

An Overview of Common Sediment Contaminants and Remediation Methods in North America and Europe

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Executive Summary

This project consists of three major deliverables:

- 1) Literature review of common sediment contaminants, regulations, and remediation techniques
- 2) Methodology and results from a preference/avoidance study with California blackworms
- 3) Short, plain language summary of the research trial

Literature Review

The management of contaminated sediments is an ongoing challenge for many regions across the globe. Common sediment contaminants include polycyclic aromatic hydrocarbons (PAHs), pesticides, polychlorinated biphenyls (PCBs), and various heavy metals such as lead, mercury, arsenic, and cadmium. Regulations surrounding sediment management are often very general and can lack the technical details necessary to make informed management decisions easily in some areas. Thus, specific regulations about suspended sediments, turbidity, and total maximum daily load are highlighted in this review. Common remediation strategies, including monitored natural recovery, passive and active capping, dredging, and a combination of techniques, are discussed in the final section of the literature review. These methods are presented with a focus on environmental impact, economic considerations, and practicality in order to provide a meaningful overview of the conditions in which the implementation of these techniques would be the most favorable. For the Saskatchewan River Delta, a combination of remediation techniques depending on site-specific factors will likely be the best approach.

Research Trial

The second component of this project consists of a preference/avoidance study with *Lumbriculus variegatus*, more commonly known as the California blackworm. Eleven potentially-contaminated sediment samples from various locations in Saskatchewan were tested to determine the behavior of the worms over 144 hours. Each sample, as well as a control, was tested in four replicates with 10 worms in each replicate, for a total of 480 worms in the trial. The results showed a very strong preference for the sediments over quartz sand that was used as a reference, with three of the samples having preferences between 80-90%, two samples between 90.1-99.9%, and six samples having 100% preference for the sediment.

Plain Language Summary

The final component of this project is a brief and straightforward summary of the research trial. This summary can be used for community presentations in the future and is useful for audiences of all backgrounds.

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1.0 Introduction

The Saskatchewan River Delta is one of the most extensive and biodiverse inland deltas in Canada. This area is situated on the traditional land of the Cumberland House Cree Nation, Peter Ballantyne Cree Nation, and Cumberland House Métis. There have been vast changes to the Saskatchewan River Delta's flow patterns in the past century, which have had a profound impact on the people who live there as well as the surrounding ecosystem. One of the most prevalent results has been an overall reduction in the amount of water that reaches the delta in the summer. These changes have negatively impacted fish production in the area, which many of the inhabitants rely on as a valuable resource. In addition to the flow changes, sediment has become trapped in the reservoirs upstream, which yields a net sediment depletion in the delta over time. This depletion causes the banks and channel beds to erode, which has negative impacts on the surrounding areas as well. Sediment restoration is needed, but possible sediment sources may be contaminated, and so sediment management techniques must incorporate remediation methods that are feasible for the area. This project is a part of a larger project that is striving to determine whether sediment restoration is a viable option for the Saskatchewan River Delta.

Partner Organization

The partner organization for this project is the Cumberland House Fishermen's Co-operative, which has been in effect since 1949. This organization consists of approximately 30 members who organize and oversee all aspects related to fishing in the area.

Project Objectives

- Highlight common contaminants in freshwater sediments, including an overview of each contaminant and their probable effect levels (PELs)
- Outline regulations surrounding sediment management, including suspended sediment, turbidity, and total maximum daily load (TMDL) information
- Provide brief summaries of common remediation techniques, including some of the benefits and drawbacks of each method and economic implications
- Determine the preference/avoidance behavior of California blackworm for various sediments collected from 11 sites in Saskatchewan
- Develop a plain-language summary of the research findings to be used in the future as a community tool and summary of the trial

2.0 Literature Review

2.1 Sediment Contaminants

Sediment contamination has been an issue that several countries have faced for many years. Contaminated sediments have become a widespread problem because of increased urbanization and industrialization, which has yielded vast amounts of organic pollutants and heavy metals being released into the sediments (Knox and Paller, 2013). These pollutants commonly enter the environment *via* point sources or through surface runoff events that feed into water bodies and groundwater sources (Knox and Paller, 2013). Despite the improvement in surface water quality over the past few decades, contaminated sediments are still a source of concern and a potential lasting legacy of the past when regulations were far less stringent. The organic contaminants commonly include polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), while the inorganic contaminants are typically metals (Cecchi et al., 2009). Some of the most significant sources of heavy metals stem from agricultural production, urban land development, industrial activities, as well as chemical spills (Knox and Paller, 2013) and can persist in the environment for many years. These metals accumulate in the sediments and can become remobilized under certain conditions where they are disturbed, which presents a continuous threat to the ecosystem. Some of the activities that may remobilize the contaminants are certain remediation techniques such as dredging or capping, and natural events such as flooding or erosion outcomes. On the other hand, polycyclic aromatic compounds often stem from fossil fuels or can be present in the atmosphere due to incomplete combustion (Heim et al., 2004). Some of the organic contaminants can be degraded *via* various transformation and loss pathways, but some constituents may still be harmful to the environment (Cruz et al., 2015) and must not be overlooked.

The chemical and physical properties of the pollutants determine their impact on the surrounding environment. More specifically, hydrophilic elements usually amass in the aqueous phase, which makes them highly mobile and subject to rapid movement, while hydrophobic pollutants predominate the solid phase and are not as easily transported in their dissolved form (Heim et al., 2004). Chemicals that are sediment-bound become a risk to sediment-dwelling organisms when the level of exposure is high enough to induce negative biological effects. However, often only a fraction of these chemicals is bioavailable, which yields a lower level of exposure for these organisms than expected based on total sediment concentrations (den Besten et al., 2003). Thus, the specific properties of the surrounding environment play a significant role in what contaminants are present and in what amounts. Further, due to the large amount of variation in the availability of the contaminant, the results of a sediment toxicity test are somewhat limited in what they can reveal. Since sediment toxicity tests are what form the basis of a risk assessment for the area, it is difficult to convey the harmfulness of the contaminant with the toxicity test value alone. Thus, the probable effect level (PEL) is used to indicate the level of a contaminant that frequently induces adverse biological effects and is listed for each contaminant below.

