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Rebuilding fish-human relationships by quantifying combined toxicity and evaluating policy related to legacy contamination

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REBUILDING FISH-HUMAN RELATIONSHIPS BY QUANTIFYING COMBINED TOXICITY AND EVALUATING POLICY RELATED TO LEGACY CONTAMINATION

By

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Author Contribution Statement

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All authors were integral to the process of developing the research guidance that is explained here. Author 3 is the wisdom underlying this manuscript. The first and second authors conducted the data gathering and all the authors contributed to drafting of the guidance. The first author was the lead in drafting this manuscript. The second and third authors supervised this process and advised on strategies, particulars of the synthesis, and ensured meaningful interpretation for KBIC. The second author reviewed and edited the manuscript through completion.

Chapter 3 will be submitted for publication in 2023. Here is a list of co-authors in no particular order: Emily Shaw, Noel Urban, Judith Perlinger, Valoree Gagnon, Cory McDonald, Evelyn Ravindran, Erin Johnston, and Dione Price. All authors were integral to the process of developing the manuscript.

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Abstract

The central theme of this dissertation is relationships – building relationships as research partnerships, disrupting relationships through chemical contamination, and upholding existing relationships (i.e., responsibilities) to address industrial legacies. In partnership with the Keweenaw Bay Indian Community Lake Superior Band of Chippewa Indians (KBIC), this dissertation focuses on rebuilding fish-human relationships within the context of chemical contamination. By quantifying combined toxicity and evaluating the efficacy of cleaning up contamination, conclusions from this work help empower people to maintain practices and knowledges related to fish.

In chapter 1, I positioned myself, a white, American settler scholar, within the context of Indigenous research grounded in Anishinaabe philosophies. My research is predicated on knowledge being a collection of practices that builds and maintains relationships with people and the environment. Being an indigenist researcher means being accountable to those relationships.

In chapter 2, I co-created a research guidance document with KBIC to provide holistic guidance and specify support that enriches their efforts to protect and restore land and life. Our guidance uses the Medicine Wheel to illustrate an interconnected system of partnership teachings that include systems of mutual expectations and responsibilities. The guidance aims for balance between and among four seasons of research: relationship building, planning and prioritization, knowledge exchange, and synthesis and application.

In chapter 3, I used a national database of fish tissue contaminant concentrations to evaluate frameworks for quantifying toxicity, spatial distributions of the components of toxicity, and variations in relative importance of chemicals in different fish types. Based on the results, I argue for using the most sensitive endpoint for components of a chemical mixture rather than the current framework that expects a shared toxic pathway. Research results show that the former is more protective and therefore represents a more appropriate strategy for protecting human health and the environment.

In chapter 4, I compared PCB trends in the Great Lakes basin to evaluate the efficacy of Canada’s 2008 PCB reduction policy. My results show that local reductions of PCB stocks significantly reduced atmospheric PCB concentrations, but a comparable response was not seen in fish tissue. I suggest that fish tissue, as the primary exposure pathway, should be the medium monitored to evaluate policy efficacy.

1 Relationships as a research method

1.1 Overview of the researcher and research on toxic contamination

The central theme of this dissertation is relationships – building relationships as research partnerships, disrupting relationships through chemical contamination, and upholding existing relationships (i.e., responsibilities) to address industrial legacies. The use of relationships as my research framework calls attention to the unique interactions between chemicals and the environment, including plants and animals. Manufacturing petrochemicals (e.g., PCBs) "…weaponises [sic] these fossil-kin, these long-dead beings, and transforms them into threats to our very existence…" (p. 104 Todd, 2017). Part of the reality of chemical contamination is learning and accepting a life of ongoing engagement with contamination, and its presence does not absolve humans of their on-going responsibilities to caretake contaminated landscapes (Hoover, 2020; Todd, 2017). Part of cultivating fish-human relationships within this context is the development of frameworks to empower people to maintain practices and knowledges related to fish.

