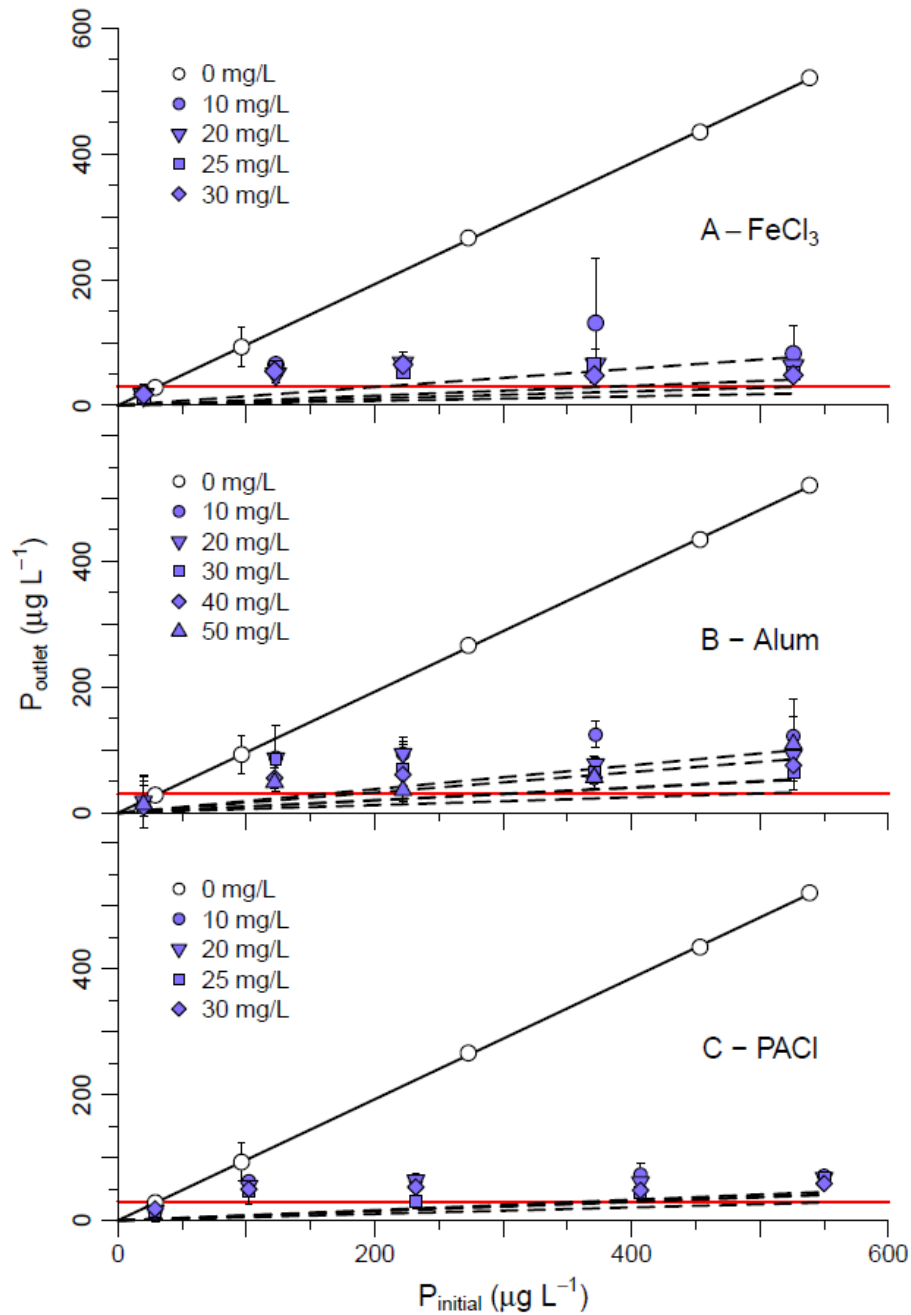
Figure 3-7: Final aqueous phase dissolved phosphorus concentration as a function of the initial concentration in the HVR source water that was inactivated with a range of chemical coagulant doses of (**A**) FeCl3, (**B**) Alum, and (**C**) PACl (mean ± 1 SD, n=3 at each aqueous phase concentration). The performance target of 30 µg L^{-1} is indicated with the solid red line. These data indicate that coagulant addition in the absence of fine sediment reduces the bioavailable phosphorus to near or below the performance target, depending on the inflow concentration of dissolved phosphorus.

