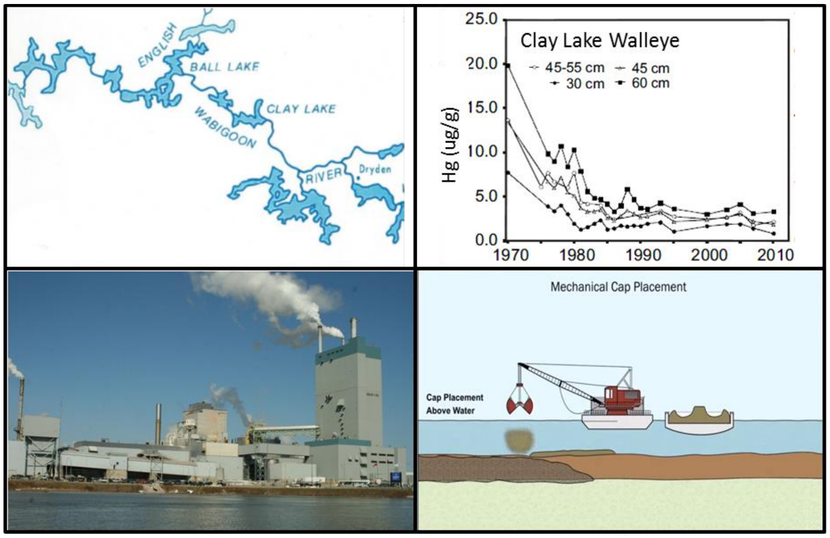
Advice on Mercury Remediation Options for the Wabigoon-English River System

Final Report



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(Grassy Narrows First Nation) - Ontario – Canada Working Group on Concerns Related to Mercury

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Summary

This report reviews approaches to reduce mercury contamination in aquatic systems, and identifies options with the potential to be applied in the Wabigoon-English River system. Approximately 10 tonnes of mercury were released to the Wabigoon River between 1962 and 1969 from a chlor-alkali facility at Dryden, Ontario, resulting in highly contaminated waters, sediments and biota. Mercury contamination in fish was observed at least as far as Tetu Lake, 250 km downstream of Dryden. After measures were carried out in the early 1970s to reduce mercury releases from the chlor-alkali facility, mercury concentrations quickly began to decline in sediments and fish, but unfortunately these concentrations stabilized or declined very slowly since the 1990s, and remained 2-10 times above regional background levels in Clay Lake walleye in 2010.

Existing data indicate that fish mercury concentrations are elevated because mercury concentrations in sediments are elevated. The persistence of mercury contamination in the sediments is likely caused by one or both of the following: (1) if mercury releases still occur at meaningful levels from the site of the former chlor-alkali facility, or (2) if contaminated river sediments between Dryden and Clay Lake are still moving slowly downstream into Clay Lake. Additional data are needed to investigate each of these possibilities.

Overall, we think that that remediation of at least some parts of mercury-contaminated Wabigoon River system is feasible. If actions were taken to reduce mercury levels in contaminated sediments, fish mercury concentrations would also be expected to decline. Actions that would reduce the efficiency of converting inorganic mercury (the form released from the former chlor-alkali facility) to methylmercury (the form in fish), or options that reduce the biological uptake of methylmercury in the food web, are also potential options to reduce mercury in fish. Careful consideration needs to be given to the potential for different responses at different locations for any given remediation scheme.

Because mercury contamination is carried downstream, a strategy is recommended that begins at Dryden. First, if recommended field studies indicate that mercury releases continue to occur from the former chlor-alkali facility site at environmentally relevant rates, steps should be taken to eliminate those releases. Second, if field studies indicate that sediments in the Wabigoon River between Dryden and Clay Lake are still an important source of inorganic mercury or methylmercury to overlying waters in the river and downstream, hydraulic dredging and/or armoured capping are potential options in localized areas of the upper river.

The third component of remediation is to focus on Clay Lake. The need for remedial measures in Clay Lake should be evaluated after remediation of the Wabigoon River upstream. We ranked the top candidates of remediation options for Clay Lake based on four criteria. These were 1) efficacy of the method, 2) possible damage caused to the ecosystem during its application, 3) time required to apply the remediation method, and 4) cost.

The top ranked method for further evaluation was Enhanced Natural Recovery (ENR). Low-mercury solids would be added to Clay Lake waters, where they would settle and be naturally mixed into surface sediments, thus diluting the mercury in the sediments. A large supply of low-mercury solids is available in Wabigoon Lake. This method is highly rated for 3 of the 4 criteria (efficacy, minimal damage and lowest cost). It ranked lower than other options in terms of the expected time required.

The second ranked option for further evaluation in Clay Lake is to directly lower mercury concentrations in Clay Lake sediments by applying a cap, which would involve adding low-mercury materials (*e.g.* 10 cm) onto the surface of Clay Lake sediments. This method is highly rated in terms of efficacy and moderately for the other 3 criteria.

The third ranked option was addition of activated carbon (*e.g.* Sedimite) to surface sediments, which would bind the mercury making it unavailable for bioaccumulation. It is not rated highly because although it has been shown to be initially effective the duration of its effectiveness is not yet known. Also, a single application to Clay Lake sediments would a cost about $1.5 billion. It is the most expensive alternative considered.

The fourth rated option is bank-to-bank dredging. It ranked lowest because of the possible damage and disruption done to the ecosystem during the dredging procedure and because of its high cost – estimated to be about $940 million.

Nitrate additions or aeration should also be further assessed for Clay Lake. These approaches would reduce methylmercury in fish but not total mercury in sediments, and are seen as supplementary options rather than primary solutions. It may be possible to address the time required for ENR to remediate the problem by combining it with aeration or nitrate additions that provide short-term benefits while ENR reduces inorganic mercury levels in Clay Lake sediments.

Other combinations of approaches could also be considered for Clay Lake, *e.g.* localized dredging, capping and nitrate additions. There is currently insufficient information to evaluate the likely success or costs of these options.

Monitored Natural Recovery is not considered to be a viable option because the system is presently either not recovering or is recovering at an imperceptible rate.

A key component of any remediation strategy is to use adaptive management. This approach is based on reviewing results as remediation progresses, and adjusting plans if the data suggest changes that would lead to improvements in performance.

Field studies are recommended to provide updated mercury information in water, sediments and biota in the Wabigoon-English River system. These data are needed to provide a measure of the present-day geographic extent of contamination, and to better understand the behaviour of mercury in the system, including areas that are ongoing sources of mercury. In some cases, data have not been updated since the 1980s (mercury in water). In other cases, updated spatial coverage is needed (mercury in sediments and fish). These data would also provide a baseline set of measurements that could be compared to data collected after remediation begins, thus allowing for the evaluation of remedial actions.

Acknowledgments

The authors are grateful for the assistance of Carol Kelly, Cody Beals, and David Hutchinson in the analysis of information and preparation of the report. We thank the University of North Carolina at Pembroke for financial support.

1 Introduction

Approximately 10 tonnes of mercury were released to the Wabigoon River between 1962 and 1969 from a chlor-alkali facility at Dryden, Ontario (Figure 1), (Parks and Hamilton, 1987). This mercury was transported in the Wabigoon River, resulting in highly contaminated waters, sediments and biota downstream. The highest levels of mercury in sediments exceeded 20,000 ng/g (parts per billion) in some samples just downstream of Dryden. These concentrations are greater than current background levels of 100 - 200 ng/g (Sellers, 2005) by a factor of roughly 100 to 200. Mercury levels in adult walleye (50 cm length) in Clay Lake, 87 km downstream of Dryden, reached concentrations of 15 parts per million (ppm) in 1970 (Parks and Hamilton, 1987), roughly 30 times higher than regional background levels estimated by Neff *et al.* (2012). Because fish mercury concentrations in the Wabigoon River system were not monitored prior to the presence of the chlor-alkali facility, the actual increase in fish mercury due to contamination is not known. Mercury contamination in fish was observed at least as far as Tetu Lake, 250 km downstream of Dryden, although at lower levels than upstream in Clay Lake (~ up to 2 ppm, Neff *et al.*, 2012).

Measures began in 1970 to prevent the pulp mill in Dryden from releasing more mercury into the Wabigoon River, but actions were not taken directly in the Wabigoon River system to lower levels of mercury already in the system. Natural recovery has resulted in a decline in mercury levels in fish (Figure 2) and sediments (Figure 3). Unfortunately, fish and sediment mercury levels in some locations remain well above background (Sellers, 2014) and are declining very slowly or stabilizing. The most recent measurements for Clay Lake sediments show that mercury concentrations in 2004 were still as much as 20 times regional background levels. Furthermore, sediment data from different locations suggest that some of the mercury contamination may be moving downstream, resulting in gradually increasing levels in sediments at the downstream end of the system. In terms of current-day fish mercury levels, adult walleye from Clay Lake in the range of 1-3 ppm had concentrations in 2010 that were 2 to 6 times higher than the 0.5 ppm guideline for commercial sale. These concentrations invoke consumption advisories to reduce health risks associated with fish consumption: no consumption of any walleye for children under 15 and women of child-bearing age, and no consumption of any walleye longer than 40 cm for the general adult population (Government of Ontario, 2015).

The persistence of elevated levels of mercury in fish and in sediment 45-50 years after the original contamination, in combination with ongoing human health problems associated with exposure to mercury through fish, was partly responsible for the formation of the Asubpeeschoseewagong Netum Anishinabek (Grassy Narrows First Nation; ANA) – Ontario – Canada Working Group in 2013. This report provides advice to the Working Group on remediation options for the Wabigoon-English River System.

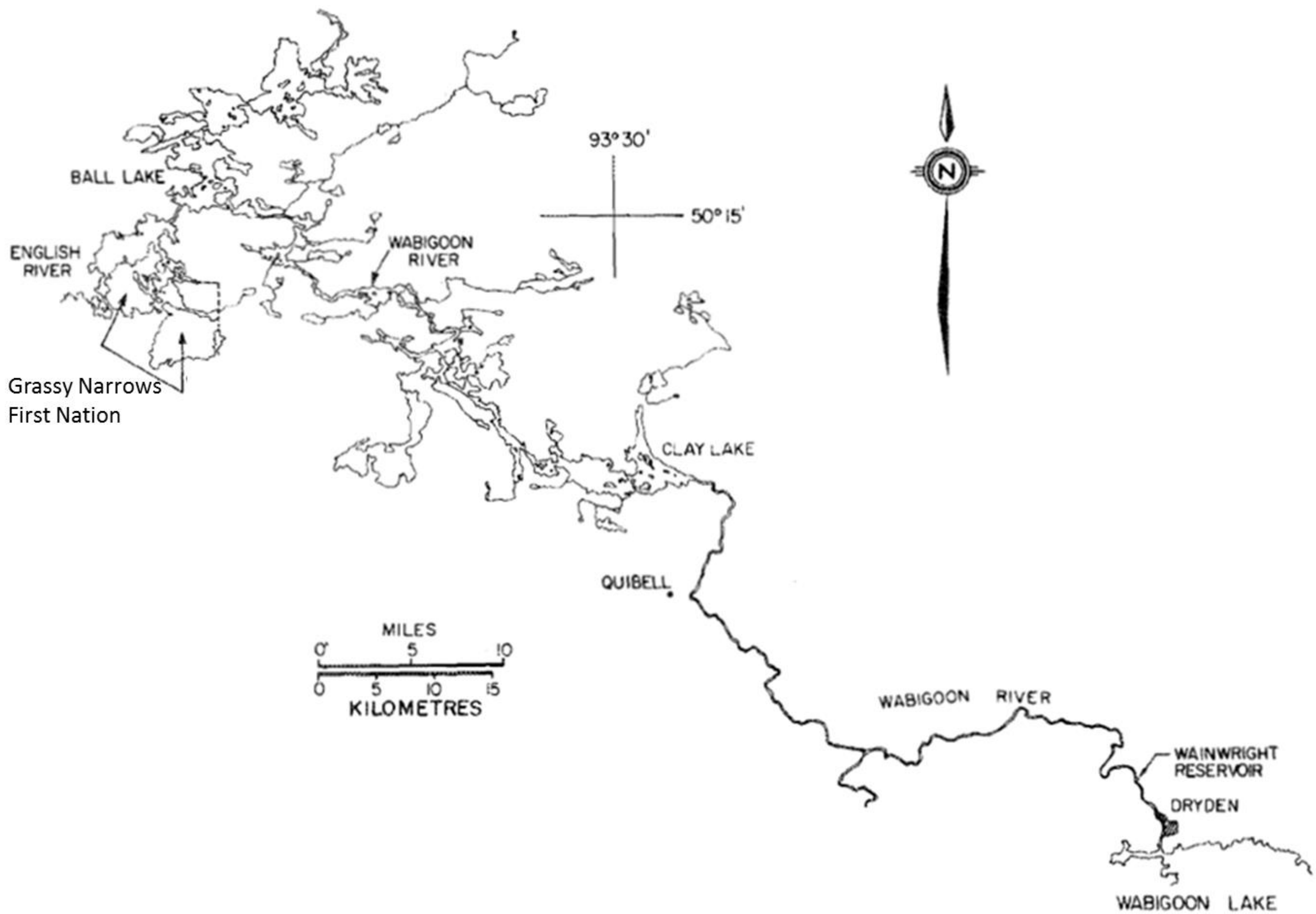

Figure 1. Map of Wabigoon-English River System downstream of Dryden. From Rudd *et al.* (1983)

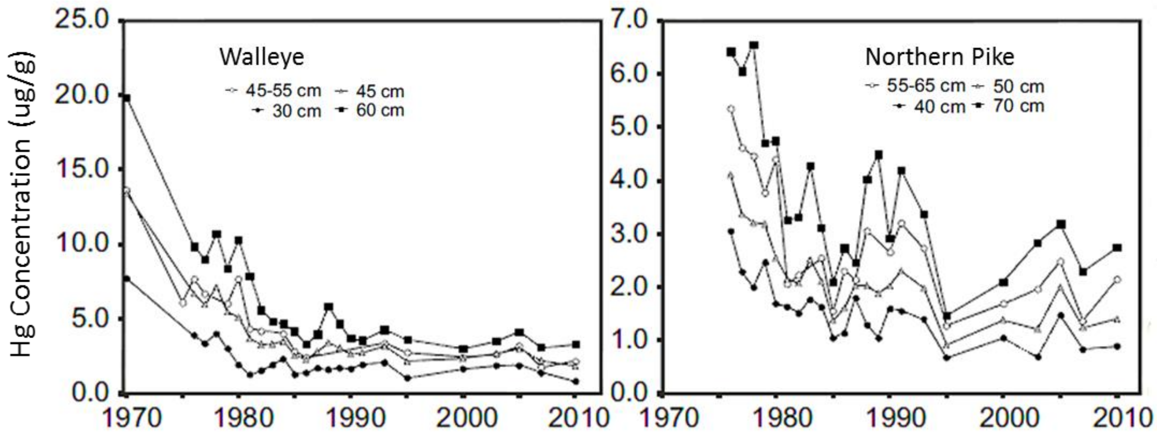

Figure 2. Mercury concentrations in walleye and northern pike of different sizes in Clay Lake, 1970-2010. Modified from Neff *et al.* (2012)