As a white, American, settler scholar with German and British ancestry, conducting Indigenous research grounded in Anishinaabe philosophies requires reflexive practices and on-going engagement and deep-thinking about my positionality in my research (Kwame, 2017). Sharing this positionality with others shows to whom I am accountable (my research partners) and reveals perspectives that influence my research. While Dominant Science emphasizes neutrality and objectivity, I agree with Absolon and Willett (2005), that in research such things do not exist. Others have referred to outsiders (i.e., non-Native researchers) doing research rooted in Indigenous philosophies as *indigenist* scholars (Wilson & Hughes, 2019). As an indigenist researcher, my work has transformed me (Absolon 2011). Beyond doing quality and ethical science, my research exists in a reality where knowledge is a practice that builds and maintains relationships with people and the environment. Being an indigenist researcher means being responsible for and accountable to those relationships.

Indigenous Knowledges are a collection of place-based practices (Cajete, 1994; Simpson, 2017; Watts, 2013) that are dynamic and able to adapt to the introduction of new relations. Faced with new relatives, or old relatives in a new shape (i.e., fossil-kin as petrochemicals), knowledge practices re-create relationships by "… embrac[ing] impure and damaged forms of life" (i.e., alterlife; p. 500 Murphy, 2017). Often delayed, invisible, and attritional, the *slow violence* of chemical exposures is "… typically not viewed as violence at all" (p. 2 Nixon, 2011). However, animals of all kinds, including humans, are vulnerable to the legacies of contaminants such as polychlorinated biphenyl (PCB) compounds, dibenzo dioxins (PCDDs), and dibenzo furans (PCDFs), as well as mercury (Hg), and are at risk of consequences of toxicity due to exposure. Place-based knowledges can be more vulnerable to contamination because of the toxic risk associated with maintaining these practices (i.e., harvesting and consuming fish).

By turning away from damage-centered research and towards desire-centered research (Tuck, 2009), this dissertation endeavors to rebuild fish-human relationships that are central to Ojibwa ways of being. In the Great Lakes basin, Superfund sites and Areas of Concern underscore places where industrial ventures exceeded ecosystem thresholds. However, it is a colonial philosophy that views the environment as a repository for human wastes (Liboiron, 2021). Embracing the *alterlife*, this work rebuilds relationships within the reality of chemical contamination. All the compounds considered in this work, even naturally occurring mercury, have environmental patterns tied to human activity. Further, these compounds are known to cause harm to humans and wildlife and therefore, amidst Ojibwa ways of being, there is the potential for major environmental and human health risks.

A major shortcoming of current strategies addressing chemical contamination is that they are based on a mistaken premise that environmental contamination is many thousands of discrete entities rather than a complicated mixture. Polychlorinated biphenyl (PCBs) compounds were manufactured as chemical mixtures with tradenames such as Aroclors (US), Kanechlors (Japan), and Delors (Czechoslovakia). Manufactured as mixtures, there are 209 possible PCB congeners that are commonly simplified into two groups: dioxin-like (DL) PCBs and non-dioxin-like (NDL) PCBs. DL-PCBs are structurally similar to dioxins in being co-planar while the NDL-PCBs are not. The U.S. was the primary producer of PCBs, but production in the U.S. ended in 1993 (Breivik et al., 2002; Melymuk et al., 2022). Polychlorinated dibenzo-p-dioxins (dioxins; PCDDs) and polychlorinated dibenzofurans (furans; PCDFs) are combustion byproducts (e.g., forest fires, metal smelting, waste incineration), and creation and distribution patterns vary among continents (Dopico & Gómez, 2015). As for PCBs, there are many congener arrangements; there are 75 dioxin and 135 furan congeners. A few (7 PCDDs and 10 PCDFs) are considered toxic and therefore quantified by monitoring agencies (Tuomisto, 2019). Mercury is a naturally occurring metal that is emitted during fossil fuel combustion and artisanal gold mining. Present in the environment in many forms (e.g., elemental (Hg(0)) and oxidized (Hg(II)), the organic form (i.e., methylmercury (MeHg)) poses the greatest risk to human and animal health, in part because it biomagnifies.