Phosphorus inactivation requires coagulant addition at doses that result in metal hydroxide precipitation (Crittenden et al., 2012). Concurrent reductions in reservoir outflow/treatment plant influent turbidity would thus also be expected given coagulant application at these doses. This co-benefit of was not explicitly evaluated here, however. This evaluation underscores that further investigation is necessary to establish the optimal frequency of dredging in relation to coagulant addition. This would depend on the timing of reservoir dredging, accumulated sludge volumes, timeframe during which coagulant is applied (e.g., seasonal), the extent of internal loading of dissolved phosphorus to the water column from previously deposited sediment on which (i) phosphorus has not been inactivated and (ii) sediment that is (seasonally) coagulated for the purpose of phosphorus inactivation is deposited. Although these operational details would require optimization, the results of this study nonetheless demonstrate that seasonal coagulant application coupled with strategically-timed reservoir dredging may offer utilities reliant on offline raw water storage reservoirs an effective phosphorus inactivation approach for risk management and climate change adaptation.

Chapter 4

Conclusions

The overall goal of this research was to develop drinking water reservoir management strategies to prevent the proliferation of cyanobacteria in raw (i.e., untreated) water storage reservoirs. The key findings of this work are:

1. To reduce the potential for cyanobacteria bloom occurrence in raw (untreated) water storage reservoirs, an aqueous total phosphorus concentration of 30 µg L^{-1} is a reasonable maximum threshold concentration that should not be exceeded.

   While it has been suggested that other setting-specific thresholds may exist for certain cyanobacterial taxa, a value of 30 µg L^{-1} is widely recognized as a general threshold for eutrophication of fresh water. Thus, it represents a reasonable performance target for the development of fine sediment management strategies focused on phosphorus inactivation in drinking water reservoirs.

2. Reservoir bottom sediments contain metal oxide (i.e., Al_2O_3, Fe_2O_3, and MnO) fractions that are known adsorptive surfaces that bind phosphorus; their distribution is generally higher in downstream reservoir cells and consistent with higher levels of bioavailable dissolved phosphorus that can promote the proliferation of cyanobacteria and other algae.

   As observed herein, these fractions are generally higher in downstream reservoir cells likely because of sediment fining (i.e., smaller-sized sediment grains settle more slowly than larger sized sediments and therefore deposit further along the flowpath in reservoirs), which may not be evident based on grain size analysis due to the large fraction of fine sediment already present in reservoir. Bioavailable particulate phosphorus forms (i.e., NAIP) are widely understood to preferentially bind to these metal oxide surfaces. Thus, these geochemically analyses are consistent with higher levels of bioavailable phosphorus contributing to the higher levels of algae proliferation that have been anecdotally observed in downstream reservoir cells (i.e., Cells #3 and #4 in this study). Furthermore, there was a higher level of the clay chlorite observed in Cell #4. In the presence of metals such as Fe, Mn, and Al, chlorites have phosphorus

sorbing metal oxy-hydroxides on their surfaces. As these are more closely associated with NAIP, their presence also suggests potential for phosphorus release increases when water moves downstream through the reservoir.

3. Sediments deposited farther along the flowpath in reservoirs are management priorities because they are more likely to have higher phosphorus release potential.

   In the absence of matrix effects attributable to competitor ions (i.e., in ultrapure Type 1 MilliQ^{TM} water), the equilibrium phosphate concentration (EPC$_0$) increased from 35 to 47 µg P L^{-1} in the first two cells of the reservoir to 108 µg P L^{-1} and 407 µg P L^{-1} in Cells #3 and #4, respectively. Fine-grained sediments can release phosphorus to the water column when aqueous phase phosphorus concentrations are below the EPC$_0$. Thus, these differences in EPC$_0$ confirm that—consistent with the geochemical analyses described above—sediments deposited further along the flowpath in the study reservoir are both (i) enriched in bioavailable particulate phosphorus (i.e., NAIP) and (ii) have a higher phosphorus release potential. Accordingly, they should be management priorities.

4. Total particulate phosphorus concentrations in the reservoir bottom sediments were over 1,400 µg P/g sediment; approximately 57% of that phosphorus was the bioavailable NAIP fraction. The NAIP content of the study reservoir is amongst the highest levels that have been reported for drinking water reservoirs, and consistent with levels that have been reported in mesotrophic lake systems. These relatively elevated NAIP levels are consistent with those reported globally for watersheds impacted by significant agricultural and urban land use.

5. The addition of common chemical coagulants to raw (untreated) water storage reservoir inflows can inactivate phosphorus and reduce the EPC$_0$. The order of performance from most to least effective was FeCl$_3$ >> alum >> PACl.

   While PACl was not especially effective, a FeCl$_3$ dose of only 10 mg L^{-1} reduced the EPC$_0$ to 25 µg L^{-1} and thus met the performance target of 30 µg L^{-1}. In contrast, an alum dose of 50 mg L^{-1} only reduced the EPC$_0$ to 39 µg L^{-1}. Thus, the application of all chemical coagulants investigated to the reservoir inflow would reduce the potential for cyanobacteria and other algae bloom

formation. A concurrent reduction in treatment plant influent turbidity would be expected; this co-benefit of was not explicitly evaluated, however. The superiority of $FeCl_3$ in reducing the EPC_0 was not surprising given the differences between iron and aluminum solubility in the pH range (i.e., 7 to 8) of most natural surface waters.