2.1.1 Polycyclic Aromatic Hydrocarbons (PAHs)

One of the major categories of sediment contaminants is hydrocarbons, with polycyclic aromatic hydrocarbons (PAHs) being the most prevalent (Cutroneo et al., 2015). These contaminants are common components of fossil fuels and are released in large volumes due to their widespread occurrence (Heim et al., 2004). There are two main categories of some of the potential sources of PAHs – pyrogenic and petrogenic. Pyrogenic sources are generally complex blends of hydrocarbons that stem from organic matter that was subjected to extreme temperatures, but lacked the amount of oxygen to yield complete combustion (EPRI, 2008). Some examples of pyrogenic sources include internal combustion engines, fires, and gas production from coal and oil products (EPRI, 2008). Petrogenic sources typically originate from refined crude oil products including coal, asphalt, and gasoline (EPRI, 2008). Cutroneo et al. (2015) note that PAHs are often of anthropogenic origin and are considered to be a priority pollutant by the United States Environmental Protection Agency (US-EPA). They are also listed as priority pollutants under the Environmental Quality Standards (EQSs) of the European Water Framework Directive. PAHs are classified as priority pollutants because they present considerable toxicity risks to some aquatic organisms due to their carcinogenic, teratogenic, and mutagenic potential (Nasher et al., 2013; Zhou et al., 2000). The US-EPA has specifically listed 16 PAHs as priority pollutants (Table 1). They have low solubility in water and can accumulate within sediments over time, with the potential of becoming resuspended if the area is disturbed (Cutroneo et al., 2015). The PEL in freshwater sediments of each PAH listed as a priority pollutant is listed in Table 1.

Table 1. The 16 PAHs listed as priority pollutants by the US-EPA and the freshwater sediment probable effect levels (PEL). Modified from Perelo (2009) and the Canadian Council of Ministers of the Environment (CCME) (1999j). A “-” symbol indicates that a value was not available.

	PEL (ppb) dw		PEL (ppb) dw		PEL (ppb) dw
Two-ring		Four-ring		Five-ring	
Naphthalene	391	Chrysene	862	Benzo[a]pyrene	782
Fluorene	144	Pyrene	875		
Acenaphthene	88.9	Benzo[a]anthracene	385	Dibenxzo[a,h]anthracene	135
Acenaphthylene	128	Benzo[b]fluoranthene	-	Indeno[1,2,3-c,d]pyrene	-
Three-ring		Benzo[k]fluoranthene	-	Six-ring	
Fluoranthene	2355			Benzo[g,h,i]perylene	-
Phenanthrene	515				
Anthracene	245				

2.1.2 Pesticides

In the past, the most commonly used pesticides in North America were organochlorines due to their prominent usage in agricultural activities. Some of the major organochlorines, according to Jayaraj et al. (2016), include dichloro-diphenyl-trichloroethane (DDT), endrin, methoxychlor, chlordane, endosulfan, heptachlor, aldrin, lindane, mirex, and toxaphene. The ability to persist in the environment for long periods of time and hydrophobicity are two properties that influence the accumulation potential of pesticides in the sediments. More specifically, the United States Geological Survey (1999) states that pesticides that have moderate-to-high persistence in the environment and moderate-to-low solubility in water are more likely to collect in the sediments. These pesticides are of particular concern for aquatic ecosystems as they often accumulate in the sediments as a result of contaminated soil and water, industrial production, and deposits from the atmosphere (Jayaraj et al., 2016). The United States Geological Survey (1999) summarized a series of previous studies and found that dieldrin, DDT, and chlordane were frequently found in sediments, despite the fact that their use was suspended in the 1970s which illustrates their long-term effects on the environment. The PELs for some of the common organochlorine pesticides are listed in Table 2.

Table 2. Common organochlorine pesticides and their respective PELs. A “-” symbol indicates that the value was not available.

Pesticide Name	PEL (ppb)	Source
Aldrin	-	
Chlordane	8.87	CCME (1999b)
DDT	4.77	CCME (1999c)
Dieldrin	6.67	CCME (1999d)
Endosulfan	-	
Endrin	62.4	CCME (1999e)
Heptachlor	2.74	CCME (1999f)
Hexachlorocyclohexane (Lindane)	1.38	CCME (1999g)
Methoxychlor	-	
Metolachlor	-	
Toxaphene	-	

2.1.3 Polychlorinated Biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are categorized based on the number of chlorine atoms within their structure. According to the CCME (2001), the number of chlorine atoms alters their physical and chemical properties, ranging from one (monochlorobiphenyls) to ten chlorine atoms (decachlorobiphenyls). Their presence in sediments can consist of one or more combinations of various PCBs, commonly known as Aroclors. These Aroclors can be difficult to distinguish within a contaminated sediment sample since they consist of a mixture of multiple PCBs. In particular, Aroclors 1254 and 1260 are difficult to remove because of their high chlorination. They have low bioavailability and require a non-aerobic remediation method (Fennell et al., 2011). PCBs are some of the most dangerous pollutants due to their

carcinogenicity, toxicity, and the gradual breakdown and release into the surrounding environment (Perelo, 2009). Despite the heavy limitations on their use in North America, they continue to be a threat from contaminated soil runoff, landfill leachates, spills, leaks, commercial and municipal effluents, and deposition from the atmosphere (Strachan, 1988; WHO, 1992). The probable effect concentration of total PCBs in freshwater sediments is 277 ppb dry weight (CCME, 2001).

2.1.4 Heavy Metals

Unlike organic contaminants, heavy metals are not able to be degraded into their chemical constituents, so their presence can be used as a stable indicator of contamination in the area (Heim et al., 2004). Some of the commonly found heavy metals present in freshwater sediments are mercury, lead, cadmium, and arsenic.

2.1.4.1 Mercury

According to the US-EPA (2016), mercury is a common contaminant found in water bodies as a result of volcanic, urban, and industrial activities. It is also possible for mercury that is present in the air to accumulate in various water bodies and become integrated into the sediments (US-EPA, 2016). Under favourable conditions, mercury in the sediments can then be converted by various microorganisms to methylmercury, which is a very toxic form that has the potential to bioaccumulate in the aquatic organisms which inhabit the area. Geographical characteristics also influence mercury and methylmercury levels. In the National Lakes Assessment (2012), researchers found that 51% of the total mercury in the sediments was found in moderately disturbed areas of the lakes. In the same study, the researchers also noted that the highest number of sediment methylmercury contamination sites (40%) were in the most disturbed sections of the lakes (National Lakes Assessment, 2012).

If the levels of mercury or methylmercury are high enough, there can be serious effects on ecosystem and human health. According to the CCME (1999i) and the National Oceanic and Atmospheric Administration in the United States (1999), the PEL for freshwater sediments is 486 ppb dry weight. Contamination levels vary considerably based on location, but sediment contamination levels in freshwater lakes and rivers have been recorded up to 15 ppm and 25 ppm, respectively (Environment Canada, 1997a). In the Great Lakes, the sediments have had up to 15 ppm of mercury present (Environment Canada, 1997a).

2.1.4.2 Lead

Historically, lead is a prominent contaminant that enters the aquatic environment mainly through runoff events or is deposited from the atmosphere. Organic sources of lead typically stem from anthropogenic activities and generally have higher toxicity than inorganic compounds. Lead is considered a toxic substance under the Canadian Environmental Protection Act (CEPA), which

means that it is entering the environment at levels that can cause adverse effects (CCME, 1999h). It is not commonly found in its elemental state and is most likely to be found in a divalent state in nature (CCME, 1999h). In aquatic settings, lead is typically found in areas where the sediments are susceptible to reduction or possess high cation exchange capacity, which can make it more bioavailable (Environment Canada, 1998). Similar to other heavy metals, environmental conditions such as sediment disturbance and location can also influence its bioavailability. The PEL for lead in freshwater sediments, according to the CCME (1999h), is 91.3 ppm dry weight.