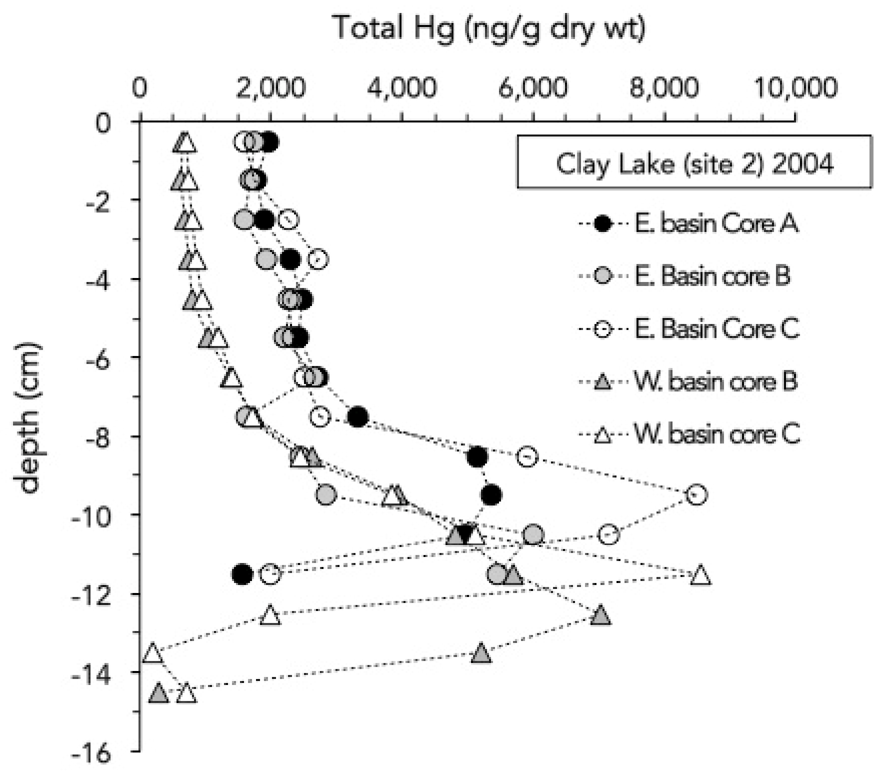

Figure 3. Mercury concentration profile in Clay Lake sediments. From Sellers (2005)

2 Study Objectives and Scope

The objectives of the study were to:

1) Research, review and analyze mercury remediation options available for the Wabigoon-English River system, and

2) Prepare a report for the Mercury Working Group that:
    i. identifies all of the mercury remediation options available, including well tested and novel approaches;
    ii. analyzes the suitability and effectiveness of each option for the Wabigoon River system;
    iii. provides recommendations on leading candidates for remediation of the Wabigoon river system, based on available information; and
    iv. identifies any further research that may be required to obtain information necessary to make a recommendation on which remediation option(s) should be used in the Wabigoon-English River system, and sets out a detailed research plan for any required further research.

It is also important to note the limits on the scope of the report:

- It is not meant to provide detailed information on any specific remediation option (technical, cost, schedule). Such analyses could follow this initial examination of remediation options if deemed warranted by the Working Group.
- Climate change and logging are both relevant issues for the Wabigoon-English River system, but they are not addressed here. Climate change has the potential to affect fish mercury concentrations (*e.g.* Harris et al., 2015, 2012; Pinkney *et al*., 2014), and increased fish mercury levels have been associated with logging (*e.g.* Garcia and Carignan (2005, 2000). Our opinion at this time, without detailed investigation, is that these factors are secondary in the upstream lakes, but possibly of importance in the downstream lakes where point source mercury is not as important.
- The study scope does not include an assessment of ecological and health risks. The Ontario Government has indicated that an ecological and human health risk assessment is required in order to clarify the need for remediation. ANA's position is that the detrimental health effects of mercury exposure are evident, some of which are documented in the work most recently published by Takoaka *et al.* (2014). These topics are being addressed in a separate proposal submitted by others.

3 Why is Mercury Still Elevated in the Wabigoon-English River System?

The basic problem in the Wabigoon-English River System is the persistence of elevated fish mercury concentrations. This is caused by the persistence of inorganic mercury contamination in surface sediments, which stimulates the production of methylmercury that is found in fish.

Actions to reduce mercury releases from the chlor-alkali facility reduced loads to the Wabigoon river by roughly 99% from peak release rates of about 1100 kg/yr. (average for 1962-1970) to approximately 10 kg/yr. by the mid to late 1970s (Government of Canada/Government of Ontario, 1984). While 10 kg/yr. is only 1% of the peak rate of mercury discharge, it would still be very relevant in terms of background mercury loads.[1] Lower estimates of ongoing releases from the Dryden chlor-alkali site were also identified (~1.1 to 1.5 kg/yr. in 1982 (Cosway, 2001)). The chlor-alkali facility was decommissioned in the 1970s and there is now a pulp and paper facility on the site. Mercury loads from some locations on the site to the Wabigoon River are currently monitored, but these data are not presently available[2]. The Ontario Ministry of Environment and Climate Change (MOECC) indicated verbally that concentrations are low at these sites. Based on a presentation by MOECC in January 2016 in Toronto, our view is that sampling locations are limited and should be expanded (especially at possible ground water seeps from the former site of the chlor-alkali facility, and or at present or former discharge outfalls) and additional data are needed to determine whether or not mercury releases from the site of the former chlor-alkali facility are important.

Mercury concentrations in sediments and biota began to decline after mercury discharges were greatly reduced in the 1970s (Figure 2, Figure 3). The decline of mercury concentrations in sediments was controlled by how quickly mercury contamination was eliminated from sediments, for example by burial, and by reduction of residual mercury loads from the site of the chlor-alkali facility. Any remedy to accelerate the recovery of the system would need to address both these issues (residual loading, if meaningful, and how quickly mercury concentrations in sediments can be returned to pre-contamination levels).

Most of the mercury contamination from chlor-alkali facilities is inorganic. A small, but very important fraction is methylmercury, a toxic organic form that is the dominant form in fish because it accumulates more effectively than inorganic mercury. Methylmercury is produced naturally from inorganic mercury by bacteria. Several studies have indicated that methylmercury concentrations increase as inorganic mercury concentrations increase in aquatic systems (*e.g.* Harris *et al.*, 2007; Rudd *et al.*, 2013a). While the chlor-alkali facility releases from the Dryden site were dominated by inorganic mercury, methylmercury concentrations in water and fish increased dramatically above background levels. This indicates that methylmercury was (and is) being produced due to the inorganic mercury contamination present in the system. It

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[1] For comparison, 10 kg/yr would be 10 fold bigger than mercury inflows from Wabigoon Lake (which is upstream of the plant site), on the order of 1 kg/yr (assuming 2 ng/L in Wabigoon Lake outflows in absence of recent data, and a mean annual flow of 14 m3/s).

[2] MOECC indicated that these mercury monitoring data are owned by third parties and cannot be released without permission. Requests have been made to release the data but permission has not yet been secured.

follows that if inorganic mercury concentrations can be reduced at sites where methylation occurs, methylmercury levels, including concentrations in fish, would decline. This explains why fish mercury levels rose to very high concentrations and subsequently declined as inorganic mercury levels in sediments declined.

In Clay Lake, the recovery trend for mercury in surface sediments (mostly inorganic) and mercury in fish (mostly methylmercury) are very similar (Figure 4). As inorganic mercury levels declined, so did methylmercury, and further reductions in inorganic mercury would likely further decrease fish mercury levels. It is not yet clear whether the decline in fish mercury in Clay Lake was due to declines in inorganic mercury concentrations and methylmercury production within Clay Lake, or a reduction in upstream sediment mercury levels, followed by decreases in upstream methylmercury loads to Clay Lake, or a combination of both.

Farther downstream, data from Ball Lake suggest that trends for inorganic mercury and methylmercury may be decoupled in the lake. In the south basin of Ball Lake, which is influenced by the Wabigoon River, sediment concentrations of inorganic mercury are slowly increasing (Sellers, 2008). In contrast, mercury levels in biota from Ball Lake (crayfish in the south basin, fish from the north basin) have declined since the 1970s (Neff et al., 2012; Sellers, 2008), the same trend seen for fish upstream in Clay Lake. Methylmercury is the dominant form of total mercury in fish, and an important component of total mercury in crayfish. Concentrations of total mercury in sediments and crayfish (which stay resident in the north basin) show no signs of contamination in the north basin, upstream of the influence of the Wabigoon River.

Decreasing mercury levels in biota and increasing concentrations in sediments in Ball Lake could occur if:

- methylmercury levels in Ball Lake biota are more influenced by high levels in inflows from upstream, including Clay Lake, rather than methylmercury production in Ball Lake itself; and/or
- fish caught in the north basin of Ball Lake spent important portions of their lifespan in contaminated areas.

The above discussions regarding sources of inorganic and methylmercury to Clay Lake and Ball Lake demonstrate the importance of understanding the extent to which methylmercury at any given location is produced locally or imported from upstream. This has important implications for the design of remediation options, and the expected benefits. For example, will remediation actions at one location (e.g. Clay Lake) quickly show benefits downstream (e.g. Ball Lake)?

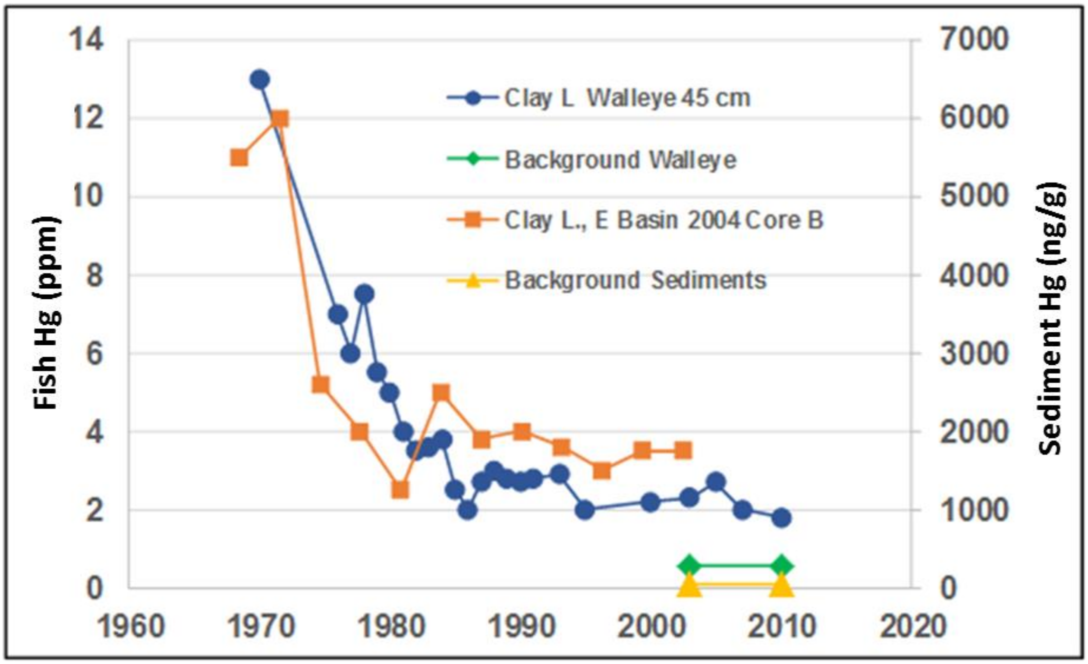

Figure 4. Time courses of mercury concentrations in surface sediments of the east basin of Clay Lake and in 45 cm walleye. The data show that fish mercury concentrations are closely related to inorganic mercury concentrations in the surface sediments. Walleye data are from Neff *et al.* (2012). Sediment Hg data are calculated from P. Sellers (unpublished data).

The above discussions do not explain why fish mercury levels in Clay Lake declined relatively quickly in the 1970s, but stabilized at levels in the range of 1-3 ppm in the 1990s and have declined little since then. The answer is that fish mercury levels have not returned to background levels because surface sediments in Clay Lake and possibly upstream of Clay Lake remain contaminated with inorganic mercury. Possibilities to explain ongoing contamination in surface sediments include:

(1) if there are significant ongoing releases of mercury from the site of the former chlor-alkali facility. Estimates of mercury releases circa 1980 were capable of sustaining mercury levels above background in the river, and there are no recently available data. Mercury releases from the site to the Wabigoon River are currently monitored at a limited number of locations at the site. Additional studies are needed to examine the potential for mercury to be released to the river from other locations at the site;

(2) after excess mercury loading to sediments stops, it takes time for surface sediments to eliminate mercury.[3] Based on limited available information[4] it would take

[3] There are two mechanisms involved: burial and fluxes to the water column. New solids with lower mercury levels settle and bury sediments with higher concentrations. Because the upper few cm of sediments mix naturally, the removal process takes longer, and follows a different pattern than if new solids were just layered on older solids from year to year. Mixing results in a recovery where the *relative* decline is expected to be the same each year (*e.g.* 10% of the ultimate response per year). At first, when concentrations are higher, the absolute decline in concentration is faster than during later years. This has been observed for Clay Lake sediments, for example (Figure 3, Figure 4) and is consistent with a mixed surface layer of sediments.

approximately 14-30 years for solids to eliminate contamination and reach sediment concentrations of 100-200 ng/g. At those rates, mercury levels in Clay Lake would have returned to background levels by now if waters entering Clay Lake had background mercury concentrations during the same period. This suggests that there is ongoing contamination entering Clay Lake from upstream. Whether the source of contamination to Clay Lake is residual seepage from the site of the former chlor-alkali facility or mercury loading from contaminated sediments in the Wabigoon River is not clear but needs to be determined; and/or

(3) it is also possible that Clay Lake has different sediment zones that eliminate mercury at different rates. The cores used to estimate mass sedimentation rates may not reflect all sediment zones in the lake. The potential exists for some zones that recovery more slowly and supply ongoing contamination to other areas.