As globally dispersed compounds, the chemicals considered in this work are of particular importance in the Great Lakes basin because their physicochemical properties frequently result in their being transported towards higher latitudes (Perlinger et al., 2016; Wania & MacKay, 1996). Four of the compound groups (PCBs, DDs, and DFs) have log K_{ow} values greater than 4 (Hawker & Connell, 1988; Sarna et al., 1984). Mercury with a log K_{oa} of 1.1 is also classified as an atmosphere-surface exchangeable pollutant or "ASEPs" (Perlinger et al. 2016). High log K_{ow} and K_{oa} values means that under equilibrium conditions, dissolved-phase and vapor-phase concentrations, respectively, are low relative to concentrations in organic material (e.g., sediment and fish tissue). These relationships mean that these contaminants tend to be passed up the food web through bioaccumulation, leading to and resulting in higher concentrations in predators (e.g., Houde et al., 2008). Lake sediments also accumulate these compounds, meaning bottom-feeding organisms are also highly exposed (Schneider et al., 2001). Waterbody characteristics strongly influence the aquatic food webs and thus exert influence on contaminant concentrations in fish (Chen & Folt, 2005; McCusker et al., 1999). Further complicating the fate and transport of these chemical groups is the diversity within each group. Although grouped into compound classes, (e,g., PCBs, dioxins, furans), there are 419 unique chemicals among these three compound classses. Differences in log K_{ow} and toxic effects make it difficult to generalize except to say that they are persistent, bioaccumulative, and toxic.

Rebuilding fish-human relationships is the primary and overarching objective of this work. To accomplish this, we co-created a research guidance document to guide university-Indigenous partnerships with an emphasis on the Keweenaw Bay Indian Community (KBIC; chapter 1). Putting our guidance into practice, we avoided focusing on disparity, deprivation, disadvantage, dysfunction, or difference (5D data; Walter, 2016) by accepting the reality of chemical exposure and establishing a combined toxicity framework to represent risk more acceptably (chapter 2). Finally, we evaluated PCB policy within the Great Lakes basin to propose additional policy action with the potential to mitigate chemical contamination harm to future generations of fish and humans (chapter 3).

1.2 Overview of dissertation chapters

Positioned as my methods, chapter 2 focuses on building relationships as the groundwork for research partnerships. Together with the KBIC Natural Resources Department staff, we co-created the seasons of research, a research guidance that provides the foundation for the remainder of this dissertation. Moving through the four seasons of research – *relationship building*, *planning and prioritization*, *knowledge exchange*, and *synthesis and application* – we participate in research partnerships with shared responsibilities and expectations for all partners. We create shared understandings and experiences to engage in solidarity to revitalize land, water, and life. Aligning research goals with community priorities facilitates work that builds community capacity and achieves shared visions of environmental relationships. Focusing on KBIC priorities means that consideration of *Anishinaabe gikendaasowin*, Anishinaabe knowledge – the place-based practices and relationships that describe their philosophies – is a necessity (Geniusz, 2009). Rather than incorporating this knowledge into Dominant Science, we strive to bridge, braid, and weave together knowledge to conduct mutually beneficial research (Reid et al., 2021; Colbourne et al., 2020; Freeman & Katwyk, 2020; Kimmerer, 2013).

Chapter 2, seasons of research, makes two important contributions. First, this guidance established shared responsibilities and expectations for research partnerships between the Keweenaw Bay Indian Community and Michigan Tech. The accompanying brochure is a useful tool that can be shared with potential partners as a first step in learning about KBIC priorities. Through shared research priorities, KBIC can build their capacity and rebuild relationships in many ways. Second, the academic publication contributes to the scholarship about bridging knowledge systems by sharing an updated table with other Tribal research guides/resources. In the article we also share context, thoughts, and reflections about the process of co-creating the guidance in hopes that other Indigenous-university partnerships might follow a similar pathway of their own.

Understanding Anishinaabe Ojibwa knowledge as a collection of relationships, chemical contamination is easily recognized as a disruptor of fish-human relationships. Exposure to PCBs has been shown to exert toxic effects on both fish (Berninger & Tillitt, 2019) and human health. Contamination embodies a failure on the part of humans to care for fish and aquatic habitats which are part of our reciprocal responsibilities to other species, a worldview articulated as *Nishnaabeg Internationalism* (Simpson, 2017). Limiting fish consumption to minimize risk interrupts practices associated with harvesting and consuming fish; that is, chemical contamination disrupts, and can sever, the knowledges that guide reciprocal relationships with fish. Chapter 3 considers chemical contamination within the context of fish-human relationships to refine the understanding of the hazards associated with fish consumption, or in other words, to represent health risk more holistically. Improved hazard frameworks empower fish harvesters to re-define acceptable levels of risk. Further, such improvements can guide future and just action to effectively manage chemical contamination.