One of the most significant threats to human and aquatic animal health is bioaccumulation of lead in aquatic organisms. In a study conducted by Gale et al. (2002), the researchers found a strong correlation between lead concentrations in the sediment and the levels found in bottom-feeding fish. The authors note that these results are in agreement with the natural feeding behaviours of these fish, as the data support much higher lead concentrations in suckers than in sunfish. In contrast, this study did not find a significant correlation between the lead concentration in sediments and concentrations in bass, which the authors attributed to the difference in feeding habits and position in the food chain.

2.1.4.3 Cadmium

Due to its toxicity even at low levels, cadmium is of concern for aquatic ecosystems. It is formed as a by-product of the refinement of zinc and lead because of its natural presence in the ores (Lenntech, 2020). In addition, the high demand for nickel/cadmium batteries worldwide has made cadmium a very prevalent contaminant across the globe. Cadmium is also used for coating materials that are subject to corrosion and can be present in some detergents, phosphate-based fertilizers, and refined oil and gas products (Lenntech, 2020). Thus, cadmium commonly enters the aquatic environment through atmospheric deposition, runoff events, and water contamination. As with other contaminants, there are site-specific factors that play a role in the bioavailability of the substance. For cadmium, this includes factors such as redox potential, rate of uptake by the ecosystem, and levels of organic matter (Environment Canada, 1997). According to the CCME (1999b), the PEL for cadmium is 3.5 ppm in freshwater sediments.

2.1.4.4 Arsenic

Similar to all other metals, arsenic cannot be broken down once it enters the aquatic environment, which makes it of considerable concern. The major source of arsenic emissions stem from the copper production industry, but zinc and lead manufacturing and some agricultural production methods contribute as well (Lenntech, 2020). Its stability is influenced by its oxidation state, with As^{+5} (arsenate), As^{+3} (arsenite), As , and As^{-3} being the most stable (CCME, 1999). However, the CCME (1999) notes that arsenic can be caught within the

crystalline structure of various minerals, including clay, and is the least bioavailable in this form. The most common form of inorganic arsenic in aerobic freshwater systems is arsenate (CCME, 1999). Recent literature (Jia et al., 2013; Xu et al., 2016) has indicated that certain microbial processes are able to convert highly toxic inorganic arsenic to an organic form under certain conditions, which can increase its bioavailability. In terms of toxicity, from most to least toxic are: arsenite, arsenate, monomethylarsonic acid, dimethylarsinic acid, and arsenobetaine (Hindmarsh and McCurdy 1986; Shiomi 1994). The site-specific factors that influence the bioavailability of arsenic are the same as for the other heavy metals – including various physicochemical, geochemical, and biological properties. The PEL in freshwater sediments is 17 ppm (CCME, 1999).

3.0 Regulations

Even though sediment is a crucial component of many aquatic habitats, it can pose a risk to the ecosystem if it is introduced in excessive quantities. When large quantities are present, the development of algal blooms can occur, as well as excessive deposition in some areas, and poor water quality (Fondriest Environmental, 2014). An abundance of suspended sediment can also negatively impact aquatic life by causing physical harm and disturbing natural migration events (Fondriest Environmental, 2014). When sediment is disturbed or transported too frequently, the quality of the water can suffer as a result of increased turbidity. More specifically, increased turbidity has the potential to increase the water temperature because of more solar heat being absorbed by the sediment than the water (Wetzel, 2001). Higher water temperatures can stress the aquatic organisms in the area since it yields lower dissolved oxygen levels, which can be critical for the ecosystem. The resuspension of anoxic sediments can also result in extreme oxygen consumption events which can have a detrimental impact on the fish in the area. It is also possible for aquatic habitats to be smothered by the increase in sediment deposition. In areas where the increased sediments are stemming from agricultural and urban activities, there is the potential for algal blooms to thrive due to more available nutrients (Fondriest Environmental, 2014). Another issue that can arise from high levels of sedimentation is changes to the waterway and directionality or flow of the water (Zaimes and Emanuel, 2006). If extreme sediment accumulation occurs, it may exceed the total maximum daily load (TMDL). The TMDL was established to highlight the negative impact that the contaminant, in this case sediment, can have on the area (Mahdi, 2001). Taken together, these factors highlight the need for regulations to be established in order to protect the aquatic environment.

3.1 Canadian Guidelines

Under section 36 of the Environment and Climate Change Canada guidelines, the addition of deleterious substances into the aquatic environment is prohibited unless an exemption is made under the Fisheries Act or any other federal legislation. Fisheries and Oceans Canada ensures that fish that inhabit Canadian waters, as well as their habitats, are protected. Under the Fisheries Act, it is a serious offense to release any deleterious substances into aquatic habitats. A deleterious substance is any material that has the potential to alter the water quality of the area such that the water may inflict harm to the fish and other species that inhabit the area

(Government of Canada, 1985). It also includes any water that contains any contaminant in a concentration that could cause adverse effects to the aquatic habitat if it was added to the water in that area (Government of Canada, 1985). Any activity that may cause harm to the aquatic environment, including habitat disruption or destruction, is also prohibited under the Fisheries Act. Sediment has the potential to cause harm to the aquatic environment and must be carefully screened for toxic materials as well as follow pre-determined guidelines for its addition. The CCME has created sediment quality guidelines that are designed to protect the aquatic environment and outline management practices to support aquatic ecosystem health (Bagherifam et al., 2019).

3.1.1 Suspended Sediments

In Canada, the CCME has published specific guidelines for suspended sediments due to their potential to cause harmful effects to fish if the concentrations are high enough in a specific timeframe. The composition of the suspended sediment affects the turbidity of the water and may consist of materials such as clay, silt, and organic and inorganic particles. According to the CCME (1999k), the freshwater concentration of suspended sediments in clear flow scenarios must not have more than a “maximum increase of 25 mg/L from background levels for any short-term exposure (e.g., a 24-hour period).” For longer-term exposures lasting up to 30 days, there must not be more than a “maximum increase of 5 mg/L from background levels.” (CCME, 1999k). For high flow scenarios, the “maximum increase [is] 25 mg/L from background levels” granted that they are between 25 and 250 mg/L (CCME, 1999k). If the background levels are higher than 250 mg/L, the CCME (1999k) states that there should not be more than a 10% increase. There is no available data for the PEL of suspended sediments from the CCME (1999k).