The slow movement of mercury contamination downstream of the original source has the potential to cause different recovery trends at different locations along the system. Locations closest to the original source would increase first, and to the greatest degree. After mercury releases from the facility stop, sediment mercury concentrations near the original source begin to decline, as low-mercury solids from upstream mix with contaminated sediments (Figure 5). Some of the contamination is buried, but some is remobilized to the water column and travels downstream, acting as a continuing source for downstream sites. Downstream locations would reach peak concentrations later than near the original source, although at lower levels. This is shown conceptually in Figure 5. There is evidence to support this idea, as mercury concentrations in the south basin of Ball Lake (Sellers, 2008) and in Tetu Lake (Sellers, 2009) appear to be increasing, although the absolute concentrations are lower than observed upstream.

[4] Based on sediment accumulation rates of 0.3 to 0.4/yr derived from mercury concentration profiles in dated sediment core from Lockhart *et al.* (2000), and an assumed mixing depth of 2 cm.

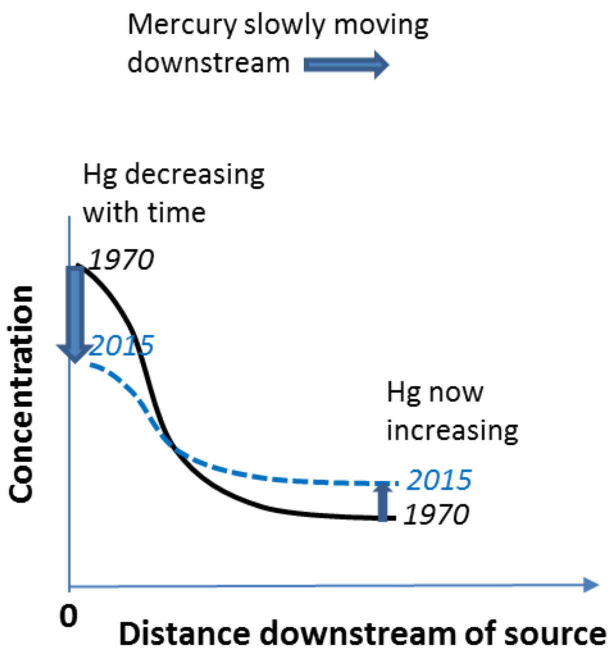

Figure 5. Conceptual sketch of mercury contamination in sediments gradually moving downstream.

A key to remediation of mercury in fish in the Wabigoon River system is to accelerate the decline of inorganic mercury concentrations in surface sediments. Careful consideration needs to be given to the potential for different responses at different locations for any given remediation scheme. Novel approaches are also under development to reduce methylmercury production or uptake in biota without reducing the overall concentration of inorganic mercury in sediments (*e.g.* addition of activated carbon, nitrate additions) but the geographic scale of contamination is a critical consideration for these and any other remediation schemes being considered. These options are discussed in more detail later in this report.

4 Mercury Remediation – What Has Been Done Elsewhere?

Remediation technologies discussed here are broadly divided into two general categories: existing and emerging approaches. Existing approaches have examples of full-scale implementation at mercury-contaminated aquatic sites, while emerging approaches have limited implementation at small or pilot scales. Examples of mercury remediation at contaminated aquatic sites are summarized in Table 1 for existing approaches and Table 2 for emerging approaches. Efforts to reduce mercury releases from chlor-alkali facility sites are not addressed here, but represent another line of pursuit if planned field studies showed that the site of the former chlor-alkali facility continues to release environmentally relevant amounts of mercury.

4.1 Existing Approaches to Reduce Mercury in Sediments

Dredging/excavation

Sediment removal technologies, including hydraulic dredging, mechanical dredging, and dry mechanical excavation have been employed to clean up mercury-contaminated sites. The chief advantage of these approaches is that contamination is removed from the aquatic environment. Disadvantages include relatively high cost, the disruption of ecosystem function, remobilization of contaminated sediments, and disrupt natural recovery processes (Environ, 2013; Randall and Chattopadhyay, 2013). Disposal of contaminated material also poses a potential risk to the surrounding environment if measures are not taken to contain the contamination (Randall and Chattopadhyay, 2013).

Hydraulic dredging typically involves a barge fitted with a boom with a rotating cutter head or a trailing suction pipe, which excavates sediment. Mechanical dredging is often conducted using a barge-mounted crane with a clamshell or dragline bucket (Environ, 2013). Dredged sediment is usually transported to a disposal location, either on land or underwater. Sediment may also undergo treatment to remove contamination, however this alternative is usually costly and therefore less common (Randall and Chattopadhyay, 2013). Dry mechanical excavation often uses water diversion structures to remove water from an area prior to excavation (Environ, 2013). Existing or temporarily dry land such as floodplains or other waterfront land that may leach mercury into the aquatic environment may also be excavated without the need for dewatering. Bank-to-bank dredging is not recommended by the US EPA as a means of mercury remediation (Committee on Sediment Dredging at Superfund Megasites, 2007).

Capping

Capping refers to the addition of material underwater to cover and isolate contaminated sediment from the water column. It also has the potential to reduce surface sediment mercury concentrations by using low-mercury cap material, separate mercury from the biologically active sediment layer, and limit transport of contaminated sediment (Environ, 2013). Contaminated floodplain soils have also been capped at some sites (Turner, 2009a). Caps may be constructed of various materials including low-mercury sediments, sand, gravel, natural/synthetic reactive material and more complex designs involving geotextiles, liners and multiple layers (Randall and Chattopadhyay, 2013). The US Navy has carried out pilot studies in Sinclair Inlet, Washington, involving caps that include activated carbon (Chadwick *et al.*, 2014), discussed in the next section. Experimental testing has shown that caps consisting of sand and finer particles are highly efficient at sorbing mercury from contaminated sediment and can act as an effective barrier to overlying water (Moo Young *et al.*, 2001). In some settings, capped mercury may still migrate through the capping layer and into the water column due to diffusion and natural physical (*e.g.*, currents, sediment consolidation, groundwater flow, tidal activity) and biological processes (*e.g.*, benthic organisms) as well as human activities (*e.g.*, shipping; Randall and

Chattopadhyay, 2013). Methylmercury production rates can increase underneath caps, creating the potential for release of methylmercury if the cap is disturbed (Johnson, 2009). Capping is typically less expensive than dredging, though there is a need for long-term monitoring to ensure cap integrity (Environ, 2013; Randall and Chattopadhyay, 2013).

Advantages of in-situ capping include a lower cost than dredging, suitability to a wide range of contaminants and less long-term environmental impact (Randall and Chattopadhyay, 2013). In-situ capping refers to on-site placement of material over contaminated sediment. Ex-situ capping involves contaminant dredging and transport to an underwater disposal site where it is then capped (Randall and Chattopadhyay, 2013). This process is known as Confined Aquatic Disposal (CAD). A cap may also be applied at a location where dredging removed some but not all contamination. Dredging and capping are often used in combination.

Water filtration

Numerous filtration technologies have been applied to extract mercury from water, including membrane separation, adsorption techniques, polymer filters, microfilters, sand filters and membrane extraction (USEPA, 1997; Ebadian *et al.*, 2001; USEPA, 2007; Santiago *et al*., 2013). Membrane separation involves passing contaminated water through a semi-permeable barrier or membrane that blocks certain constituents (USEPA, 2007). Adsorption techniques rely on compounds, usually packed into a column, which sorb mercury from the liquid phase as contaminated water is passed through the column (USEPA, 2007). Mercury extraction from solid debris can be conducted via a polymer filtration technique developed by the Los Alamos National Laboratory (Turner, 2009a). The technique employs a leaching solution and multiple ultra-filtration applications. Contaminated water can also be passed through sand filters to extract mercury, using either pumps or holding ponds in which gravity draws water through the sand filter (Santiago *et al*., 2013; AECOM, 2012). Multiple filtration techniques are often used in conjunction to maximize the efficacy of mercury extraction. Little information was available on the relative costs of various filtration technologies.

Erosion control

Various measures can be applied to limit the erosion of contaminated sediments, which can be a source of mercury to aquatic systems. Erosion control measures have been successfully employed at mercury-contaminated sites, though limited information was found describing the particular techniques used. Erosion controls at a mine near Walker Creek, California led to a 90% reduction in mercury loading from the site (Turner, 2009a). Sediment, waterfront and/or floodplain material could be stabilized using physical (*e.g.*, riprap/rock lining, grading, compaction), biological (*e.g.*, vegetative buffer strip, phytostabilization) or chemical (*e.g.*, surface treatments) approaches. Pilot scale bank stabilization studies were carried out in the South River, Virginia, where mercury was released from 1929-1950 from a DuPont facility that produced Rayon (Stahl, 2013). Plans are under development to initially stabilize areas within a 3 km stretch of river near the facility that released mercury.

Monitored Natural Recovery

Monitored Natural Recovery (MNR) relies on natural recovery processes to reduce contaminant levels, combined with a monitoring program over time. For mercury in aquatic systems, the primary natural removal mechanisms are burial, downstream export and evasion to the atmosphere. Downstream export eliminates mercury from one location, but transfers that mercury downstream. MNR was selected as a preferred option to address contaminated sediments in the St. Lawrence River near Cornwall, Ontario (deBarros and Anderson, 2010). This choice was partly based on findings that "Since contaminated sediments along the Cornwall waterfront are not toxic to sediment dwelling organisms and fish, since the sediments are stable and there is no risk to people or the environment, natural recovery was considered a suitable option for the three zones". Furthermore: "Since mercury in sediment is not a major contributor to mercury in fish, to dredge or cap the sediments would not result in a measurable benefit to the fish. Thus the Natural Recovery option is the most suitable option for dealing with the contaminated sediments within the St. Lawrence (Cornwall) AOC." These statements suggest that level of contamination, influence of sediments on fish Hg, and the associated concern was different for the Cornwall case and the Wabigoon-English River system.

MNR is also used in combination with other remediation techniques. Mercury remediation in Onondaga Lake, NY, for example used a combination of methods including dredging, capping, nitrate additions and MNR (US EPA, 21015c). MNR was also a component of the overall remediation strategy employed at the Wyckoff/Eagle Harbor Superfund site in Washington State (Merritt et al., 2009). MNR has the advantages of being the least disruptive option for an ecosystem and has the lowest cost. The disadvantage of MNR is that it takes longer than active remediation, based on the rate that natural processes can eliminate contamination.

We do not consider MNR to be a viable standalone option for the Wabigoon-English System because fish mercury concentrations in Clay Lake are presently not declining or are declining at an imperceptible rate.

Table 1. Existing approaches to actively remediate mercury contamination in aquatic systems. These technologies have full-scale implementation at mercury contaminated aquatic sites.

Option	Description	Site applications	Reference
Dredging	Hydraulic dredging: Sediments are pumped to a barge, then a disposal location on land or underwater. Mechanical dredging: Excavation with clamshell or bucket.	St. Clair River, Ontario, Canada Acid Brook, NJ Onondaga Lake, NY	Environ (2013); Turner (2009a) Turner (2009a) US EPA (2015c)
Dry mechanical excavation	Dewatering, mechanical excavation and disposal. Also, removal of contaminated soil in floodplain or adjacent waterbody.	North Fork Holston River, VA Poplar Creek, TN Acid Brook, NJ Squamish, BC Carson River, NV Guilderland and Colonie, New York East Fork Poplar Creek, Oak Ridge, TN Sydney, Austria (Orica Botany)	Turner (2009a) Turner (2009a) Turner (2009a) Turner (2009b) US EPA (2015a) US EPA (2015b) Lora *et al.* (2011) NSW EPA (2014)
Capping	Addition of low mercury solids on top of contaminated sediments. May also include biologically or chemically reactive layers, geotextiles or liners.	Peninsula Harbor, Lake Superior Elliott Bay, Seattle, WA Whatcom Waterway, Bellingham, WA Middle Waterway Commencement Bay, Tacoma, WA Onondaga Lake, NY Lake Turingen, Sweden	Wilson *et al*. (2014) Reible (2005) Reible (2005) Reible (2005) US EPA (2015c) Reible (2005)

Option	**Description**	**Site applications**	**Reference**
Dredging and capping	Combination of dredging followed by capping.	Minimata Bay, Japan Elliott Bay, Seattle, WA West Eagle Harbor/Wyckoff Bainbridge Island, WA Middle Waterway Commencement Bay, Tacoma, WA Duwamish River/Elliott Bay Seattle, WA Puget Sound Naval Shipyard Bremerton, Seattle, WA ALCOA Lavaca Bay, Point Comfort, TX North Fork Holston River, VA Geddes Brook/Ninemile Creek, NY	Hosokawa (1994) Reible (2005) Reible (2005) Reible (2005) Reible (2005) Reible (2005) Reible (2005) Turner (2009a) Turner (2009a)
Water filtration	Mercury is removed from water passing through a filtration system.	North Fork Holston River, VA Abbotts Creek, NC East Fork Poplar Creek, Oak Ridge, TN Hamilton Harbor, Ontario, Canada St. Clair River, Ontario, Canada	Turner (2009a) Turner (2009a) Lora *et al.* (2011) AECOM (2012) Santiago *et al.* (2013)
Erosion control	Stabilization of contaminated sediment, waterfront or floodplain material.	South River, VA Cache Creek, CA Walker Creek, CA Guadalupe River, CA East Fork Poplar Creek, Oak Ridge, TN	Stahl (2013) Turner (2009a) Turner (2009a) Turner (2009a) Lora *et al.* (2011)
Monitored Natural Recovery	Reliance on natural processes to reduce contamination, in combination with long term monitoring.	Onondaga Lake, NY (in combination with other options). Wyckoff/Eagle Harbor, Washington State (in combination with other options) Cornwall, ON Area of Concern	US EPA (2015c) Merritt et al. (2009) deBarros and Anderson (2010)

4.2 Emerging Approaches to Reduce Mercury in Aquatic Systems

Enhanced Natural Recovery (ENR)

Enhanced natural recovery of mercury contamination in surface sediments has been proposed as potential technique to reduce mercury methylation and bioaccumulation (Rudd et al 2013a). This approach involves the addition of low-mercury solids (such as clean sediment) to the water column. This would increase the natural sedimentation rate and reduce mercury concentrations mercury in surface sediments. Production of methylmercury would be reduced in sediments and potentially in overlying waters. The added solids may also bind mercury in the water column, and lower mercury concentrations that are relevant to bioaccumulation.