6. The combination of reservoir dredging and coagulant application not only inactivates phosphorus, but also eliminates the potential for phosphorus re-release to the reservoir water column.

As discussed above, concurrent reduction in treatment plant influent turbidity would be expected; this co-benefit of was not explicitly evaluated, however. Thus, the results of this study demonstrate that seasonal coagulant application coupled with strategically-timed reservoir dredging may offer utilities reliant on offline raw water storage reservoirs an effective phosphorus inactivation approach for risk management and climate change adaptation.

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Appendix A

SRP results in ultrapure water across four reservoir cells

The following table provides a summary of the sorption data from all four reservoir cells with ultrapure water.

The EPC_0 value is where the model line crosses the zero on the y-axis and represents the concentration where sediment and water are at equilibrium. If the aqueous concentration is above the EPC_0, the sediment will adsorb phosphorus from the water; if the aqueous concentration is below the EPC_0, the sediment will release phosphorus into the water column to establish equilibrium.

Table A1: Sorption data for all four reservoir cells in ultrapure water.

Cell Number	Initial [P] ($\mu g\ L^{-1}$)	Volume of water (L)	Mass of sediment (g)	Number of samples (n)	P sorbed by sediment ($\mu g\ g^{-1}$)	
					Mean	Standard error
Cell 1	0	2.5×10^{-2}	25	9	-9.00	2.20
	25	2.5×10^{-2}	25	9	-4.00	1.30
	50	2.5×10^{-2}	25	9	1.40	1.40
	100	2.5×10^{-2}	25	9	9.40	1.40
	200	2.5×10^{-2}	25	9	27.30	1.10
Cell 2	0	2.5×10^{-2}	25	9	-6.10	1.20
	25	2.5×10^{-2}	25	9	-1.60	1.30
	50	2.5×10^{-2}	25	9	2.50	1.20
	100	2.5×10^{-2}	25	9	12.00	1.60
	200	2.5×10^{-2}	25	9	29.50	1.80
Cell 3	0	2.5×10^{-2}	25	9	-9.80	1.60
	25	2.5×10^{-2}	25	9	-7.10	1.60
	50	2.5×10^{-2}	25	9	-4.90	1.90
	100	2.5×10^{-2}	25	9	-0.70	1.40
	200	2.5×10^{-2}	25	9	8.10	1.60
Cell 4	0	2.5×10^{-2}	25	9	-38.60	1.20
	25	2.5×10^{-2}	25	9	-34.02	4.30
	50	2.5×10^{-2}	25	9	-30.73	4.20
	100	2.5×10^{-2}	25	9	-25.20	3.80
	200	2.5×10^{-2}	25	9	-20.00	4.30

Appendix B

Particulate phosphorus composition

Particulate phosphorus composition with standard deviation of the deposited sediment within the Hidden Valley Reservoir (HVR) Cell #1 collected in September 2020. Sample locations were from the left edge, middle, and right edge of the Cell to observe any variation in particulate phosphorus composition within the cell.

Table B1: Particulate phosphorus composition of the HVR sediment within Cell #1

| | Non-apatite inorganic phosphorus | | | Apatite phosphorus $(\mu g\ g^{-1})$ | Organic phopshorus $(\mu g\ g^{-1})$ | Total particulate phosphorus $(\mu g\ g^{-1})$ |
	Loosly sorbed $(\mu g\ g^{-1})$	Reductant soluble $(\mu g\ g^{-1})$	Metal oxide bound $(\mu g\ g^{-1})$			
Number of samples (n)	3	3	3	3	3	
Mean	22	459	333	376	218	1409
Standard error	5	30	24	15	11	74

Appendix C

Sediment oxidation reduction potential (ORP)

Sediment oxidation reduction potential (ORP) with standard deviation taken from the HVR Cell before sediment collection. Sample locations were from the left edge, middle, and right edge of the Cell to observe any variation in sediment ORP within the cell.

Table C1: ORP of the HVR sediment

Cell 1 Location	Sample No. (n)	ORP reading (mV)
Left	1	-230
	2	-248
	3	-212
Centre	1	-232
	2	-240
	3	-224
Right	1	-236
	2	-248
	3	-224

Appendix D

SRP results in reservoir water amended with coagulant

Appendix D provides the initial (~ 20 – 600 µg L^{-1}) and final phosphorus data after fine sediment sorption. Samples were treated with typical ranges of ferric chloride (10, 20, 25, and 30 mg L^{-1}), aluminum sulfate (10, 20, 30, 40, and 50 mg L^{-1}), and poly-aluminum chloride (10, 20, 25, and 30 mg L^{-1}). Data are presented in three tables below.