3.1.2 Turbidity

The turbidity of a water sample refers to how transparent or clear the sample appears. A higher turbidity value indicates a higher level of suspended or dissolved matter within the sample. Some examples of materials that can increase turbidity are very small inorganic and organic materials, plankton and other microorganisms as well as microalgae (USGS, 1965). This parameter is important because it reflects the amount of material within the sample that may affect the surrounding environment. The CCME (2002) has developed guidelines for freshwater environments to ensure that certain levels are not exceeded that may inflict harm on the ecosystem. For clear flow scenarios, there must not be an increase greater than 8 nephelometric turbidity units (NTUs) from the background level for a 24-hour period (CCME, 2002). In longer-term situations, the highest average increase must not exceed 2 NTUs from the background level (CCME, 2002). The CCME (2002) also states that if the background levels are between 8 and 80 NTUs that there is no more than an 8 NTU increase from background levels, and if the background level exceeds 80 NTUs, there should not be more than a 10% increase. There is no data available to determine the PEL for turbidity from the CCME (2002).

3.2 Total Maximum Daily Load

The total maximum daily load (TMDL) reflects the maximum amount of sediment that can be present without compromising the water quality of the area, (Mahdi, 2001) and falls under section 303 of the Clean Water Act (US-EPA, 2018). The overall goal of the TMDL is to establish the loading capacity of an area in order to effectively distribute the contaminant load and implement the most appropriate control measures (US-EPA, 2018). Thus, a TMDL can be established for various aspects of the sediment situation, such as suspended solids, contaminants, nutrient levels, and fine sediment deposition (Fondriest Environmental, 2014). According to the US-EPA (2018), the development of a TMDL is important because it acts as a link between the water quality guidelines and the implementation of proper control methods. During the development of the TMDL's, it is the responsibility of the state to present the proposed TMDL to the EPA for approval, even if third party stakeholders were involved (US-EPA, 2018). The Michigan Department of Environment, Great Lakes, and Energy (2020) list the EPA-approved total maximum daily loads for various water bodies in the United States for different parameters including dissolved oxygen, *E.coli*, mercury, nitrate, PCBs, phosphorus, and sediments.

3.3 Sediment Contamination

In addition to the risks presented with quantity, sediment contamination is another factor that should be considered. There is a large degree of variation between countries with respect to the development and implementation of regulatory guidelines for sediment contamination. The most common measure of contaminant levels stems from the total contaminant concentrations, which are typically used to develop screening frameworks and specific regulatory guidelines (Bagherifam et al., 2019). However, there has been criticism in the past for developing guidelines based on the total concentrations because there are often site-specific factors that influence the bioavailability of the contaminant (Bagherifam et al., 2019). Thus, the bioavailability information has been deemed more appropriate as a measure of the potential impact that the contaminant may impose. Sediment-biota accumulation factors (BSAFs) are another indicator that is used for depicting the bioaccumulation of contaminants in aquatic organisms and in sediments (Burkhard, 2009). Unlike total contaminant concentrations, BSAFs are designed to take bioavailability into consideration. In the United States, the EPA has allocated many resources towards increasing the awareness and knowledge surrounding sediment contamination as well as the best management and remediation options (Majone et al., 2015). In Europe, an organization called SedNet has been instrumental in developing management strategies for contaminated sediments (Majone et al., 2015). Furthermore, the European Water Framework Directive also establishes water quality guidelines which contaminated sediments may adversely effect in some areas. Environmental quality standards (EQSs) have also been developed for 33 priority substances and 8 other contaminants under the European Water Framework Directive which defines allowed limits of contaminant concentrations in surface waters (European Commission, 2008).

4.0 Remediation Methods

Due to the vast amount of sediment contamination in various sites worldwide, many different remediation techniques attempt to minimize contaminant levels. The more traditional options typically consist of no action, monitored natural recovery (MNR), and various *in situ* and *ex situ* management techniques (Knox and Paller, 2013). Majone et al. (2015) report that oftentimes, the massive volume of sediments at certain sites can exceed one million cubic meters, which is incredibly difficult to tackle in conjunction with the enormous volumes of water that the sediments occupy. These authors also estimate that around 15,000 sites that have been contaminated across Europe will need some form of management or remediation in the next decade, which could cost up to 30 billion dollars (Majone et al. 2015). In Europe, the Water Framework Directive (WFD) established the Directive on Environmental Quality Standards (EQSD), which allows for each member state to create environmental quality standards for sediments. However, there is no single complete source of sediment remediation pursuits in Europe (Majone et al., 2015). The Great Lakes Water Quality Agreement (1978) and the Canadian Environmental Protection Act (1999) discuss some remediation methods and the factors that must be taken into consideration when selecting an appropriate method in Canada. There has also been a lack of recognition of the need to properly manage areas with large volumes of contaminated sediments in the past, especially in certain areas of the United States such as the Hudson River (Majone et al., 2015). Factors such as sediment types, depth, and various site-specific factors all play an important role in determining which technology is best suited for a particular area (Reis et al., 2007).

Another factor that is highly variable and difficult to present accurately is the cost of the different remediation technologies. While it is possible to locate some specific project costs, oftentimes, these figures are not broken down and thus are difficult to compare. Jersak et al. (2016) have provided an estimated cost for various sediment remediation methods internationally. The authors estimate that dredging costs can range from \$19 - \$2,089 USD per cubic meter, \$2 - \$209 USD per square meter for *in situ* capping, and less than \$1 USD per square meter for MNR (Jersak et al., 2016). Taken together, these factors present many challenges to finding an effective and economical solution to manage these contaminated sites.

4.1 *In situ* Technologies

Remediation methods that do not involve removing sediments from their natural environment are characterized as *in situ*. Examples of this technique include various capping, treatment, and containment methods that sometimes incorporate the mixing or strategic placement of microorganisms within the contaminated areas of the sediment (Reis et al., 2007). These methods are generally more straightforward and less expensive to carry out due to the sediments being able to remain in place for treatment (Majone et al., 2015). There is also a reduced risk of disturbing the contaminants, which could increase the exposure risk to the surrounding environment with *in situ* technologies (Reis et al., 2007). One downside of these techniques is that they require extensive knowledge and planning to ensure that they are carried out effectively (Majone et al., 2015), which could limit their use in some areas. Some other limiting factors include sediment type, lack of control over environmental conditions, length of time required, and reduced

efficacy when compared to some *ex situ* methods (Reis et al., 2007). According to Reis et al. (2007), *in situ* remediation methods are best suited for areas with low flow and in areas where the flow can be diverted during treatment.