Rudd and Turner (1983a) conducted pilot scale experiments in large enclosures located in Clay Lake. Addition and resuspension of low organic content sediments were observed to significantly reduce mercury bioaccumulation by zooplankton, crayfish, clams and pearl dace. The authors suggested that continuous or periodic resuspension of low-mercury sediments by dredges followed by downstream deposition and dilution of mercury in contaminated surface sediment might be a feasible mercury remediation strategy for the Wabigoon-English River system. This procedure is now also under consideration for the remediating the mercury pollution of the Penobscot River/Estuary (Rudd et al., 2013a,b).

Activated carbon

Techniques involving activated carbon are commonly applied to remove mercury from industrial waste (Ebadian *et al.*, 2001; US EPA, 1997). Many laboratory studies have also investigated activated carbon technologies for aqueous mercury removal (*e.g.*, Anirudhan *et al*., 2008; Zabihi *et al*., 2010; Di Natale *et al.*, 2011). Mercury contaminated water can be treated by passing through a system of columns containing activated carbon, usually in granular form (USEPA, 1997; Ebadian *et al.*, 2001). This technique was applied to the outflow of a mercury contaminated waste pond from an historic chlor-alkali facility on the banks of the North Fork Holston River, VA resulting in a 98% reduction in mercury loading to the river (Turner, 2009a).

Activated carbon can also be applied in-situ to contaminated sediments, either alone or as a component of capping material. Microcosm studies investigating in-situ activated carbon treatments at freshwater and estuarine sites found that the technique significantly reduced dissolved methylmercury concentrations in sediment pore water and bioaccumulation of methylmercury by a species of aquatic worm (Gilmour *et al.*, 2013a). Gilmour *et al.* (2013b) also conducted microcosm studies in a marsh in the Penobscot River estuary with activated carbon, biochar, iron, and lime treatments. Activated carbon amendments reduced porewater methylmercury concentrations by an average of 60-70% and total mercury concentrations by 50-60%, with biochar being nearly as effective (50-70%, 35-55%). A full-scale demonstration of a proprietary activated carbon aggregate was conducted at the Puget Sound Naval Shipyard Bremerton, WA. The cap was approximately 5-15 cm thick. Preliminary results showed a significant decrease in total mercury concentrations in sediments, but little short-term effect on methylmercury in sediments and biota (Chadwick *et al.*, 2014). The need for additional applications if efficacy decreases with time remains to be assessed. Multi-walled carbon nanotubes are another emerging technology that has shown the ability to remove mercury from solutions in a recent study (Yaghmaeian *et al.*, 2015).

Phytoremediation

Phytoremediation is a general term that refers to various approaches involving the use of plants to remediate contaminated sites. Advantages of phytoremediation include its relatively low cost and minimal intrusiveness compared to conventional remediation approaches (Jadia and Fulekar, 2008; Henry, 2000; Fitzgerald, 2014). Phytoextraction refers to the use of mercury-accumulating plants to absorb, concentrate and store mercury in biomass. Mercury enriched plant material can be harvested and treated as hazardous waste, removing mercury from the site (Fitzgerald, 2014). Phytovolatilization is another approach involving plants which absorb aqueous mercury species, transform them into volatile forms and transpiring them into the atmosphere (Jadia and Fulekar, 2008). A disadvantage of phytovolatilization is that the atmospheric mercury will find its way back into terrestrial or aquatic environments via dry or wet deposition. A third approach, phytostabilization, is the use of plant roots to minimize the mobility of mercury in sediment. The plants may act to limit water percolating through the sediment, act as a barrier limiting contact with contaminated sediment, and/or minimize erosion (Jadia and Fulekar, 2008). A primary disadvantage of this method is that contamination is not removed from the site.

Coagulation

Coagulation is the aggregation of material in the water column via the formation of particles (flocculate) that precipitate out of solution. Coagulation has been applied in the treatment of drinking water, wastewater and storm water, as well as whole-lake phosphorous removal (Kraus, 2013). Recent laboratory and field studies have shown that coagulation techniques can effectively remove both inorganic mercury and methylmercury from solution in contaminated natural waters (Henneberry *et al.* 2015, 2011; Kraus *et al.*, 2013). Coagulants that can be used to remove mercury include metal-based salts, in particular iron and aluminum compounds (Henneberry *et al.* 2015; Ebadian *et al.*, 2001; USEPA, 1997). Removal efficiency is dependent on the coagulant concentration or dosage and the initial mercury concentration and species (Ebadian *et al.*, 2001). Laboratory studies by Henneberry *et al.* (2015, 2011) have shown that treatment with metal-based salt coagulants can remove greater than 75% of filtered inorganic mercury from water samples. Field studies by the same group (Kraus *et al.*, 2013) demonstrated that coagulation treatments can be scaled up to an in-situ wetland application with similar efficacy (60-85% reduction in filtered total mercury and methylmercury).

Nitrate addition

Nitrate addition has been proposed as a viable remediation approach to reduce methylmercury production in certain mercury contaminated aquatic systems (Matthews *et al.*, 2013). Recent studies suggest that nitrate addition can suppress methylation by promoting metabolic pathways that are more energetically favoured than sulfate reduction (Todorova *et al.*, 2009) or iron reduction, processes identified with methylmercury production. This phenomenon has been studied in Onondaga Lake, a seasonally stratified, eutrophic, sulfate-rich lake in New York with mercury contamination from two historic chlor-alkali facilities (US EPA, 2015c, Todorova *et al.*, 2009). Todorova *et al.* (2009) observed that increasing long-term nitrate supply to the lake from a wastewater treatment correlated with marked decreases in methylmercury accumulation in the hypolimnion. In a subsequent study, Matthews *et al.* (2013) conducted a whole-lake nitrate addition pilot test in Onondaga Lake. A liquid calcium-nitrate solution was added to the hypolimnion three times weekly throughout the period of summer stratification. Hypolimnetic methylmercury concentrations decreased by 94% compared to the baseline, which eliminated the usual spike in methylmercury concentrations in surface waters following fall turnover.

Selenium addition

Aquatic biota have been shown to rapidly bioaccumulate selenium with an accompanying decrease in mercury bioaccumulation (Turner and Rudd, 1983). Therefore, the addition of selenium to mercury contaminated aquatic ecosystems has been proposed as a remediation strategy to reduce methylmercury concentrations in biota. This concept is supported by pilot studies in Clay Lake (Turner and Rudd, 1983; Rudd *et al.*, 1980), which found the addition of selenium led to a decline in mercury concentrations in all fish species studied. Selenium's antagonistic effect on mercury bioaccumulation in aquatic organisms is well-established with several studies observing an inverse relationship between mercury and selenium concentrations in aquatic biota (Chen *et al*., 2001; Belzile *et al.*, 2005; Khan and Wang, 2009). A disadvantage of this approach is the potential toxicity of selenium to aquatic organisms (Turner and Rudd, 1983).

Aeration

Mercury methylation occurs primarily under anaerobic conditions. Aerating anoxic bottom water and sediments that are hotspots for mercury methylation can suppress methylmercury production. The feasibility of this approach has been demonstrated in pilot-scale studies (Mailman *et al.*, 2006; Wang *et al.*, 2004) and at least one application at a mercury contaminated site. A small (25 acre) reservoir on the Guadalupe River, California with contamination from historic mercury mining operations was aerated, leading to a significant (>95%) reduction in the methylmercury concentrations (Turner, 2009a). Hypolimnetic oxygenation can carry number other water quality benefits besides remediation of mercury contaminated sites (Beutel and Horne, 1999). Deep oxygen injection systems have been in use since the early 1980s, with applications in several Swiss lakes to remediate eutrophication, as well as other applications in hydroelectric reservoirs in the southern United States (Beutel and Horne, 1999). One potential drawback to this approach is negative effects on organisms living in sediments (Fitzgerald, 2014).

Increase load of limiting nutrients

Increasing the load of limiting nutrients to stimulate primary production has been investigated as a potential mercury ameliorating procedure. Rudd and Turner (1983b) carried out a microcosm experiment in which enclosures were enriched with nutrients ($NaN0_3$ and NaH_2P0_4) at two different levels. The highly enriched enclosure showed increases in primary productivity, fish growth rates and pH, and a reduction in fish mercury concentrations. The moderately nutrient-enriched enclosure also showed increases in primary productivity and fish growth rates, but no change in pH and an increase in fish mercury concentrations. Based on these mixed results, the authors did not recommend this approach as a mercury remediation procedure.

Other approaches and considerations

Other emerging approaches were identified as potential remediation options for mercury contaminated sites, including reducing the load organic matter, various measures to enhance demethylation and trophic modifications to limit mercury bioaccumulation. Little to no discussion of these approaches was found in the literature. They were deemed to be at a conceptual stage and likely do not merit consideration for the Wabigoon-English River System until further research and testing have been conducted.

Upstream activities, such as logging and nutrient loading from effluent discharge, have the potential to affect water chemistry such that methylation rates are also affected. The potential for these to be of significant influence in parts of the Wabigoon-English River system warrants assessment in the future.

Table 2. Emerging approaches to actively remediate mercury contamination in aquatic systems. These technologies have limited or pilot-scale implementation at mercury contaminated aquatic sites, or have been investigated in laboratory studies.

Option	Description	Site applications	Reference
Activated carbon	Activated carbon is added to the sediment surface to bind mercury and reduce bioavailable methylmercury.	North Fork Holston River, VA South River, VA (pilot study) Puget Sound Naval Shipyard Bremerton, Seattle, WA Penobscot River estuary, ME (microcosm study) Freshwater and estuarine microcosm study Many laboratory studies	Turner (2009a) Patmont (2015) Chadwick *et al.* (2014) Gilmour *et al.* (2013a) Gilmour *et al.* (2013b) See discussion
Phytoremediation	Use of plants to absorb and retain mercury (phytoextraction), volatilize mercury (phytovolatilization) or stabilize contaminated sediment (phytostabilization).	Idaho Falls, ID	USEPA (2002)
Coagulation	Removal of mercury from water via coagulation (formation of solids via chemical additions)	Twitchell Island, CA (field study) Laboratory studies	Kraus *et al.* (2013) Henneberry *et al.* (2015, 2011)
Nitrate addition	Addition of nitrate to suppress activity of mercury methylating microbes.	Onondaga Lake, NY	US EPA (2015c) Matthews *et al.* (2013)
Selenium addition	Organism bioaccumulation of selenium and a reduction in mercury bioaccumulation.	Clay Lake, Ontario (pilot studies)	Turner and Rudd (1983) Rudd *et al.* (1980)
Aeration	Aeration to suppress methylation.	Guadalupe River, CA	Turner (2009a)
Dilution of mercury in surface sediments (ENR)	Added clean material is naturally deposited and mixed into surface sediments thus reducing mercury and methyl mercury concentrations	Clay Lake, Ontario (pilot studies)) Penobscot Estuary, under consideration	Rudd and Turner (1983a) Rudd *et al*. 2013a,b
Increase load of limiting nutrients	Stimulation of primary production and fish growth rates	Clay Lake, Ontario (Pilot study)	Rudd and Turner (1983b)

5 Remediating the Wabigoon-English River System

The previous section summarized remediation approaches that have been used in a variety of other settings. In this section we address approaches that are best suited to the Wabigoon-English River System.

Based on available scientific evidence, it is likely that recovery of mercury contamination could be accelerated in at least some parts of the Wabigoon-English River system. More is known now about aquatic mercury pollution than in the early 1980's when the last in-depth studies of the Wabigoon-English River System were done. As stated earlier, the basic problem in the Wabigoon-English River System is the persistence of elevated fish mercury concentrations. This is caused by the persistence of inorganic mercury contamination in surface sediments, which stimulates the production of methylmercury that is found in fish (Chapter 3). If the reasons for this persistence could be identified, and surface sediment mercury concentrations in the system could be lowered to near background concentrations of about 100 – 150 ng/g dry weight (Sellers, 2005), the problem would be solved. For perspective, surface sediment concentrations in Clay Lake were about 700-2000 ng/g when last sampled in 2004.

In addition to recommendations for specific remediation methods, we also recommend that any remediation plan use an adaptive management approach. Adaptive management is an iterative process of decision making in the face of the uncertainty associated with complex systems. This approach improves the likelihood of success by monitoring the ecosystem as remediation proceeds, so that adjustments can be made if necessary to the remediation plan as work proceeds. Adaptive Management approaches needs to be accompanied by an appropriate funding framework *i.e.* one that allows remediation goals to be reached without interruption or delays.

5.1 Basis for Determining the Best Approach to Remediation

Before any remediation plan can be designed, it is necessary to know the geographic extent of the present-day contamination and the reason why recovery of fish mercury concentrations has stalled 45 years after controls were imposed at the Dryden facility. It is critical to understand these two issues so future remediation actions can be identified that are feasible and based on sound science.

5.1.1 Geographic Scale of Contamination[5]

The widespread downstream dispersal of mercury since it was added to the system between 1962-1970, limits options that are available for remediation. Widespread dispersal is often one

[5] We define contamination in this study to be mercury concentrations that are higher *today* than they would be if the chlor-alkali plant had not released mercury to the Wabigoon River system.

of the most difficult aspects of mercury pollution, as seen before at several other sites with large point-source releases of mercury (*e.g.* Penobscot River estuary ME, Lavaca Bay TX, Sudbury River MA, South River, VA). The first step in understanding the present-day severity of contamination is to establish current concentrations of mercury in sediments and fish throughout the system. The second step is to compare these concentrations to benchmark concentrations that define contamination. We define benchmarks in this report as concentrations that would exist if there had been no mercury release from the former chlor-alkali facility.

<u>Extent of Contamination Based on Mercury in Fish</u>

Mercury concentrations in adult sportfish (walleye and northern pike) were selected to examine mercury contamination in fish, as they represent the higher trophic level fish species with the highest concentrations, and are popular subsistence and sportfish.

We do not know what fish mercury levels were prior to the presence of the chlor-alkali facility at Dryden. Current fish concentrations in the Wabigoon River system can be compared to concentrations in other lakes in areas of northwestern Ontario where there are no documented point sources of mercury contamination. However, Clay Lake and lakes downstream are atypical of Canadian Shield Lakes in the region because they are richer in glacial clays and silts. Clays bind mercury very tightly and so, in absence of pollution, fish in Clay Lake are expected to have lower fish Hg levels than lakes without clay (Rudd and Turner, 1983). Thus, while northwest Ontario northern pike (50 cm) and walleye (45 cm) average 0.4 - 0.5 ppm mercury respectively (Neff et al., 2012), we think that the targets for fish in Clay Lake should be lower.