Table D1: Sorption data amended with ferric chloride

Coagulant addition	[FeCl$_3$] = 10 mg L^{-1}			[FeCl$_3$] = 20 mg L^{-1}			[FeCl$_3$] = 25 mg L^{-1}			[FeCl$_3$] = 30 mg L^{-1}		
	Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})	
	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	tandard error
	20	2.03	0.03	20	2.01	0.03	20	2.03	0.03	20	2.00	0.01
	29	-7.27	2.03	29	-3.02	0.03	29	-1.80	1.58	29	-0.27	1.42
	96	7.99	0.47	96	4.08	1.50	96	5.71	1.10	96	5.64	2.06
	123	12.38	0.14	123	12.11	0.24	123	12.35	0.23	122	12.43	0.56
	222	23.02	0.88	222	22.34	0.11	222	23.19	0.80	222	22.80	0.34
	273	22.82	3.38	232	21.81	1.10	232	21.93	1.14	232	22.79	1.56
	372	37.27	1.05	372	37.76	0.82	371	38.44	1.60	371	37.66	0.76
	453	43.37	8.27	407	37.87	1.73	407	38.56	2.31	407	38.34	3.13
	526	56.53	2.94	526	53.58	0.35	526	52.89	0.85	526	52.14	0.75
	539	41.50	3.55	539	47.28	2.47	539	47.71	1.12	539	49.67	3.71

Table D2: Sorption data amended with aluminum sulfate.

Coagulant addition	[Alum] = 10 mg L^{-1}			[Alum] = 20 mg L^{-1}			[Alum] = 30 mg L^{-1}			[Alum] = 40 mg L^{-1}			[Alum] = 50 mg L^{-1}		
	Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})	
	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error
	20	-7.89	3.03	20	-9.16	2.85	20	-9.32	4.83	20	-1.84	4.99	20	-3.44	3.83
	29	-17.52	0.86	29	-12.32	3.14	29	-7.02	3.67	29	-3.37	1.91	29	-4.94	1.88
	96	-8.93	2.09	96	-3.78	3.37	96	0.10	3.94	96	8.27	0.70	96	9.33	0.08
	123	3.51	0.25	123	-2.60	5.50	123	4.93	0.97	122	6.71	1.55	122	7.54	0.16
	222	12.99	1.58	222	10.99	1.87	222	13.48	4.91	222	17.03	0.81	222	18.46	2.39
	232	18.93	1.21	232	18.99	1.80	232	17.45	0.98	232	20.22	1.45	232	23.65	2.65
	372	26.93	4.31	372	29.71	2.76	371	33.10	2.85	371	32.21	0.90	371	31.63	0.74
	407	35.18	2.27	407	38.38	2.32	407	38.37	1.10	407	39.28	6.61	407	42.87	3.24
	526	40.70	2.85	526	44.76	2.91	526	43.56	1.57	526	45.65	0.97	526	41.64	7.66
	539	40.11	5.52	539	40.37	2.41	539	41.52	1.32	539	53.00	4.94	539	52.57	1.69
	721	58.21	5.37	721	54.80	2.09				721	70.26	0.38	721	70.89	1.93
	1132	94.84	8.57	1132	96.13	2.81									

Table D3: Sorption data amended with poly-aluminum chloride.

Coagulant addition	[PACl] = 10 mg L^{-1}			[PACl] = 20 mg L^{-1}			[PACl] = 25 mg L^{-1}			[PACl] = 30 mg L^{-1}		
	Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})		Initial [P]	P sorbed by sediment (µg g^{-1})	
	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error	(µg L^{-1})	Mean	Standard error
	29	-2.40	0.26	29	-2.07	1.19	29	-2.46	0.51	29	-2.89	2.00
	102	3.92	0.94	102	4.84	0.56	102	5.32	2.41	102	5.00	0.54
	232	17.88	2.36	232	17.07	2.13	232	21.13	1.87	232	16.78	1.92
	407	32.06	1.00	407	34.94	2.44	407	36.25	1.46	407	35.04	2.88
	595	55.61	6.23	595	55.99	0.58	595	54.45	3.95	595	56.52	1.36

Appendix E

Initial and final phosphorus concentrations reservoir water amended with coagulant

Data from the dredging efficacy experiments as a standalone treatment method and tested in conjunction with fine sediment.