4.1.1 Monitored Natural Recovery

Monitored Natural Recovery (MNR) is a form of bioremediation that capitalizes on native microbial populations and some of the biological, chemical, and physical processes that occur naturally within the sediments. Perelo (2009) describes this technique as making use of the “self-healing” properties that exist in the natural environment. More specifically, MNR allows the sediments to remain in place while permitting activities such as biological and chemical transformation and aquatic sedimentation to occur, which have the potential to reduce the bioavailability of the contaminants (Magar, 2001). Hence, transformation processes can reduce the toxicity of certain contaminants, and sorption processes can reduce bioavailability – these methods are typically preferred over burial methods or those that incorporate mixing techniques since these methods generally have a more permanent impact on the aquatic environment (US-EPA, 2005). For the contaminants that cannot be reduced to a less toxic form, e.g., metals, this method is not ideal. In these cases, a natural burial option is more common and preferred over a dispersal technique as an alternative (US-EPA, 2005). It is considered ideal to be used for low-risk areas that contain minimal contamination levels and in areas where there is little risk to human and environmental health (Magar and Wenning, 2006).

One feature specific to MNR that can make it appealing is that there is no action needed to start or maintain the process; however, to maximize the results, it requires careful decision making that results from a thorough site assessment (Perelo, 2009). Some natural processes can yield an increased risk to other areas or present additional challenges to a particular area if a proper risk assessment is not conducted prior to selecting this method (US EPA, 2005). It is considered a suitable strategy in areas where the contaminants have been buried or will be converted into a less toxic form, and where other remediation techniques would have more adverse impacts (Magar and Wenning, 2006). Another potential advantage of this technique is that it is considerably less expensive than methods like dredging and capping; however, it needs heavily involved monitoring over the long run (Perelo, 2009). One of the disadvantages of this method is that it leaves the contaminants undisturbed in their original location, which could leave the potential for reexposure in the future (US-EPA, 2005). It also can be a slow process and may not be ideal in situations that require a rapid solution. According to the US-EPA (2005), MNR has been chosen as a part of the remediation methods that are being used to treat contaminated sediment at a dozen US Superfund sites. It has also been used in combination with other remediation methods such as capping or dredging in some areas. The US-EPA (2005) has reported some success with this method as it has been able to reduce the contamination of some sediments, but long-term monitoring data is limited.

4.1.2 Capping Technologies

Capping technologies use specific isolation techniques to cover or produce a cap over the contaminated sediments. One of the benefits of these methods is that they reduce the interaction between the contaminant and the surrounding aquatic environment, in addition to minimizing the potential for pollutant transmission downstream (Perelo, 2009). The cost to implement a cap is also generally less expensive than other remediation technologies such as dredging (Perelo, 2009). Depending on the area, these caps are specially designed with various layers of certain permeable and impermeable components, and often incorporate granular materials such as sand, gravel or clean sediment (Reis et al., 2007). One unique property of capping techniques is that their suitability is largely defined by factors other than contaminant concentration or type, unlike other remediation methods. The major determining factors for whether capping is suitable for a certain location are site-specific, including the potential for cap disturbance or erosion (Reis et al., 2007). These factors also influence the composition and thickness of the cap required (Reis et al., 2007). Capping does have the potential to alter the native benthic community composition, so care must be taken to incorporate materials that will minimize this disruption (Perelo, 2009). There are two types of capping techniques that are used for addressing sediment contamination – passive or inactive capping and active or reactive capping.

4.1.3 Passive (Inactive) Capping

Passive capping involves developing a cap that consists of clean and nonreactive materials to isolate the contaminated sediments. The thickness of these caps is quite variable, but most are between 30-100 cm thick (Zhang et al., 2016). Thus, passive caps are not suitable for areas that are shallow or ecologically sensitive as a result of their thickness (Knox et al., 2012). The commonly used materials such as sand and gravel do not yield a perfect seal, so these caps are still susceptible to the effects of leaching and any disturbance events in the area (Knox and Paller, 2013). Nevertheless, this method is useful in areas that are not subject to severe disturbance events. Zhang et al. (2016) also report that this method is often used in areas with short-term risk potential. These caps do, however, have the potential to severely disturb the natural benthic ecosystems (Knox and Paller, 2013), and care must be taken to minimize the disturbance. But, this technique is quite economical with materials yielding reasonable costs (Zhang et al., 2016) when compared to other more involved methods. In a United States-based study, passive caps of varying thickness were shown to effectively isolate contaminated sediments that were previously dredged as a means of long-term storage and disposal (Ruiz and Schroeder, 2007). In Canada, a 35 cm passive cap was also used to contain heavy metal contamination in Lake Ontario (Azcue et al., 1998), which cost approximately \$300,000 CAD to implement (Zarull et al., 1999).

4.1.4 Active (Reactive) Capping

On the other hand, active caps are a newer technology that use chemically reactive materials to develop the cap. These caps are designed to reduce the bioavailability of contaminants, thereby stabilizing the contaminated sediment areas (Knox and Paller, 2013). Commonly used materials to develop active caps include organoclay, zeolite, apatite, biopolymer, iron, and activated carbon (Zhang et al., 2016). These caps are generally much thinner than passive caps, with thicknesses ranging from 10-30 cm (Zhang et al., 2016). This method has great potential to provide a cost-effective and reasonably permanent solution by sealing off the contaminants from the surrounding environment in areas that cannot implement a more traditional thick passive cap (Knox et al., 2012). Zeolites, which are brittle, solid rocks with easily controlled grain sizes, have the potential to be a powerful sorptive agent (Jacobs and Foerstner, 1999). If zeolites are ground relatively coarsely, Jacobs and Foerstner (1999) found that they accumulate on top of the sediments quite freely, which is ideal for implementation in existing water bodies. The same study showed that if the surfaces are treated with surfactants with cationic properties before application, zeolites can capture and contain various non-polar pollutants (Jacobs and Foerstner, 1999). In another study, Murphy et al. (2006) tested the effect of adding a 1.25 cm layer of an active sorbent such as activated carbon or organic soil in the middle of the polluted sediment and 15 cm sand layers. The authors found that with 1 cm of groundwater seepage per day, the activated carbon was able to seal the contaminated sediment for more than 60 years (Murphy et al., 2006). These authors also found that without any groundwater seepage, this method was effective with both activated carbon and organic soil for longer than 100 years and had a reduced PCB concentration in the bioactive area when compared to traditional sand caps (Murphy et al., 2006).

4.2 *Ex situ* Technologies

Ex situ techniques are those that are applied at an off-site location and require the sediments to be moved from their original area. Dredging is one of the most common forms of this technology and has been widely used throughout many areas of the world. This technology is significantly more expensive than some of the *in situ* methods (Reis et al., 2007), but is highly effective in areas with high contaminant levels. This method requires the sediments to be removed from their original location and to either be stored at an alternate location to be treated or to be disposed of off-site. One of the advantages of this technology is that it allows for a permanent solution for the affected area since the sediments are removed from the area. In the dredge and treat approach, these sediments can sometimes be treated and returned at a later date, which makes it a valuable tool for some applications. The dredge and dispose method is useful for very contaminated sediments but can be challenging in areas with dense human populations and a lack of available land as it would require an area dedicated to disposal. In addition to the high cost, this method also presents a high potential for disturbance of the area and potential

for remobilization of some contaminants during the process (Pourabadehei and Mulligan, 2016).