In the absence of fish mercury data for Clay Lake without the influence of the former chlor-alkali facility, it is necessary to predict what the fish mercury concentration was before pollution. The best available way to do this is to use existing data from Clay Lake, namely the past relationship between mercury in fish and sediments in Clay Lake (Figure 4). If the sediments were lowered to their pre-pollution mercury levels (100 - 150 ng/g), which is about 10 times lower than they are today, the walleye mercury would also be lowered by about 10 times[6] to 0.2 ppm. As such, 0.2 ppm in 45 cm walleye is a reasonable estimate of a target in remediation efforts for Clay Lake, based on available data. Recognizing uncertainty in this estimate, available data can be interpreted to suggest that if the effects of contamination from the chlor-alkali facility could be fully remediated, fish mercury concentrations would be expected to be lower than regional average values.

There may be extenuating factors that we are currently unaware of that could prevent the achievement of this target and place it closer to regional average concentrations. Such factors will be revealed when we collect more data during the field program.

[6] It should be also noted that proportional responses do often occur (e.g. Penobscot) but there are examples of non-linear responses in highly contaminated systems (*e.g.* South River, VA).

Insufficient data are available to predict benchmarks for mercury in fish in lakes downstream of Clay Lake. In the absence of mercury from the chlor-alkali facility, 45 cm walleye in these lakes could have benchmark concentrations similar to the value predicted to Clay Lake (0.2 ppm), or similar to the regional background (0.5 ppm).

Mercury concentrations in Clay Lake walleye (45 cm) and northern pike (50 cm) were 2.4 and 1.5 ppm in 2014 (Table 3). These **concentrations were clearly above any reasonable estimate of benchmark concentrations and are outside the range for other water bodies reported by Neff *et al.* (2012).** Farther downstream, in Ball Lake, Separation Lake, and Tetu Lake, walleye mercury concentrations (45 cm) were not as high as Clay Lake and were within the background range, but were above the regional background average of 0.5 ppm and the 0.2 ppm benchmark for Clay Lake. Mercury concentrations in 50 cm northern pike in 2014 are also shown in Table 3. Mercury concentrations were higher in the south basin of Ball Lake in 2014 for both walleye and northern pike.

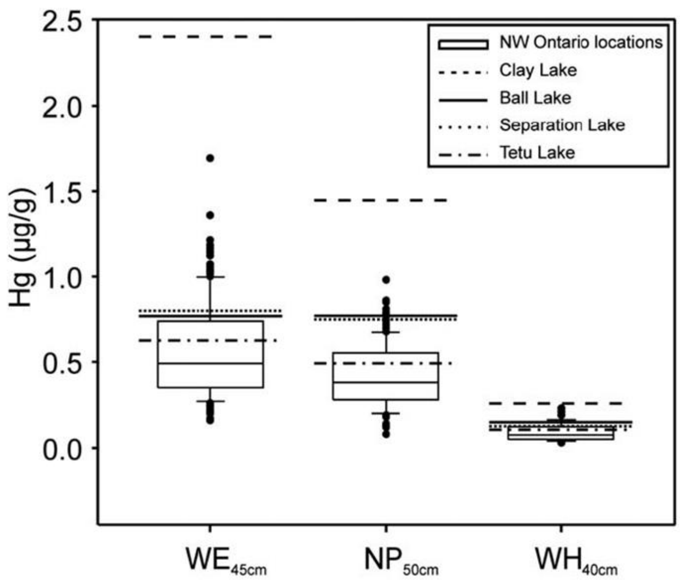

Figure 6. Box plots of recent (2000–2010) mercury levels in in the Wabigoon-English River system and other Northwestern Ontario locations. Figure is from Neff *et al.* (2012). Northwestern locations (north of 48° N and west of 85° W) are compared to lakes in the Wabigoon-English system at standardized mercury concentrations for (a) 45 cm walleye (n = 143 locations), (b) 50 cm northern pike (n = 123 locations) and (c) 40 cm lake whitefish (n = 38 locations). Lines in each box represent the median concentration, boxes indicate the 25th and 75th quartile values, and whiskers indicate the upper and lower values not classified as statistical outliers or extremes. Horizontal lines indicated mean mercury concentrations for Clay, Ball, Separation and Tetu Lakes for 2000–2010, for each respective fish length.

Table 3. Estimated mercury concentrations in four lakes in 2014. Derived from data provided by MOECC. Standard length Hg concentrations estimated using power series fits to data.

Lake	**Mercury concentration (µg/g)**	
	Walleye Hg (45 cm)	**Northern Pike Hg (50 cm)**
Clay Lake	2.41	1.51
Ball Lake (north basin)	0.57	0.28
Ball Lake (south basin	0.92	1.05
Separation Lake	0.88	0.74
Tetu Lake	0.56	0.34

Routine monitoring of fish mercury levels is carried out in 4 lakes in the Wabigoon-English River system (Figure 6, Table 3). A survey that included more lakes has been done, but it was 12 years ago, and the data were not standardized to fish length for comparison from lake to lake (Kinghorn et al 2007; data from 2003). Thus, a one-time survey of present-day concentrations of mercury in fish is recommended in 2017. This survey should include all lakes on the mainstem of the river as well as lakes used by First Nations for subsistence fishing.

Extent of Contamination Based on Mercury in Sediments

Sediment core data from Clay Lake provide information on what mercury concentrations were prior to the industrial period, on the order of 50 ng Hg/g (Sellers, 2005). Due to increased mercury releases regionally and globally, sediment mercury levels today are often 2-3x higher than pre-industrial levels. As a result, the benchmark for what the sediments would be if the chlor-alkali facility had not contaminated the system is estimated to be 100-150 ng/g.

Mercury concentrations in surface sediments in Clay Lake and in the Wabigoon River between Dryden and Clay Lake are still clearly contaminated. Recent surface sediment concentrations in Shallow Lake, which is a small lake between Dryden and Clay Lake, are 2200 ng/g (*i.e.* ~10-20 times above background). Concentrations in the east basin of Clay Lake are also about 2000 ng/g, and the most recent (2004) data show little decrease in the preceding 20-30 years. Downstream of Clay Lake, surface sediment concentrations are lower, and were in the range of 300-400 ng/g in the 1980s (Parks and Hamilton, 1987). More recently, Sellers (2005) reported concentrations in the south basin of Ball Lake of approximately 320 ng/g. The sharp decline in sediment mercury levels downstream of Clay Lake was likely because Clay Lake is an effective trap for particles, and mercury can bind strongly to solids. While sediment mercury concentrations downstream of Clay Lake are within the high end of the natural range of mercury concentrations observed in freshwater systems, they are above the 100-150 ng/g estimate of current background for this system and likely reflect contamination.

A one-time survey of surface sediment mercury concentrations is recommended in conjunction with fish sampling. We also recommend that as soon as possible sediment and fish sampling be done in the adjoining lakes that are most frequently used or desired to be used by First Nations people for subsistence fishing. When these data are compared to benchmark values, decisions can be made about which sites are of highest priority for remediation. A study plan for assessing the geographic extent of contamination is outlined in Chapter 7.

Extent of Contamination Based on Mercury in Water[7]

Mercury concentrations in surface waters sampled from 1978-1980 were reported as high as 26.5 ng/L upstream of Clay Lake, and were lower downstream of Clay Lake (Parks and Hamilton, 1987). Methylmercury concentrations in surface waters reported by Parks and Hamilton (1987) were in the range of 1-1.5 ng/L from Dryden to the inflow to Ball Lake. No recent data are available. The concentrations reported in the 1980's are roughly 5-10X above background levels of roughly 2 ng/L for total mercury and 0.1 ng/L for methylmercury in freshwaters, based on a survey of US rivers (Scudder *et al.*, 2009). It is worth noting that the Health Canada guideline for mercury concentration in drinking water is 1000 ng/L (Health Canada, 2014), and available data indicate that concentrations are expected to be far below levels of concern for drinking water. Federal guidelines for the protection of aquatic life are more restrictive: 26 ng/L for inorganic mercury and 4 ng/L for methylmercury (CCME, 2003). While concentrations of mercury and methylmercury are likely lower in surface waters now than in the 1970s and 1980s, this should be confirmed with new measurements (Chapter 7, Task D).

Overall, considering available data for mercury levels in fish and sediments, concentrations remain contaminated from the former chlor-alkali site downstream to at least Clay Lake and likely farther downstream to at least Ball Lake.

5.1.2 Has Recovery Stalled Because of Ongoing Sources of Mercury Between Dryden and Clay Lake?

Figure 4 shows that since about 1985 the concentrations of mercury in 45 cm walleye taken from Clay Lake appear to have plateaued (i.e. stopped declining) or are declining at an imperceptible rate. This suggested to us that there is likely an ongoing source of mercury to Clay Lake originating either from the former chlor-alkali facility itself or from the contaminated river sediments above Clay Lake – or both. To demonstrate this we modelled expected surface sediment mercury concentrations in Clay Lake by assuming that suspended sediments entering

[7] Dr. Rudd was directly involved in the English-Wabigoon water sampling for mercury concentrations during the 1980's. Samples were analyzed in the metals lab at the Freshwater Institute in Winnipeg under the direction of Drs. A. Lutz and F.A.J. Armstrong. This analytical laboratory was the gold standard lab of its time. Low detection levels were achieved by taking large volume (20 L) samples and concentrating them in the lab. Decades later it is Dr. Rudd's opinion that these analyses are reliable and comparable to results produced by modern methods.

Clay Lake from the Wabigoon River upstream of Clay Lake had been at near background concentrations (100 or 200 ng/g dw) since 1970. These modelled results are compared to actual surface sediment mercury concentrations in cores taken from Clay Lake (Figure 7). The modeled data show that mercury concentrations in the surface sediments should have recovered to near background concentrations by about year 2000. Instead measured concentrations were about 10 fold higher that we expected (2000 ng/g dw). Because of the close correspondence of surface sediment mercury concentrations to fish mercury concentrations (Figure 4) we expect that fish mercury concentrations should also have recovered in Clay Lake by about year 2000, if there was no ongoing source(s) to Clay Lake.

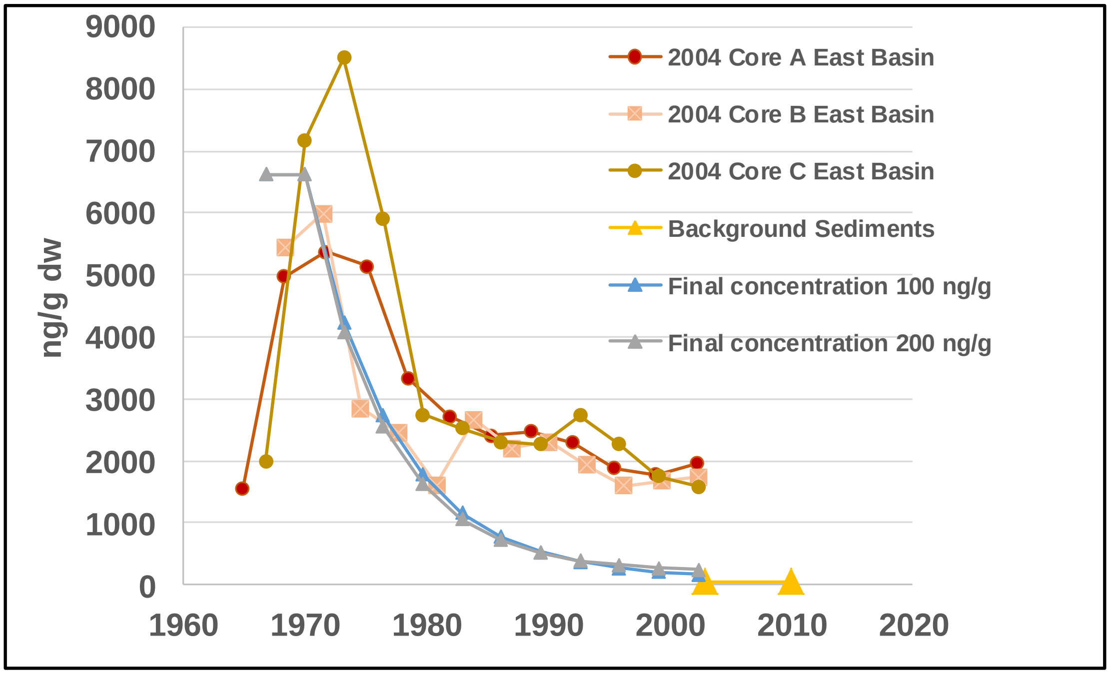

Figure 7. THg concentrations in Cores A, B, and C (circles and squares) taken from the East Basin of Clay Lake (P. Sellers, unpublished data), and THg concentrations predicted (triangles) in sediments if clean sediments (100-200 ng/g) were accumulating in Clay Lake since the peak contamination in 1970. Surface mixing depth was estimated to be 2 cm

The two potential present-day sources of elevated/legacy mercury to the Wabigoon River above Clay Lake are discussed below.

Present-day releases of legacy mercury from the former chlor-alkali site in Dryden

If there are still environmentally meaningful releases of legacy[8] mercury into the river from the site of the former chlor-alkali facility in Dryden, efforts to reduce mercury concentrations in sediments would only be temporary solutions. Eventually ongoing residual loads from the facility site would re-contaminate the system to some degree.

Fifty-seven years after mercury releases started, mercury concentrations in surface sediments of Shallow Lake and Clay Lake are still highly elevated (2200 ng/g and 2000 ng/g respectively). These data could be explained by an ongoing elevated source of mercury from the plant site to the river and/or by a slow recovery of legacy mercury contamination in sediments in the upstream river (Figure 7). We propose to investigate both of these possibilities with additional work.

This stalling of recovery in Clay Lake is likely also affecting recovery rates in downstream lakes because water and contaminated particles are constantly flowing out of Clay Lake towards the downstream lakes. This concern is supported by data in (Neff *et al*., 2012), which shows that walleye and northern pike mercury levels at standard lengths in Ball Lake and Separation Lake have stabilized above 0.5 ppm.

Although controls on mercury releases were initiated at the Dryden facility in 1970, measurements of plant effluent in the 1970's and 1980's showed that mercury losses to the river were still occurring (Wilkins, 1978; Parks and Hamilton 1987). Continued losses of mercury from chlor-alkali facilities, where controls are already in place, are frequent (for example, HoltraChem-Penobscot River MA; ALCOA-Lavaca Bay TX; Squamish BC). At some locations, further controls have been instituted more recently, further lowering ongoing inputs. For example, at the HoltraChem site high-level treatment of the plant effluent is now in place, and ground water pumping is ongoing to capture mercury contaminated ground water before it can seep into the Penobscot River. As a result, estimated losses of mercury from the HoltraChem site to the river have now been lowered to about 2 kg per year, which was taken to be an acceptable level of input for the Penobscot system (Rudd *et al*., 2013). As noted in Chapter 3, even 1% of the peak mercury release rate from the facility site at Dryden could affect mercury levels in the Wabigoon-English River system.