Table E1: Initial ($\sim$20 – 600 μg L^{-1}) and final phosphorus concentrations in the presence and absence of fine sediment. All initial samples are HVR raw water spiked with various concentrations of phosphorus

	Sediment added			Dredged	
		Final [P] (μg L^{-1})			Final [P] (μg L^{-1})
Initial [P] (μg L^{-1})	Mean	Standard error	Initial [P] (μg L^{-1})	Mean	Standard error
20	199.18	22.91	29.13	28.47	1.52
29	129.40	39.75	96.27	92.94	30.37
96	108.86	12.03	272.90	266.33	1.53
123	237.50	17.42	453.36	434.67	1.15
222	230.97	14.12	538.63	521.00	4.00
273	150.83	17.56			
372	287.47	63.60			
453	138.33	51.98			
526	187.21	110.29			
539	146.68	7.72			

Table E2: Initial (~20 – 600 µg L^{-1}) and final phosphorus concentrations in the absence of fine

sediment. All initial samples are HVR raw water spiked with various concentrations of

phosphorus. Samples were treated with typical various ranges of ferric chloride (10, 20,

25, and 30 mg L^{-1}), aluminum sulfate (10, 20, 30, 40, and 50 mg L^{-1}), and poly-aluminum

chloride (10, 20, 25, and 30 mg L^{-1})

Coagulant

[FeCl₃] = 10 mg L⁻¹			[FeCl₃] = 20 mg L⁻¹			[FeCl₃] = 25 mg L⁻¹			[FeCl₃] = 30 mg L⁻¹		
Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error
20	18.65	14.62	20	16.72	1.89	20	14.31	7.86	20	17.29	16.43
123	65.06	7.74	123	49.70	13.77	123	59.19	12.83	122	54.30	9.36
222	65.02	8.72	222	66.97	17.30	222	54.47	4.76	222	64.91	6.63
372	131.03	101.69	372	65.22	23.73	371	63.59	9.68	371	47.38	10.94
526	82.48	44.08	526	62.55	15.96	526	51.36	12.36	526	48.10	5.32

Coagulant

[Alum] = 10 mg L⁻¹			[Alum] = 20 mg L⁻¹			[Alum] = 30 mg L⁻¹			[Alum] = 40 mg L⁻¹			[Alum] = 50 mg L⁻¹		
Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error
20	15.47	9.25	20	18.43	24.73	20	9.56	50.26	20	8.83	49.69	20	13.63	37.56
123	87.76	2.33	123	87.21	51.89	123	84.93	8.43	122	55.60	15.68	122	48.46	2.00
222	94.06	15.25	222	94.20	20.47	222	70.24	51.08	222	60.92	5.76	222	36.10	22.37
372	124.43	20.83	372	78.06	10.82	371	64.75	25.67	371	56.36	9.82	371	56.43	5.49
526	122.18	31.45	526	92.09	25.30	526	64.38	13.23	526	75.77	12.95	526	109.45	71.94

Coagulant

[PACl] = 10 mg L⁻¹			[PACl] = 20 mg L⁻¹			[PACl] = 25 mg L⁻¹			[PACl] = 30 mg L⁻¹		
Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error	Initial [P] (μg L⁻¹)	Final [P] (μg L⁻¹) Mean	Standard error
29	14.19	2.11	29	15.91	12.91	29	14.28	6.42	29	18.40	20.54
102	62.41	8.48	102	55.14	5.76	102	47.33	22.34	102	49.70	4.71
232	64.36	10.12	232	63.91	5.90	232	31.26	12.03	232	53.13	18.27
407	73.00	18.52	407	61.00	3.61	407	44.67	12.06	407	48.00	13.00
550	71.00	9.17	550	68.33	8.08	550	68.67	3.21	550	58.67	7.09

Appendix F
Quality control

A set of phosphorus standards (0, 25, 50, 100, 200, 400, 800 µg P L^{-1}) were used to create a standard calibration curve by which all samples were benchmarked against. Samples were analyzed when the coefficient of determination (R^2) for the calibration curve was greater than 0.995. To ensure there was no analytical drift during the SRP analysis, random known standards were used every 15th interval of samples. If there was a greater than 5% deviation from the known concentration, that section of samples would be re-analyzed.

Table F1: Interval samples of known phosphorus concentration and percent (%) difference from measured value

Sample No.	Phosphorus concentration measured ($\mu g\ L^{-1}$)		
	Expected concentration	Measured concentration	Percent difference
15	50	48	4%
30	100	97	3%
45	200	198	1%
60	400	400	0%
75	50	48	5%
90	100	95	5%
105	200	196	2%
120	400	392	2%
135	50	50	0%
150	100	98	2%
165	200	190	5%
180	400	385	4%
195	25	25	2%
210	25	25	0%
225	25	24	3%
240	50	48	4%
255	50	50	0%
270	50	50	1%
285	100	95	5%
300	100	97	3%
315	100	98	2%
330	200	198	1%

Sample No.	Phosphorus concentration measured (μg L^{-1})		
	Expected concentration	Measured concentration	Percent difference
345	200	192	4%
360	200	200	0%
375	400	400	0%
390	400	385	4%
405	400	388	3%
420	400	385	4%
435	400	385	4%
450	400	392	2%
465	200	198	1%
480	200	196	2%
495	200	198	1%
510	100	96	4%
525	100	98	2%
540	100	97	3%
555	50	48	4%
570	50	48	4%
585	50	49	2%
600	25	25	2%
615	25	24	4%
630	25	24	4%
645	25	24	4%
660	25	25	0%
675	25	25	1%

| Sample No.	Phosphorus concentration measured (µg L^{-1})		
	Expected concentration	Measured concentration	Percent difference
690	50	48	4%
705	50	50	0%
720	50	49	3%
735	100	100	0%
750	100	99	1%
765	100	96	4%
780	200	190	5%
795	200	194	3%
810	200	192	4%
825	400	396	1%
840	400	396	1%
855	400	385	4%
870	400	388	3%
885	400	381	5%
900	400	392	2%
915	200	196	2%
930	200	190	5%
945	200	194	3%
960	100	98	2%
975	100	99	1%
990	100	99	1%
1005	100	95	5%