4.2.1 Dredging

Dredging consists of removing debris and sediments that have built up or have been contaminated from the bottom of various water bodies. In some areas, such as in major waterways, it is a routine process to keep waters navigable, and that counteracts the effects of sedimentation (NOAA, 2019). It is also used to address contaminated sediments in areas where aquatic ecosystems or human health may be at risk. Dredging is a popular remediation technique in the United States and Europe (Majone et al., 2015). In the United States, a paper by Mulligan et al. (2001) showed that roughly 10% of the sediments are contaminated, with 300 Mm³ being dredged annually. This paper also showed that between 3 and 12 Mm³ of these sediments were heavily contaminated (Mulligan et al., 2001). In Europe, it was estimated that 100-200 Mm³ were dredged every year as a result of heavy contamination levels (Bartone et al., 2004). Despite its advantages and popularity in some areas, it is very expensive to conduct. More specifically, the cost to dispose of the dredged sediments is very high, especially if the contamination levels exceed the approved guidelines in the area (Walker et al., 2013). Sediments contaminated with PCBs from the Manistique, Shiawassee, and Maumee Rivers as well as from the River Raisin were dredged, and the costs ranged from \$5 to \$25 million USD in these areas (Zarull et al., 1999). In addition, PAHs, mercury, and lead in the Kalamazoo River were present, which cost \$900,000 USD to dredge (Zarull et al., 1999). The Niagra River also cost \$14 million USD to dredge as a result of dioxin contamination (Zarull et al., 1999). The other option is to treat and reuse the contaminated sediments, but this option has been adopted to a much smaller extent due to the even higher costs and uncertainty about the reuse potential for the sediments (Majone et al., 2015). Another potential issue with dredging is the requirement for space to dispose of the contaminated sediments – not every area has enough space to dedicate an area for contaminated sediments. Dredging also has the potential to disturb and spread the contaminants in the affected areas, which increases their risk through enhanced mobility and availability (Pourabadehei and Mulligan, 2016). Perelo (2009) also points out that this technique does not actually eliminate the contaminants but instead moves them to another location, where it can have adverse effects as well.

4.3 Remediation Technique Combinations

Under certain circumstances, a combination of more than one remediation technique might yield the best results. A combination of methods is usually used in large areas where the site-specific factors vary at different points along the body of water. In some extensive cases, a combination approach may also be used to reduce the costs or environmental impact associated with the chosen techniques. As this review has

previously noted, a common combination of techniques is the dredging of an area followed by capping the contaminated sediments. In some cases, this combination of techniques is referred to as *ex situ* capping (Wang et al., 2004). Another combination might be dredging or capping in one area of a river, while MNR is carried out at another (Jersak et al., 2016). In the end, the most suitable method for tackling contaminated sediments varies depending on the specific factors that pertain to that area, as well as the anticipated environmental and economic impacts involved.

5.0 Laboratory Study

5.1. Introduction

This study was conducted as a preliminary indicator of the potential toxicity of various sediment samples based on the preference and avoidance behaviours of *Lumbriculus variegatus*. The purpose of this study was to determine if the worms preferred a sediment sample over a quartz sand control. If the worms avoided a particular sediment this would indicate that there were potential biological effects of that sample. The results from this study will be used to inform future research on the potential biological effects of the sediments in these areas.

The sediment samples that were used in this study were collected from various sites in the Saskatchewan River Basin. The sites from upstream to downstream include Lake Diefenbaker (including Saskatchewan Landing (SL) and Miry Bay (M)), Codette Lake (C, two replicates), Tobin Lake (T, three replicates), and Cumberland Lake which includes Mossy Mouth (MM), Mossy Fan Out (MFO), North End (NE) and South End (SE). Each sample was analyzed previously for the presence of 24 trace elements by ICPMS. The second Codette Lake sample, the Saskatchewan Landing sample, and the North and South End samples were the only ones that contained a concentration of nickel that exceeded the CCME PEL guidelines. All of the other samples were below the available CCME PEL guidelines for all of the elements.

5.2 Experimental Design

The assay was conducted using a modified version of the methods described in West and Ankley (1998). The test organism *L. variegatus* was used for this trial because of its natural characteristics that make it easy to handle, straightforward to monitor and recover from the sediments, its ability to move freely between the sediments and surrounding water, and its status as a benthic organism (West and Ankley, 1998).

Prior to the beginning of the experiment, a representative sample of all 11 sediment samples and a quartz sand control were weighed and then dried in an incubator (Binder model BF 53-UL) at 105°C for 72 hours. Once dried, the samples were weighed again, and the dry weight equivalent of 10 g of each sediment and the control were calculated (Equation 1 and 2).

$$\text{Water Content} = \frac{\text{Wet Sediment Mass} - \text{Dry Sediment Mass}}{\text{Wet Sediment Mass}} \times 100$$

Equation 1. The formula used to calculate the percent water content in each sediment sample.

$$\text{Wet Mass of Sample Needed} = \frac{\text{Desired Dry Sediment Mass}}{(1 - \% \text{ water in sediment})}$$

Equation 2. The formula used to determine the appropriate mass of the wet sediment samples needed in order to have a 10 g dry weight equivalent of each sample.

The experiment was designed to include $n=4$ replicates of each sample for a total of 48 samples. Each sample was housed in a 125-ml glass jar which was divided in half by a plastic divider that allowed for a shared water source within each jar. For each sediment sample, there was a 10-g dry weight equivalent on one side of the divider, and on the other side, there was a 10-g equivalent of quartz sand. For each of the control samples, there was a 10-g dry weight equivalent of quartz sand on both sides of the divider. The jars were set up the night before the start of the experiment and were initially filled with approximately 100 ml of water (pH of ~ 7.8 ; DO between 75-95%; ammonia >0.1 mg/L; chlorine, nitrite and nitrate < 0.1 mg/L) and left to equilibrate overnight. The following morning, five randomly selected worms were added on each side of the dividers and given approximately 5 minutes to settle. The dividers were then carefully removed, and the jars were randomly distributed on both shelves of a two-tiered metal rack. The lighting source on each tier of the rack was a Wills 165-W aquarium light that was set to deliver 16 hours of light and 8 hours of darkness. At the 48- and 96-hour mark, approximately 25 ml of water was carefully removed from each jar and replaced with approximately 50 ml of freshwater to account for evaporation. The temperature in the laboratory for the duration of the experiment was $20.0^{\circ}\text{C} \pm 0.3^{\circ}\text{C}$. At the end of a 144-hour period, the worms were collected by replacing the divider and slowly decanting half of the water from each jar while holding the divider in place. The sand was carefully removed from one side of the jar and placed into a large plastic weigh boat where the worms were counted. The divider was then removed, and the worms in the sediment were placed into a separate plastic weigh boat to be counted. For the control jars, the sand on each side of the divider was removed, and the worms were counted. The preference behaviour of the worms was calculated using Equation 3.

$$\% \text{ Preference} = \frac{\text{Number of Worms in Sediment} - \text{Number of Worms in Sand}^*}{\text{Total Number of Worms Recovered}} \times 100$$

Equation 3. The formula used to determine the preference of the worms in each jar. The “*” indicates that for the control jars the same formula was used with one side of the jar being subtracted from the other.