We recommend that a full assessment be carried out of present-day losses of mercury from the former chlor-alkali facility site in Dryden to the Wabigoon River. While mercury monitoring is presently carried out associated with a disposal site near the former chlor-alkali facility, there could also be ongoing releases to the river from ground water locations along the shoreline of the property at the site of the former chlor-alkali facility. A description of field work that will investigate these sources to the river is provided in Chapter 7, Task F.

[8] We define the term "legacy mercury" as mercury that was used in the chlor-alkali facility until controls were instituted in 1970. Some remnants of the legacy mercury may still be entering the river from the facility site, or legacy mercury previously deposited in the river sediments or river banks may still be moving down to Clay Lake, especially at times of high river flow.

Contamination from river sediments and banks between Dryden and Clay Lake

Parks and Hamilton (1987) estimated that the mass of mercury between Dryden and the inflow to Clay Lake was between 2.3 and 4.9 tonnes around 1980. It is not known how much of this mercury still remains in the Wabigoon River upstream of Clay Lake, or how much moves downstream to Clay Lake each year. It is also not known how much of the mercury coming into Clay Lake is methylmercury that was produced in the river between Dryden and Clay Lake. There is concern that some of this previously deposited mercury is still being transported downstream as inorganic mercury or methylmercury, especially during high flow events when erosion and resuspension of contaminated sediments (and of river bank material) would be greatest.

Field studies are needed to assess the importance of the river between Dryden and Clay Lake as a source of methylmercury and/or inorganic mercury downstream. We recommend that these studies be carried out over a two year period[9]. Measurements of mercury in water and in sediments along this reach are needed to better identify key sediment areas needing remediation. Methylmercury and inorganic mercury data are both needed to help identify the extent to which methylmercury is produced locally and exported downstream, versus the downstream transport of inorganic mercury to sites where methylation happens (*e.g.* in Clay Lake). Hydrometric data for the Wabigoon River at Quibel are available through Environment Canada; such data can be used to estimate mass transport of mercury along the River.

Bank erosion is another potential source of mercury along the river, and an aerial survey is proposed to identify areas, if any, where significant bank erosion is occurring. Better information on sediment and river bank "hotspots" has the potential to influence the feasibility and cost of different remediation approaches related to these sources of mercury. Additional information on proposed studies in the Wabigoon River between Dryden and Clay Lake are outlined in Chapter 7.

5.2 Discussion of Selected Remediation Options:

We recommend that the remediation of the Wabigoon-English River System should focus initially on the portion of the system between Dryden and the downstream end of Clay Lake. A completely successful clean-up of the Wabigoon River System will likely require that more than one method be applied in order to reduce fish mercury concentrations to desired benchmark levels. We recognize that achieving this goal is subject to uncertainties in our understanding of the system, but we do believe the present situation could be substantially improved. Achieving a mercury concentration of 0.2 ppm in Clay Lake for example would enable women of child

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[9] Two years of measurements of mercury in Clay Lake inflow is a minimum because flow during any one year may be either unusually high or low, and therefore could skew longer term estimates.

bearing age and children under age 15 to eat 8 meals[10] of fish per month without being at risk to mercury exposure (Government of Ontario, 2015).

Three geographic areas are identified as initial candidates for further evaluation of remediation options:

1. The former chlor-alkali facility site.
2. The Wabigoon River between Dryden and Clay Lake.
3. Clay Lake.

Remediation of each of these areas is discussed below. Remediation of these three areas could also benefit downstream sites (such as Ball Lake), but additional data are needed to better evaluate the likelihood of this occurring, and monitoring will be important in assessing possible downstream benefits.

5.2.1 Reduce Present-day Mercury Discharges from the Former Chlor-alkali Site

Remediation of the Wabigoon-English River mercury problem would be simpler if it was found that the former chlor-alkali site in Dryden is important and the dominant present-day source of mercury to the river system. This is because the task of reducing downstream transport of legacy mercury from sediments or bank materials would be more difficult than stopping possible present-day inputs of mercury from the facility site.

It would be comparatively easy and cost-effective to treat a confined point source at Dryden. This could be done by the removal of mercury from end-of-pipe effluents and/or by pumping and treating of contaminated groundwater before it can seep into the Wabigoon River, as was done for the HoltraChem site on the Penobscot River. Another example of successful treatment of a chlor-alkali facility site is at Lavaca Bay, TX, (Bloom *et al.* 1999; Gill *et al.* 1999). At this site, fish mercury concentrations were lowered by preventing contaminated groundwater at the facility site from seeping into Lavaca Bay by the pumping of groundwater to reverse ground water flow.

5.2.2 Reduce Surface Sediment Mercury concentrations Between Dryden and the Inflow to Clay Lake

If proposed field studies show that the stretch of the Wabigoon River between Dryden and Clay Lake is an important source of inorganic mercury or methylmercury to the water column and downstream sites, remediation of this source of mercury could be attempted by locating depositional sites and treating them with targeted "hot spot" dredging, or armoured capping to prevent resuspension during future high flow events. Armouring involves actions to prevent

[10] A meal is defined as 227 grams (eight ounces; a fillet of dinner plate length).

erosion such as adding riprap rock, adding vegetation, landscaping, etc. Dredging the river between Dryden and Clay Lake was investigated by Acres (1983), who indicated the option was feasible using small floating hydraulic dredges with mechanical cutterheads (Parks and Hamilton, 1987). The estimated cost in 1981 dollars was $19 million ($48 million in 2014 dollars) over a 3 year period during the ice free season. Costs could differ significantly now from the 1980s, depending on the present distribution of mercury contamination in river sediments.

5.2.3 Reduce Methylmercury Production in Clay Lake

If proposed field studies show that in-lake methylation is an important source of methylmercury in Clay Lake, there are options to reduce this source; including,

- Reduce inputs of inorganic mercury from upstream (discussed above).
- Reduce the concentration of legacy inorganic mercury in Clay Lake sediments.
- Reduce the in-lake efficiency of converting inorganic mercury to methylmercury.

<u>Enhanced Natural Recovery (ENR)[11]: *Rough estimated cost of $6M/yr.[12].*</u>

Surface sediment mercury concentrations could be lowered by diluting the mercury in surface sediments with clean particles (low-mercury clays and silts) as recommended previously (Rudd *et al.*, 1983; Parks and Hamilton, 1987; Rudd *et al.*, 2013a,b).

For example, to treat Clay Lake, Rudd *et al.* (1983) recommended taking advantage of the large quantity of clean clays and silts in Wabigoon Lake upstream of the mill in Dryden. (Wabigoon Lake sediments are very low in mercury (80 ng/gdw)). It was recommended that a hydraulic dredge be situated in Wabigoon Lake and sediment slurry be pumped/piped down to the Wabigoon River and released below the Wainwright Dam (to prevent trapping of the clean materials behind the Wainwright Dam). The dredge would be operated during times of high river flow, so that the clean resuspended material would be carried down the river from the below the Wainwright Dam and deposited in the surface sediments of Clay Lake.

There are two scenarios whereby ENR could be applied to Clay Lake. First, if it is found that inputs of legacy mercury from upstream of Clay Lake are presently significant and cannot be controlled, then mercury concentrations in fish in Clay Lake could be lowered by about a factor of 2 by doubling the natural sedimentation by the addition of clean clay/silts dredged from Wabigoon Lake. This could be done at an estimated cost of about $6M/yr. based on dredging

[11] This remediation option should not be confused with capping, which covers existing sediments with a new layer of low-contaminant solids.
[12] Based on 3000 ha, 0.3 to 0.4 cm/yr sediment accumulation, roughly 42,000 tonnes per year at $150 per tonne. The number of years of additions would be dependent on target sediment concentrations and sediment mixing depths.

approximately 40,000 tonnes per year at $150 per tonne of clean materials from Wabigoon Lake during times of high flow in the river. Additions would be needed as long as elevated mercury inputs continued from upstream.

Second, if inputs of legacy mercury to Clay Lake could be controlled, Clay Lake would begin to recover naturally, but based on limited data[13], this recovery would take about 14-18 years to reach 200 ng/g and 17-30 years to reach 100-150 ng/g. The natural recovery rate could be accelerated by adding clean Wabigoon Lake sediments to dilute the concentration of mercury in surface sediments more quickly. For example, if the natural sedimentation rate were doubled with the addition of 40,000 tonnes of clean sediment, then it would take 7-9 years to attain a sediment concentration of 200 ng/g at a cost of $42 million to $54 million. Recovery could be faster if the sediment additions were greater. Optimal timing (*i.e.* rates of sediment additions) for this procedure remains to be worked out.

As discussed previously, remediation of Clay Lake may speed the recovery of downstream lakes because concentrations of dissolved and suspended particulate mercury flowing downstream would be lowered. If it was decided to further increase the rate of recovery of downstream lakes, such as Ball Lake, local sources of clean materials could be resuspended in Ball Lake as suggested by Parks and Hamilton (1987). Additional field studies are proposed to assess the effects at downstream locations of localized actions versus reducing methylmercury inputs from upstream.

ENR is considered the leading candidate for mercury remediation in Clay Lake and is recommended for further evaluation. We recommend this procedure because of its likely effectiveness, because it is least disruptive to ecosystem, and because of its comparatively low cost. Depending on how quickly sediments are added, this procedure may take somewhat longer than others to apply.

Capping of surface sediments in both basins of Clay Lake. ***Rough estimated cost of $160M[14]***. This estimate is based on the placement of 10 cm of clean materials onto the surface sediments of Clay Lake. The likelihood of success with this option is high – with two caveats: 1) a cap on the east basin of Clay Lake may need to armoured because this basin of the lake is shallow and unstratified; and 2) recently it has been found that production of methylmercury can be stimulated on the underside of the cap after placement of the cap, so any disturbance of the cap would be problematic (Johnson, 2009). This would be more of a concern in shallow areas, such as the east basin of Clay Lake than deep zones.

We rank capping of Clay Lake sediments as being second in order of priority and it should be considered if ENR is found to be deficient after further investigations. Capping would likely be

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[13] Assuming a mixed surface layer of 2 cm and using the observed sedimentation rate of roughly 0.3-0.4 cm/yr. Mixing depth and sedimentation rate estimates are needed to improve estimates of recovery times.

[14] Based on 3000 ha, 10 cm cap, 1.05 million tonnes of solids at $225 per tonne.

effective, but it is moderately disruptive to the benthic community, and it is more expensive than ENR.

<u>Nitrate additions or aeration of deep anoxic waters of lakes</u>

Production of methylmercury by bacteria occurs at sites of low or no oxygen. Methylmercury production in many lakes is enhanced by the presence of deep waters depleted in oxygen during periods of summer or winter stratification. It has been proposed that methylmercury production could be reduced in lakes with these conditions by increasing oxygen levels via aeration, or by adding nitrate. Nitrate addition studies were carried out in Onondaga Lake, a mercury-contaminated lake in New York state, with the same surface area (3000 ha) as Clay Lake. Nitrate was added near the sediment/water interface in the deep water portions of the Lake in pilot studies from 2011-2013. The nitrate additions effectively inhibited the release of methylmercury from sediment in the deep water portions of the lake, resulting in lower concentrations of methylmercury in lake water and in zooplankton. Lower methylmercury concentrations in zooplankton are expected to subsequently lower fish mercury levels (USEPA, 2015c). Based on the pilot studies, nitrate addition was preferred to oxygenation.

We recommend that oxygen depth profiles be obtained during late summer in all lakes on the Wabigoon-English River System lakes that have high mercury concentrations. If it is found that these lakes have anoxic hypolimnia, aeration or nitrate additions to these lakes on an annual basis should be considered.

Monitored Natural Recovery (MNR): MNR should only be considered if the suspected ongoing inputs of mercury, as discussed above, have been identified and controlled. MNR could be considered if field studies showed that Clay Lake, left untreated, could respond to changes in mercury loading faster than presently expected.

Targeted actions in parts of Clay Lake: Combination of approaches discussed above could be applied to Clay Lake. Targeted approaches in parts of the lake could be employed but insufficient information is currently available to determine if such an approach would succeed. An example of this approach is Onondaga Lake, NY, where localized dredging, capping and nitrate additions are all being employed. Localized dredging was completed in 2014. All dredged areas are being capped. Some areas are being capped only, and capping will be completed in 2016. Habitat restoration and monitored natural recovery are also being used in some areas of the lake (US EPA, 2015c). These in-lake actions were proposed to cost approximately $451 million, excluding nitrate additions (US EPA *et al.*, 2012). At present the success of this approach at Onodoga Lake remains to be determined.

Remediation options ranked with a low suitability

Bank-to-bank dredging of Clay Lake. ***Rough estimated cost of $940M***[15]. This cost estimate is based on sediment removal, transportation, and disposal to a depth of 20 cm in Clay Lake (Sellers, 2005). Costs are based on dredging costs estimated in Rudd *et al.* (2013b).

In addition to its high cost, this option is ranked at the lower level because of the risk of aggravation of the mercury contamination by sediment disturbances. The US EPA generally does not recommend dredging on a bank-to-bank basis for the same reason (Committee on Sediment Dredging at Superfund Megasites, 2007).

Sedimite application to the surface sediments of Clay Lake. ***Estimated cost for a single application $1.5B***[16]***.*** Sedimite is a proprietary form of activated carbon, which prevents methylmercury that has been produced in the surface sediments from being bioaccumulated into the food chain and fish (*e.g.* Gomez-Eyles *et al.*, 2013). This treatment has been applied in Mirror Lake, a 2 hectare pond located in Dover, Delaware (Delaware Online, 2015). Pilot studies have recently been completed on the Penobscot system (Gilmour et al 2013b). That study suggested that treatments would need to be repeated every 5 years (Rudd et al., 2013a,b). This is a quite new procedure that has not yet been applied to lakes the size of Clay Lake. The long term efficacy of this treatment is still not well understood. We give this procedure a low ranking because of its unknown long term efficacy and because of its high cost.

Whole ecosystem selenium additions: Additions of selenium have been found to lower the bioaccumulation of MeHg into the food web and fish (Rudd *et al*., 1983). While this would be a cost effective option for the Wabigoon-English River System, this option is not recommended because of the possible toxicity of the added selenium.