Appendix G
Experiment photos

**Figure G1:** HVR cell#1 on by-pass for reservoir dredging

Figure G2: Sediment accumulated on the reservoir floor, along with an example of ORP probe in

use.

A

B

C

D
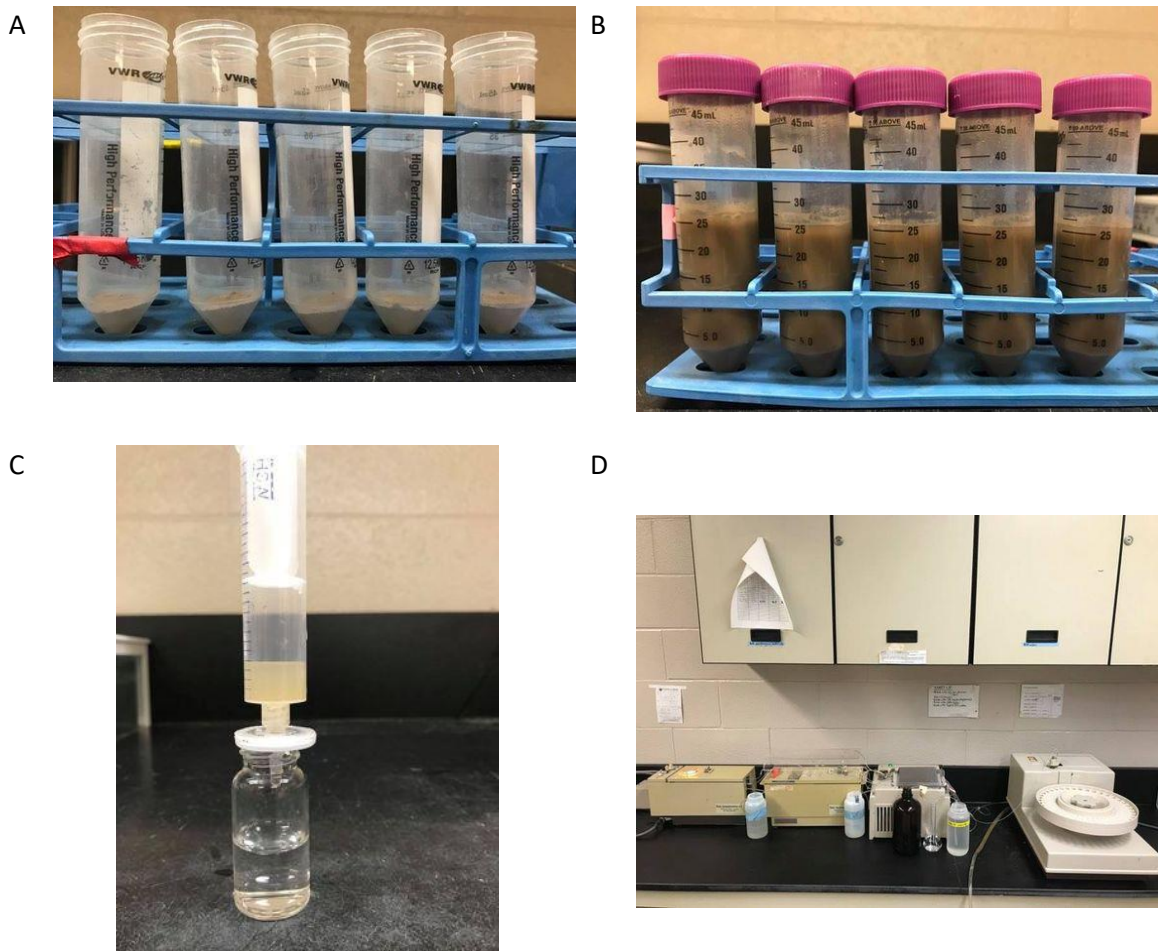
Figure G3: Sorption experiment process: A) add sediment to centrifuge tube, B) add water to
centrifuge tube spiked with various phosphate concentrations and agitate for 18 hours
on shaker table, C) filter sample through 0.45 µm filter, and D) analyze final
phosphorus concentration using ammonium molybdate/stannous chloride method
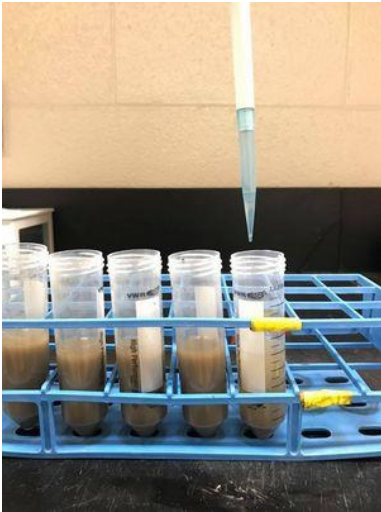
III IV