5.3 Results

The preference and avoidance behavior of the *L. variegatus* worms was analyzed in this study. The number of worms on each side of the divider was counted after 144 hours to determine their preference or avoidance of each sediment and sand sample. A total of 453 worms (94%) were recovered at the end of the experiment. Overall, the worms showed a strong preference for the sediment samples over the quartz sand controls. The worms showed an average of $90\% \pm 12\%$ preference for the first Codette Lake sample (C1) and a $100\% \pm 0\%$ preference for the second Codette Lake sample (C2). The Miry Bay sample (M) yielded a $100\% \pm 0\%$ preference, as did the Mossy Fan Out (MFO) and Mossy Mouth (MM) samples. The North End (NE) sample yielded an average $88.9\% \pm 13\%$ preference, while the South End (SE) sample showed an average $93.8\% \pm 13\%$ preference. Saskatchewan Landing (SL) had an average $85\% \pm 13\%$ preference for the sediment. Lastly, the Tobin Lake samples showed an average of $80\% \pm$

16% (T4), 100% ± 0% (T5), and 100% ± 0% (T6) preference for the sediments. The control samples yielded an average of 15% ± 70% preference for the quartz sand. These results are shown in Figure 1.

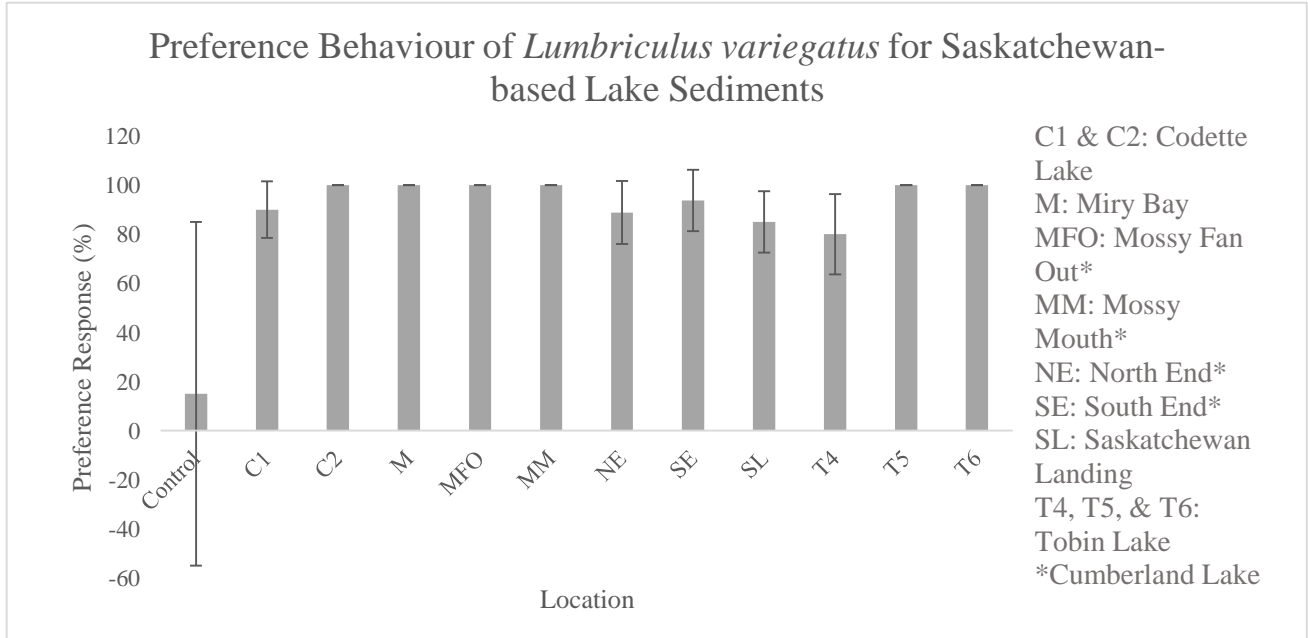
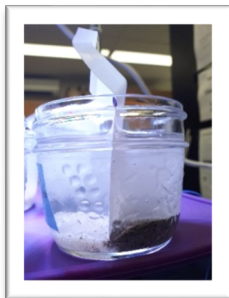


Figure 1. Preference behavior of *L. variegatus* for 11 sediment samples from Saskatchewan lakes. 0% indicates no preference, while positive numbers indicate preference for the natural sediments and negative values indicate preference for the controls.

6.0 Plain Language Summary

The Saskatchewan River Delta has changed over the past century. Some of these changes include different water flow patterns, less water reaching the delta, and loss of sediment. All of these changes have affected the people who live in the delta, as well as the ability of the area to thrive. Replenishing the depleted sediments is needed in this area, but some sediments are contaminated which presents challenges to restoring a healthy environment. Sediments are an important habitat and help the aquatic environment to function, but they could have negative effects on the organisms that rely on the delta if contaminants are present at harmful levels.

The California blackworm (*Lumbriculus variegatus*) is a sediment-dwelling organism that has been used to learn about the specific composition of certain sediments. These worms show obvious preference and avoidance behaviors based on particular features of the sediments such as nutrient availability and the presence of contaminants. As part of a summer MSEM project, a study was designed to assess the behavior of a total of 480 worms in 11 different types of sediment from Saskatchewan lakes with known contaminant levels and quartz sand.



The lake locations included Codette Lake, Tobin Lake, Saskatchewan Landing, Cumberland Lake, which includes Mossy Mouth, Mossy Fan Out, North End, and South End, and Miry Bay. There were 44 glass jars with a plastic divider in the middle that separated equal amounts of one sediment variety from a sample of quartz sand. There were also another 4 glass jars with dividers that had equal amounts of quartz sand on either side. Each jar contained a total of 10 worms, with 5 being placed on either side of the divider at the beginning of the trial. Once the trial began, the dividers were removed so that the worms had full access to both sides. All of the jars also contained a set volume of water that was changed on days 2 and 4 of the experiment. The study was conducted over a period of 6 days, with the dividers being replaced at the end of the study and the worms on each side being counted to indicate their preference for certain samples and avoidance of others. The results of the study showed a very strong preference for the sediment side of each jar, with values of 80% and above.