[15] Based on 3000 ha, 20 cm depth, 2.1 million tonnes of sediment at $450 per tonne.
[16] Based on 3000 ha, at $49 per m2 (from a Delaware study, Delaware Newszap (2015))

Table 4. Selected Mercury Remediation Options for the Wabigoon River System

Option	Background	Comments	Recommend for further consideration?
1. Lowering present-day sources of mercury to the Wabigoon-English River System			
Reduce ongoing mercury discharges from the site of the former chlor-alkali facility	If significant present-day discharges site of the former chlor-alkali facility are occurring, this would elevate mercury concentrations in sedimenting particles downstream and elevated methylmercury production in surface sediments.	Various approaches have been successfully used to reduce ongoing mercury discharges from other pulp and paper facilities. If ongoing inputs were large enough and were stopped, this approach alone could hasten recovery (as was the case for Lavaca Bay, TX). If significant, should be done prior to any other remediation measures.	Yes
Reduce downstream supply of total mercury and methylmercury from Wabigoon River sediments above Clay Lake.	Downstream transport of total mercury and methylmercury in this river reach could be occurring particularly at times of high river flow.	Dredging of “hotspot” locations may be warranted. If hotspot erosion is significant, removal should be done prior to any other remediation measures. If significant, should be done prior to any other remediation measures. Capping or river bank armouring at selected locations are also possibilities.	Yes

Option	Background	Comments	Recommend for further consideration?
2. Traditional Engineering Approaches			
Bank-to-bank dredging	Involves removal and disposal of surface contaminated sediments to depths of acceptable mercury concentration.	Dispersion of mercury throughout the Wabigoon –English River System is now so widespread that that dredging and disposal of dredge spoils is impractical for cost reasons. If not done correctly dredging can also re-contaminate surface sediments.	No
Capping	Placement of clean materials over contaminated sediments to depths of 5-10 cm.	Expensive on a very wide geographic scale, but might be considered for Clay Lake. However recent studies have found that capping contaminated sediments can enhance methylmercury production under the cap	Yes, but targeted
Monitored Natural Recovery	Rely on natural processes, combined with long term monitoring	Lowest disturbance to ecosystem and lowest cost. Longest time required.	Not as a standalone option. The time of recovery too long for ANA people after having already waited 54 years. Our opinion is that Monitored Natural Recovery will be too slow, which is why we tentatively recommend ENR to enhance the rate of natural recovery.

Option	Background	Comments	Recommend for further consideration?
		3. Whole-ecosystem additions	
Enhanced Natural Recovery (ENR)	Mercury concentrations in fish are controlled by the mercury concentration of surface sediments. This option dilutes mercury in surface sediment by decreasing the concentration of mercury in sedimenting particles by the addition of clean clay to lake inflows.	Two advantages to this approach are: 1) Relatively low cost, and 2) ENR could improve the mercury situation even if mercury inputs from the river upstream of Clay Lake could not be lowered.	Yes
Nitrate additions	Reduces methylation in some systems where oxygen is depleted during summer stratification	Would only lower methylmercury production in the anoxic hypolimnia of some lakes.	Yes
Aeration	Lakes with anoxic bottom waters tend to have elevated mercury concentrations in fish because of enhanced hypolimnetic methylation.	Would only lower methylmercury production in the anoxic hypolimnia of some lakes.	Yes
Selenium additions	The addition of selenium to ecosystems interferes with the uptake of mercury at all levels of the food web.	This approach is not recommended because of the small window between the beneficial concentrations of selenium and toxic concentrations of selenium.	No
		4. Application of Sorbents	
Sorbent additions to surface sediments	Various forms of activated carbon (*e.g.* Sedimite, biochar) are applied to surface sediments to bind mercury making it less available for bioaccumulation.	The cost of a single application is high on a whole-lake basis. Also, based on pilot studies, applications would need to be repeated every few years.	Not on whole lake basis

6 Adequacy of Existing Information.

The following discussion addresses information needed to assess options to remediate the effects of mercury releases from the former chlor-alkali facility at Dryden. Other human influences on mercury such as logging, climate change, and sewage effluent discharge could be secondary factors affecting fish mercury concentrations, but are outside the scope of this report and the evaluation of data presented below.

6.1 Knowledge Gaps

To determine the best adaptive management approach to remediation, we need to fill in some gaps in our understanding of the present-day mercury situation of the Wabigoon River system. These gaps in our knowledge are presented as questions here. They include:

1. what is the present-day, geographic extent of mercury contamination of the Wabigoon-English River system?
2. are there still significant sources of mercury released into the Wabigoon River at Dryden?
3. is legacy mercury in Wabigoon River sediments between Dryden and Clay Lake still an important source of mercury downstream?
4. is mercury released from Clay Lake important downstream?
5. are there low oxygen zones in Wabigoon-English River lakes that are sites of high methylmercury production?

Clay Lake is considered an important part of the system to examine because

a) the highest measured fish mercury concentrations are in Clay Lake.
b) lowering fish mercury concentrations in Clay Lake would enable the ANA people to re-establish a fishery there.
c) improving the situation in Clay Lake could have beneficial effects downstream, particularly in Seguise Lake and Ball Lake. This would happen at no extra cost if Clay Lake was to be remediated. It is well established from studies elsewhere that methylmercury produced at an upstream location can elevate fish mercury concentrations downstream. Conversely, attempting remedies downstream without addressing Clay Lake may not achieve the desired results downstream.

To fill in the knowledge gaps, specific questions (within the above questions) need to be answered and are listed in the following table. For the sake of linking this section of the report with the next section (Section 7, Recommended Field Sampling Program), a summary description of the field program that would be needed to answer the specific questions is included.

Table 5. Questions and Field Studies Associated with Knowledge Gap 1.

Knowledge Gap 1 The current geographical extent of Hg pollution in the Wabigoon-River system		
Main Question	Specific questions	Data or field program needed
1. What is the present-day geographical extent of mercury contamination of the Wabigoon-English River system?	1.1. What is the present-day, upstream-downstream pattern of mercury pollution in the Wabigoon River between Dryden and Tetu Lake?	1.1. One-time sampling of sediment, fish, crayfish, and water along the Wabigoon River between Dryden and Tetu Lake.
	1.2. Is the pollution between Dryden and Clay Lake lower today than it was 45 years ago? Are the sediments near Dryden cleaning up?	1.2. Return to the same sediment and crayfish sites that were sampled 45 years ago and compare values.
	1.3. Are there “hot spots” of crayfish mercury in the River channel between Dryden and Clay Lake?	1.3. Sampling of crayfish up- and downstream of suspected hot spots of river mercury.
	1.4. What is the mercury status of the Wabigoon-English River prior to remediation?	1.4. Establish a baseline for a monitoring program before and after remediation.

Table 6. Questions and Field Studies Associated with Knowledge Gap 2

Knowledge Gap 2 The extent to which the former chlor-alkali facility site is releasing mercury to the Wabigoon River		
Main Question	Specific questions	Summary field program needed
2. Are there still significant sources of mercury released into the Wabigoon River at Dryden? Questions 2.2 and 2.3 will require access to the site, which will require negotiation. In the meantime we propose to address the question 2.1 and 2.4, which can be addressed by river sampling.	2.1 Does the level of mercury and chloride in the Wabigoon River change as it passes by the former chlor-alkali site?	2.1 Measurement of mercury and chloride in the surface water of the Wabigoon River at Dryden at key sites.
	2.2. Was the concrete pad on which mercury cells were stored removed and the ground below excavated to also remove mercury that leaked through the concrete? Are local disposal sites leaking mercury?	2.2. Assessment of former chlor-alkali facility and associated local disposal sites; review of historical and available documents.
	2.3. Is mercury entering the Wabigoon River via groundwater and/ or seepage from containment walls?	2.3. Measurement of mercury entering the river via groundwater and visible seeps; Engage a hydrologist and an engineering firm to assess the former chlor-alkali facility and carry out ground water remediation.
	2.4. Is mercury coming out of old pipes remaining on-site or presently operating outfalls?	2.4. Measurement of mercury in water flowing out of old pipes and present outfalls at the site.

Table 7. Questions and Field Studies Associated with Knowledge Gap 3

Knowledge Gap 3 The extent to which legacy mercury in Wabigoon River sediments continues to be an important source of mercury downstream		
Main Question	Specific questions	Summary field program needed
3. Is legacy mercury in Wabigoon River sediments between Dryden and Clay Lake still an important source of mercury downstream?	3.1. Are there "hot-spots" of resuspended sediment or bank erosion along the Wabigoon River that are enriching the downstream transport of mercury?	3.1.1. Aerial survey of the Wabigoon River during a high-water period (*i.e.* spring melt) to visually identify and geo-reference sites of resuspension and erosion; and 3.1.2. Measurements of total mercury and methylmercury in the water upstream and downstream of suspected hotspots during high water flow.
	3.2. How important is high water in transporting river sediment, total mercury and methylmercury to downstream lakes?	3.2.1. Measurement of mercury in water entering Clay and Ball Lakes during a range of flow conditions; and 3.2.2. Measurements of water flow at relevant sites along the Wabigoon River.
	3.3. Is the Hg entering Clay Lake sustaining present-day mercury concentrations in the sediments of the lake?	3.3. Measurements of the mercury in water (particles and dissolved) entering Clay Lake and in the surface sediments of the east basin.

Table 8. Questions and Field Studies Associated with Knowledge Gap 4

Knowledge Gap 4 The extent to which mercury released from Clay Lake is important downstream		
Main Question	Specific questions	Summary field program needed
4. Is mercury released from Clay Lake important downstream?	4.1. How much mercury is leaving Clay Lake?	4.1. Measurement of methylmercury and total mercury in water flowing out of Clay Lake especially during fall overturn.
	4.2. How does mercury leaving Clay Lake compare to mercury entering Seguise and Ball Lakes?	4.2. Measurement of methylmercury entering Seguise and Ball Lakes.

Table 9. Questions and Field Studies Associated with Knowledge Gap 5

Knowledge Gaps 5 The importance of anoxia hypolimnia (in Wabigoon River lakes) as sites of methylmercury production		
Main Question	Specific questions	Summary field program needed
5. Are low oxygen zones (in Wabigoon-English River lakes) sites of high methylmercury production?	5.1. To what extent are the deep waters of lakes depleted in oxygen during summer stratification?	5.1. Measurement of dissolved oxygen profiles in Clay, Ball, and Seguise lakes during the summer.

6.2 Data Gaps

Some components of the Wabigoon-English River system have been sampled more often than others. For example, we have long-term data of mercury in fish from Clay Lake, but not in Indian Lake or the Wabigoon River between Dryden and Clay Lake. We have no recent measurements of mercury in crayfish from Clay Lake. We have no recent measurements of mercury concentrations or water quality in the water of the Wabigoon River or in any of its lakes.

Table 10 summarizes where we have data and where we do not. This is emphasized with "N" (no data) and "NR" (no recent data). The table shows that we are lacking data for the Wabigoon River between Dryden and Clay Lake and for mercury levels in water.

Table 10. Summary table of available data in Wabigoon River System.
G = good data, S = some data, NR = no recent data, N = no data.

Compartment	Wabigoon River			Wabigoon and English Rivers		English River
	Wabigoon River between Dryden and Clay Lake	Clay Lake	Ball Lake South basin	Indian Lake	Separation Lake	Ball Lake North basin
Walleye and northern pike	NR	G[i]	S[ii]	S[iii]	S[iii]	G[i]
Crayfish	NR	N[iv]	G[iv,v]	S[v]	NR	S[v]
Sediment	NR	G[v]	G[v]	S[v]	NR	G[v]
Water	NR	NR	NR	NR	NR	NR

[i] see Neff et al, 2012

[ii] data not included for this basin in Neff *et al.* 2012.

[iii] sampled in 2003 only (see Kinghorn *et al.* 2007).

[iv] Published data for 1971 and 1985 (See Parks *et al.,* 1991); unpublished data for 1970 – 1989 (McCrae and Hamilton) and 1997 (Lockhart and DeLaronde).

[v] sampled in 2004 and/or 2007 (see Sellers, 2005 and 2008).

7 Recommended Field Sampling Program

7.1 Full Field Program

The main objective of this section of the report is to describe tasks required to answer the questions presented in Chapter 6. Some of the proposed fieldwork is an update to sampling conducted over the past 45 years but for which there are no recent data. This is particularly the case for mercury measurements in water.

With the execution of the field program we expect that the understanding of mercury movement in river systems generally, and in the Wabigoon River specifically, will be greatly enhanced. What we learn will be applicable not only to the Wabigoon River system and its remediation, but likely to other mercury contaminated sites and systems. As such this project will benefit other researchers, projects and communities dealing with similar issues.

Nine tasks related to field work are:

A. finalizing the field program outlined here,
B. training of youth from ANA in Environmental Monitoring,
C. aerial survey,
D. water sampling for several parameters from Dryden to Ball Lake,
E. sampling for the determination of mercury levels in fish and crayfish,
F. sampling of surface, seepage and ground-water at Dryden for mercury and chloride to determine present-day mercury loads from the former chlor-alkali facility,
G. surface sediment sampling, and
H. estimation of water flow rates.

Three additional tasks are also proposed that are not field-based:

I. Assessment of decommissioning of chlor-alkali facility,
J. Data synthesis and analysis, and
K. Reporting.

We envision this field program as one that will be adaptive. By this we mean that tasks and subtasks will be prioritized, span more than one field season, and be modified as data emerge and questions are answered. What is learned from data collected in the first field season will likely result in adjustments to tasks for subsequent field seasons, as is the nature of this type of work. An adaptive approach may also reveal that supplementary sampling (beyond that what is proposed here) will be required as new questions arise. Recognizing the interest in carrying out remediation, we would only recommend supplementary sampling considered essential.

TASK A: Finalize the Field Program

Frequency: Once

A.1. Review existing technical reports for useful field information (*e.g.* previous sampling sites and sampling times, water flow) that will help develop a detailed plan with respect to sampling, river travel, specific equipment required, time required etc. This review will also ensure the data are comparable to past data;

A.2. Review past and present effluent release data for the facility at Dryden to better understand practices of effluent release;

A.3. Review existing maps and images to identify key areas for aerial photography and sampling;

A.4. Identify and engage local people and expertise for implementation of field program;

A.5. Identify youth from ANA for training in Environmental Monitoring;

A.6. Trip to ANA (and perhaps neighboring communities appropriate for different sections of the River) to study maps, engage local expertise in planning, coordinate with youth and guides, and finalize plan; and

A.7. Secure equipment, supplies and personnel as necessary.