Figure G4: Sorption experiment process with coagulant amendment. Steps A) and B) the same as
figure G3-A and G3-B, III) after 18 hours of agitation, set centrifuge tubes upright and
add range of coagulant dose and type, rapid swirl for 10 seconds, and IV) filter sample
through 0.45 μm filter, before analyzing final phosphorus concentration using
ammonium molybdate/stannous chloride method
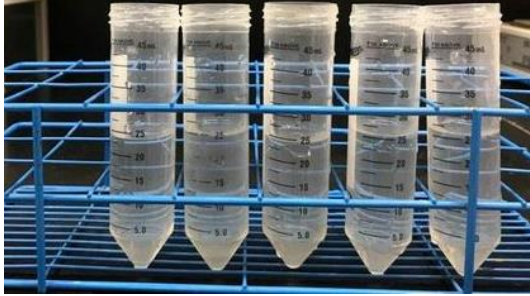
Figure G5: Dredging efficacy experiment process. A) add water to centrifuge tube spiked with various phosphate concentrations and agitate for 18 hours on shaker table, B) if coagulant is to be added, after 18 hours of agitation set centrifuge tubes upright and add range of coagulant dose and type, rapid swirl for 10 seconds, and C) filter sample through 0.45 μm filter before analyzing final phosphorus concentration using ammonium molybdate/stannous chloride method
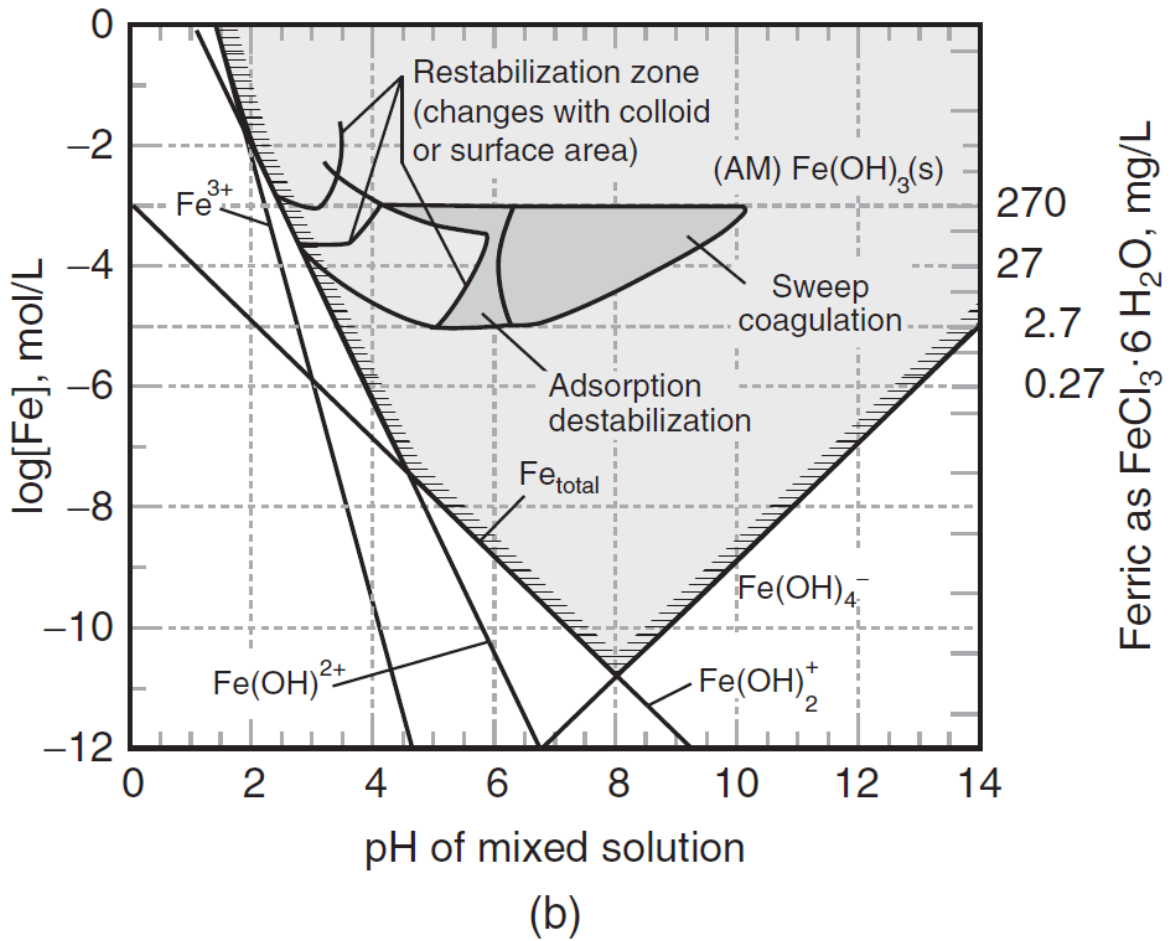
Appendix H