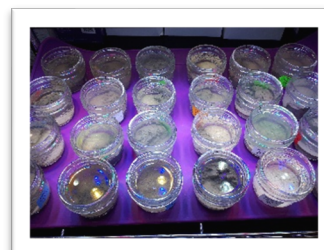
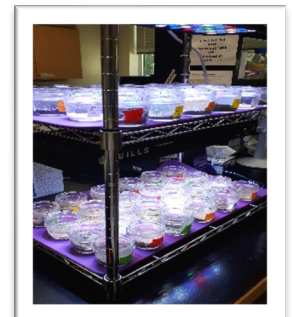
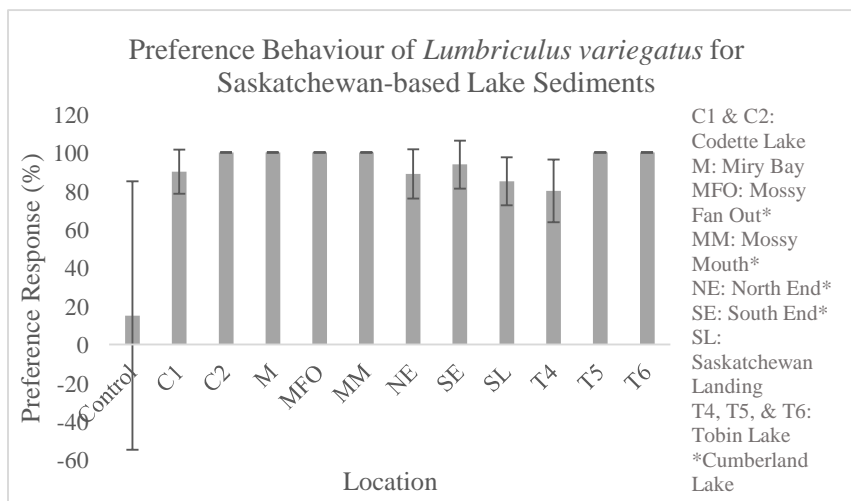


Figure 2. Preference behavior of *L. variegatus* for 11 sediment samples from Saskatchewan lakes. 0% indicates no preference, while positive numbers indicate preference for the natural sediments and negative values indicate preference for the controls.

7.0 Towards a Solution

The remediation technique that is best suited for the Saskatchewan River Delta is largely dependent on the site-specific factors of the particular area, such as sediment type, level of contamination, environmental conditions, and economic considerations. Consideration must also be given to the stakeholders and rights-holders involved, as they will ultimately be the ones whom these decisions impact the most.

If economic considerations are the highest priority and financial resources are lacking in the immediate future, then MNR might be the best choice for areas with low levels of contamination. MNR is ideal in areas that do not possess a high ecotoxicological risk and that do not require a rapid solution. However, it is important to note that this method is only effective for contaminants that are able to be reduced to a less toxic form. This method also requires an extensive site assessment to be conducted in addition to long term monitoring, which means that qualified personnel needs to be available and involved. As a result, the long-term monitoring that is necessary for this method will incur some costs in the future, but it could be implemented in areas of the delta that fit the criteria, and that may have funding available in the future.

If the area has moderate to high levels of contamination or contains contaminants that are not easily reduced, then one of the *in situ* capping methods might be better suited. These methods are ideal in areas that are not subject to large levels of disturbances and contain a water level that is adequate to allow for the cap. A passive cap is generally the more inexpensive of the two options but will require a deeper area due to the increased thickness compared to an active cap. It is ideal for areas with high ecological sensitivity, as the cap contains inert materials that will allow for minimal disruption to the area. Active caps are useful for contaminated areas that have the potential to be reduced from the materials contained within the cap. These caps are a newer technology and can be significantly thinner than the more traditional passive cap, which makes them useful in shallower areas. This option is more expensive than passive capping, but it allows for specific contaminants to be targeted and reduced or, in some cases, even neutralized. Thus, either capping variation might be a practical and reasonably effective solution for the relatively undisturbed areas of the Saskatchewan River Delta.

For areas that are subject to high levels of disturbance and contain high concentrations of contaminants, dredging might be the most suitable option since it allows for the removal of the contaminated sediments. While this option is more costly than *in situ* methods, it might be the most feasible for large and highly contaminated areas in the long run so that the contaminants are not constantly remobilized. This method provides an alternative for the areas of the delta that are not ideal for MNR and/or capping techniques and allows for a dredge and treat or dredge and dispose option.

Another option is to bring sediments in from another location to fill in the depleted areas. These sediments would have to be brought in via *ex situ* methods, but would provide an alternative that allows for clean sediments to be introduced. This method would likely be more costly than some of the other *in situ* methods, but would allow for uncontaminated sediments to be brought in to replenish areas with high contamination levels or in areas that do not have other sources of sediments available.

Overall, the Saskatchewan River Delta is an incredibly diverse and unique area. For the sections that contain contaminated sediments, there are a variety of remediation options available that could provide a reasonable solution. It is likely that one remediation technique will not be best suited for all areas, and that a combination approach will be the best option for this area.

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Appendices

Appendix A - ENVS992: Project Plan

Student: *Elaine Bird*

Partner: *Gary Carriere*

Faculty Advisor: *Tim Jardine & Markus Brinkmann*

Date: *April 13, 2020*

Project title

An Overview of Common Sediment Contaminants and Remediation Methods in North America and Europe

Partner organization name, location, and type (Private, Government, Plural)

Cumberland House Fishermen's Co-operative

P.O. Box 69

Cumberland House, SK S0E 0S0

Partner contact details

E-mail: gcarriere@sasktel.net

Phone: 306-888-7530

Faculty advisor

Tim Jardine: tim.jardine@usask.ca

Markus Brinkmann: markus.brinkmann@usask.ca

Project location, and working arrangements if students not based on campus

Project to be completed remotely or on campus if the campus reopens in the summer months

Location and approximate timing of any field work

N/A unless campus reopens

Brief description of the problem to be addressed and tentative objectives

There are vast differences in sediment remediation approaches based on the geographic location of the area. These differences are a result of variations in availability of resources – such as physical space and capital – in these areas. This literature review will explore the major remediation methods that are currently used in North America and Europe. It will also outline the most common sediment contaminants in these areas as well as any guidelines that are in effect to

regulate their impacts. The overall goal is to gain a greater understanding of the most widely used sediment remediation techniques and to define any benchmark values with respect to contaminant impacts and economic considerations. This information will then be available for the Saskatchewan River Delta sediment restoration efforts as part of a larger ongoing project.

Objectives:

- Outline the most commonly used sediment remediation techniques in North America and Europe
- Establish threshold values for toxicity by contaminant
- Highlight economic implications of each technique

Brief overview of the methods and disciplines that will be employed and tentative deliverables

This project consists of a literature review that will provide an overview of common sediment remediation methods in North America and Europe. The information obtained is designed to contribute to a larger project being conducted in the Saskatchewan River Delta.

Deliverables:

1. Literature review of sediment remediation techniques
2. Create an informative tool to be used in the community

Gantt Chart that lays out deliverables for partners and project requirements

See attached Excel document

List of any required resources, and how these will be provided (e.g. travel funding, equipment, software, etc.).

None