Products:

- Refined field program and budget;
- Youth selected by ANA for training in environmental monitoring; and
- Equipment, personnel, and supplies to support field program.

TASK B: Training of Youth from ANA in Environmental Monitoring.

Frequency: Ongoing

B.1. On-site training in sample collection, handling, storage, and shipment; data recording, data-sharing, equipment maintenance and staging;

B.2. Production of training manual with photos, text, and web-links; and

B.3. Training in other aspects of science according to expressed interest.

TASK C. Aerial Survey of the Wabigoon River System

Frequency: Once

C.1. Charter an airplane and fly over the Wabigoon-English River system with a photographer capable of producing geo-referencing photographs.

Products:

- Detailed images of the Wabigoon and English Rivers from Dryden to the western edge of ANA's Territory;
- Geo-referenced images of areas of concern or sampling interest (*e.g.* logging, inflows, suspended sediment or erosional areas);
- Geo-referenced points of entry onto the River from logging roads;
- Photos and videos of the Wabigoon River that could be used in communication and education; and
- Photos and GIS shape files (*.shp) that can be used in future mapping projects.

TASK D. Water Sampling along the Wabigoon River System

Frequency: As described below

D1. Water sampling will be carried out as described in Table 11.

Table 11. Proposed Water Sampling along Wabigoon River system

Parameter	Sites not yet determined*		Targeted sites	
	Number	Frequency	Lake	Frequency
Total mercury** (particulate and dissolved) during range of flow conditions	10	9-12	Clay, Seguise, and Ball Lake inflows and outflows	10/site
Methylmercury **	2	3	Clay, Seguise, and Ball Lake inflows and outflows	5/site
Dissolved organic carbon	2	3	Clay, Seguise, and Ball Lake inflows and outflows	5/site
Dissolved oxygen	2	3	Clay, Seguise, and Ball Lakes	8-10/site
Total Suspended Solids	10	9-12	Clay, Seguise, and Ball Lake inflows and outflows	10/site
Chloride	10	9-12	Clay, Seguise, and Ball Lake inflows	10/site

* To be determined based on Task A.

** Mercury samples to be sampled in duplicate

Products:

Data that will be used to determine:

- the geographic scope and baseline of mercury in the water;
- mass movement of mercury along the river and into specific lakes;
- whether methylmercury produced at the bottom of lakes during the summer is a) a function of anoxia and b) released downstream during fall turnover;
- the extent of anoxia in the deep water of Clay, Seguise and Ball Lakes. These data are needed to determine if nitrate additions or aeration might be useful remediation techniques; and
- water flows and tributary dilution at key locations along the Wabigoon River, *e.g.* inflows to Clay, Seguise, and Ball Lakes

TASK E. Sampling for determination of mercury levels in fish and crayfish

E.1. Fish Sampling

Frequency and location: Once, likely in 4 lakes that are not sites included in routine MOECC sampling)

Collect walleye and northern pike for mercury analysis. Attempt to catch at least 30 fish spanning a range of sizes (thus allowing estimates of mercury in fish of a standard size).

Products: Data that will be used to:

- Help determine the geographic scope of elevated fish mercury, and
- Set pre-remediation baselines for fish mercury.

E.2. Crayfish sampling for determination of Hg in crayfish

Frequency and location: Once at 16-20 sites along the Wabigoon-English River and associated Lakes, repeating some of the 24 sites sampled by Freshwater Institute scientists in the 1970s, 6 of the sites sampled by Parks et al (1991) and 6 of the sites sampled by Sellers (2005).

Collect crayfish for analysis of THg

Products: Data that will be used to:

- determine the geographic scope of elevated crayfish Hg,
- determine hotspots of crayfish mercury along the Wabigoon River,
- set pre-remediation baselines for crayfish, and
- allow for assessment of the natural recovery of the Wabigoon River during the past 45 years.

TASK F. Sampling at Dryden to determine present-day mercury loading into the Wabigoon River

Ongoing losses of mercury from the former facility site have not been estimated since the 1970's - 1980's. Continued losses of mercury from former chlor-alkali facilities, where controls are already in place, are common (for example, HoltraChem-Penobscot River MA; ALCOA-Lavaca Bay TX; Squamish BC). At some locations, additional controls have been instituted after the initial controls in the 1970's more recently further lowering ongoing inputs. These have been successful. For example, at the HoltraChem site high-level treatment of the facility effluent is now in place, and ground water pumping is ongoing to capture mercury contaminated ground water before it can seep into the Penobscot River. As a result, estimated losses of mercury from the HoltraChem site to the river have now been lowered to about 2 kg per year (Rudd *et al*., 2013).

Three sub-tasks are recommended to determine present-day mercury loads to the Wabigoon River from the site of the former chlor-alkali facility. The data collected will help to determine:

- mercury in ground water compared to benchmark levels,
- loading of mercury into the Wabigoon River from groundwater and seeps,
- loading of mercury from into the Wabigoon River from pipes, and
- the influence of the former chlor-alkali facility site & disposal site on mercury levels in the water.

F.1. Wabigoon River up-and downstream of key sites

Frequency: 9-12 times.

F.3.1. Sample surface water up- and downstream of the former chlor-alkali facility (and its disposal area) for mercury and chloride.

F.2. Groundwater and above-ground seeps

Frequency: To be determined once groundwater hydrologist is consulted.

F.1.1. Install ground water wells for determination of groundwater flow and to facilitate groundwater sampling for mercury (both needed to determine mercury flowing into the river via groundwater);

F.1.2. Sample for radon in ground and surface water to detect the occurrence of seepage; and

F.1.3. Measure mercury in seeps from river channel, berms or containment walls.

F.3. Discharge Pipes

Frequency: 9-12 times.

F.2.1. Measure end-of-pipe discharges from the facility site.

TASK G. Surface sediment sampling (using coring technique) from Wabigoon to Tetu Lake

Frequency and location: Once at 24-30 selected sites that encompass lakes and the Wabigoon River channel, repeating some of the 24 – 30 previous sampling sites.

G.1. Collect sediment cores.

Products: Data that will be used to determine:

- the present-day geographic scope of elevated sediment mercury,
- whether sediments near Dryden have cleaned up,
- if the geographic peak of sediment mercury has migrated downstream since the 1970s,
- the natural rate of recovery of mercury in the Wabigoon River system during the past 45 years,
- a pre-remediation baseline for sediment mercury levels, and
- the mixing depth in the surface sediments of Clay Lake.

TASK H. Estimation of Water Flow rates

<u>Frequency and location: Once for 4 sites along the Wabigoon River</u>

H1. Monitor water flow rate and chloride ion concentration at Dryden and targeted sites along the River.

<u>Products:</u>

- Water flow data needed to determine mass movement and dilution of mercury concentrations along the river and into Clay, Seguise, and Ball Lakes.

7.2 The First Field Season (2016-2017)

Within the field program described above there are priority tasks to reflect the priority questions. Therefore what we propose for the first field season will address several (but not all) of the specific questions among the main questions (see chapter 6). In the first field season we will focus on the first part of Wabigoon River between Wabigoon Lake and the outflow of Clay Lake.

Our first priority for sampling will be surface water between Wabigoon Lake and the outflow of Clay Lake. We propose to sample at 13 sites that will be accessible from a boat launch or float plane. Sampling is designed such that the data will allow us to determine:

a) if the former chlor-alkali site or disposal area is releasing mercury to the Wabigoon River,
b) the amount and significance of total mercury and methylmercury entering Clay Lake from the Wabigoon River,
c) the amount and significance of total mercury and methylmercury leaving Clay Lake (with a priority being during fall overturn), and
d) the amount of total mercury and methylmercury entering Ball Lake.

Our second priority would be the sampling of surface sediment, using the coring technique at sites where this is possible. Sampling will be designed such that the data will allow us to determine:

a) the present-day upstream-downstream pattern of sediment mercury between Wabigoon and Clay Lake (where is the highest sediment mercury?),
b) if the mercury at selected sites in this stretch are lower today than they were 45 years ago, and
c) a pre-remediation baseline for mercury in sediment.

Our third priority would be the sampling of crayfish at selected sites along the River, most of which would repeat the sites sampled 45 years ago. Sampling will be designed such that the data will allow us to determine:

a) the present-day upstream-downstream pattern of crayfish mercury between Wabigoon and Clay Lake,
b) if there are "hot-spots" along the river where mercury is getting into the food web,
c) if the upstream downstream pattern of crayfish mercury has changed in 45 years, and
d) a pre-remediation baseline for crayfish mercury.

At the time of writing the final version of this report, the cost associated with these three priority sampling tasks is estimated at $293 000 (CDN). This cost includes the professional fees necessary to direct and execute the field program, field and lab technical support, data analyses and writing, and community meetings. This budget was submitted to ANA in March of 2016.

8 Remediation Costs

Remediation of the Wabigoon-English River system would likely require a combination of activities. It would not make sense for example to pursue sediment remediation in the Wabigoon River or contaminated lakes if mercury releases are found to continue at meaningful rates from the site of the former chlor-alkali facility. This source should be investigated and if necessary, eliminated, before other remediation is pursued. Similarly it would be desirable to reduce ongoing mercury supply to the river from sediments in the Wabigoon River, if important, then evaluate the need to take further action in Clay Lake.

Additional information is required to establish the need and scope of effort to reduce mercury sources from the former chlor-alkali facility site and sediments in the upper Wabigoon River. Until that time, accurate cost estimates are not possible for an overall remediation strategy. Sufficient information is available however to demonstrate that some remediation activities would be expensive, such as bank-to-bank dredging (~$940 million) or Sedimite application ($1.5 billion) in Clay Lake. The least expensive option to reduce concentrations of total mercury in Clay Lake is ENR with low-mercury solids at a rough cost of $6 million per year. Additional studies recommended would help to better estimate how many years of application would be required. Costs associated with supplementary actions such as aeration or nitrate addition in Clay Lake should also be further investigated.

9 Conclusions

Recovery of fish mercury concentrations in the Wabigoon-English system from mercury pollution, which began in in the 1960's, has stalled in the most contaminated part of the system, and appears to be spreading to the lower reaches of the system. However we believe that recovery could be restarted and accelerated, using methods described in this report.

Potential causes of the stalled recovery include: (1) if there are present-day inputs of mercury from the former chlor-alkali facility to the Wabigoon River at Dryden, and/or (2) if there is ongoing transport of contaminated sediments from the river between Dryden and Clay Lake into Clay Lake. It is also possible that the ability of Clay Lake sediments to eliminate mercury contamination is slower than estimated with available data. If meaningful amounts of mercury are still being lost from the former chlor-alkali facility and/or from the river sediments between Dryden and Clay Lake, the first step in remediation should be to reduce these ongoing inputs to Clay Lake by site remediation of the former chlor-alkali facility, and/or by hot spot dredging or capping of the Wabigoon River above Clay Lake.

A second step of remediation should be treatment of Clay Lake itself, which is the most contaminated lake in the Wabigoon-English system. Several remediation methods were considered. Enhanced Natural Recovery (ENR) in surface sediments is the leading candidate. It offers the advantages of being least disruptive to the ecosystem because natural processes and materials are employed, and it is also least expensive, but its application time would likely be longer than other methods, such as capping. If on further investigation of ENR is found to be deficient, capping of Clay Lake by clean sediments should be considered, although it would be more disruptive to the ecosystem and have a higher cost than ENR. A third option to further speed recovery, which could be used in conjunction with ENR or capping, would be to inhibit methylmercury production in the anoxic hypolimnia of lakes in the system by additions of nitrate or by aeration. However at this point we do not know if anoxic hypolimnia are present in any of the lakes in the Wabigoon-English system.

A third step of remediation would be the lowering of fish mercury concentrations in the lakes downstream of Clay Lake. There are data suggesting that fish mercury concentrations in Ball Lake and the lower lakes are being sustained by downstream transport of methylmercury from Clay Lake. Further investigations are needed to establish if this is true. If it is, fish mercury concentrations in Ball Lake and in the lower lakes should begin to decline, with no further treatments necessary, after Clay Lake has been remediated. If it is found that methylmercury transport from Clay Lake is unimportant, then local treatment of Ball Lake by ENR, capping or nitrate additions could be considered at that point in time.

Field studies are recommended to provide updated mercury information in water, sediments, fish and crayfish in the Wabigoon-English River system. These data are needed to provide a measure of the present-day geographic extent of contamination, and to better understand the behaviour of mercury in the system, including areas that may be ongoing sources mercury. These data would narrow the focus of remediation options and provide a baseline set of measurements that represent the beginning of a monitoring program needed to evaluate the effectiveness of remediation.

10 Recommendations

Overall, it is recommended that several remediation approaches be further evaluated for the potential application in the Wabigoon-English River system.

General recommendations:

I. Begin with targeted field studies to better select and prioritize the appropriate remediation options;
II. Follow an adaptive management approach that is supported by an appropriate funding model; and
III. Begin a long term monitoring program to establish baseline conditions and follow the success of remediation as it proceeds.

Specific recommendations in order of their application to the system:

1. Start at Dryden and proceed downstream;

2. Determine the geographic extent and severity of mercury pollution in the river system (see Chapter 5.1.1 and Chapter 7);

3. Determine if present-day sources of mercury in the Wabigoon River upstream of Clay Lake are responsible for the stalling of recovery of high mercury concentrations in fish and sediments in the river, Clay Lake and downstream lakes (see Chapter 5.1.2 and Chapter 7);

4. If present-day sources of mercury upstream of Clay Lake are found to be important, control/reduce these sources before any other specific remediation measures are applied (See Chapter 5.2.1 and 5.2.2). If present-day mercury sources upstream of Clay Lake are found to be small, proceed to 5;

5. If certain remediation procedures are deemed acceptable for Clay Lake, apply them using the adaptive management approach discussed above;

6. To establish the long term efficacy of applied remediation procedures begin an annual monitoring program for mercury concentrations in fish, sediments and water the Wabigoon-English River System;

7. A panel, including Reed Harris, John Rudd, Patricia Sellers and a representative from ANA are recommended to guide the overall program and make recommendations to ANA. Other experts may need to be involved on an as-needed basis. If/when an engineering firm is retained, it would make sense to add a member from that firm;

8. Establish a coordinator position to lead the field studies. We recommend Patricia Sellers, who has extensive experience in the field of mercury pollution, the Wabigoon River system, and working with ANA and neighbouring First Nations;

9. Initiate efforts to involve youth from ANA for the purposes of capacity building, particularly in the field of environmental monitoring; and

10. Clarify the need for remediation of mercury contamination in the Wabigoon River system with the Ontario Government, and in what geographic areas.

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