Solubility diagrams
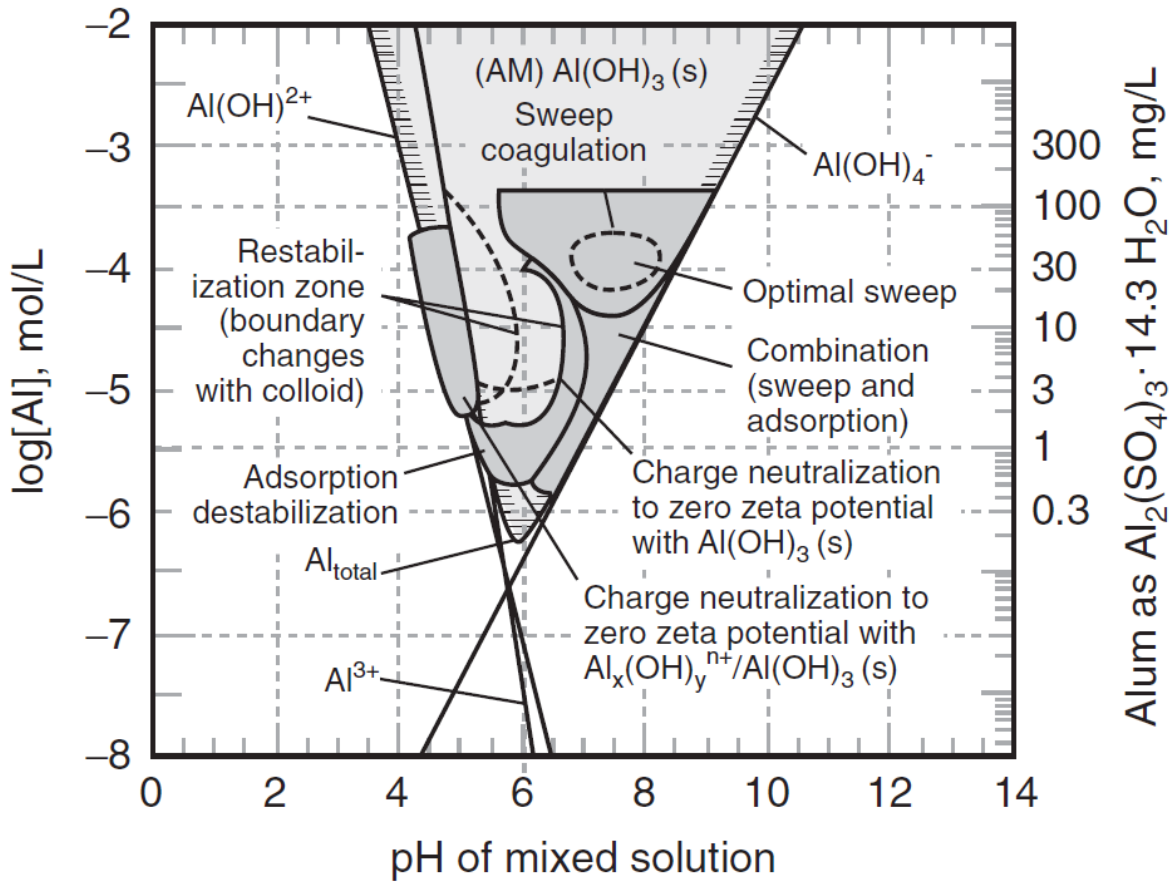
(a)

Figure H1: Solubility diagram for (a) alum and (b) ferric chloride at 25ºC (Crittenden et al; 2012).

Appendix I

Reservoir sediment calculations

	Dimensions		
	Area (m^2)	Depth (m)	Volume (m^3)
Cell 1	2866	12.54	35940
Deposited sediment (4")	2866	0.10	291
Water	2866	12.44	35648
Sediment dry-weight density	818	$(kg\ m^{-3})$	
	$(kg_{sed}\ m^{-3}_{water})$	$(g_{sed}\ L^{-1}_{water})$	
Sediment:water ratio (HVR)	6.68	6.68	
Sediment:water ratio (lab)	10	10	

Figure I1: Calculations detailing the approximate sediment and water volumes inside the drinking water reservoir cell, as well as the ratio of sediment to water in the HVR versus the experimental ratio.