Chapter 3 advances the science of toxicity by illustrating and contrasting frameworks for defining mixture and combined toxicity. Consideration of both similar and dissimilar effects of five different compound classes facilitates the evaluation of the variation of relative and absolute concentrations across U.S. ecological regions and between fish composite types. This work addresses three questions: *Why should managers and scientists quantify the mixture and combined toxicity of exposure to PCBs, dioxins, furans, and mercury?, What is the variation in relative and absolute contributions to combined toxicity across ecoregions and between fish composite types?,* and, *What constitutes an effective strategy for quantifying mixture and combined toxicity of chemical mixtures?* Our results reveal that consideration for the most sensitive toxic effect (i.e., dissimilar toxic effects) is more protective than a shared reproductive effect. This work provides evidence that the current accepted framework for mixture toxicity, requiring shared endpoints, may not be the most protective strategy. Further, regional variability in the magnitude and contribution of combined toxicity reiterates the need for localized strategies to remediate chemical contamination and manage risks. In assessing combined toxicity methods across disciplines, I aim to refocus considerations on the intended primary objectives of risk assessments: protecting human health and the environment.

Transboundary pollution (e.g., PCBs) requires international cooperation to mitigate existing risks and develop effective solutions (Norman 2015). Abundant in the Great Lakes basin, PCB contamination continues to be the primary reason for creating and distributing fish consumption advisories. Eliminating ongoing sources of chemicals (e.g., of PCBs) is a possible strategy for re-building fish-human relationships in the presence of chemical contamination (e.g., Todd 2017). Localized sources of PCBs have a localized impact on the environment (Totten et al., 2006; Venier et al., 2014). Putting the *seasons of research* into practice, chapter 4 evaluated the effect of localized PCB removal. This is particularly important because this analysis recognizes PCBs as an important contributor to combined toxicity, especially for bottom-dwellers, Chapter 4 evaluates the efficacy of Canada's PCB Regulations (SOR 2008-273) policy, in reducing atmosphere and fish tissue PCB concentrations in the Great Lakes basin. Aligned with the Stockholm Convention, SOR 2008-273 accelerated the safe destruction and storage of PCBs, an atmospheric-surface exchange pollutant (ASEP) that moves through the environment. Specifically, this work seeks to address one question: *Can local regulation of ASEPs significantly reduce local exposure to these contaminants, and, if so, in what environmental reservoirs?* Our results show that local reductions of PCB stocks can significantly reduce atmospheric PCB concentrations. However, after 10 years of implementation in Canada, a comparable response is not yet seen in fish tissue. The different policy impacts complicate considerations in determining whether a policy was successful. Fish consumption is the primary exposure pathway, and for many within the Great Lakes basin their PCB body burden may be more relevant than atmospheric PCB concentrations.

Centering relationships as a research method, when practiced in a good way (see Shaw et al. 2022), can be mutually beneficial because it advances community priorities (Whyte, 2017) and creates opportunities for shared learning by/from all research partners. While finalizing chapter 3 for publication, a group meeting between me, Evelyn Ravindran, Dione Price, Erin Johnston, and Valoree Gagnon involved discussions about the details of the toxicity frameworks and the value of fish-human relationships. This discussion improved the discussion of our results by embedding them within Anishinaabe philosophies of responsibility and relationships. Further, evidence shows that diversity in research groups (e.g., cultural, gender, orientation, racial, etc.) leads to better science (Warkentin, 2019). Doing this kind of collaborative research better ensures that Dominant Science tools consider, and be directed by, community priorities.

Collectively, this research both articulates and enacts an ethical research partnership framework that prioritizes relationships. Chapter 2 is co-created with guidance from KBIC Natural Resources Department staff; the ideas are also shared more widely in a trifold brochure designed for the public, as well as an academic publication (Shaw et al. 2022) to reach multiple audiences. In chapter 3, I consider chemical contamination as a disruptor of fish-human relationships and endeavor to support the rebuilding of those relationships by representing risk more holistically. In chapter 4, I apply what was learned about the magnitude of contamination and evaluate the efficacy of localized reductions of PCB stocks in reducing fish tissue PCBs. Similar policy-driven strategies have the potential to decrease vapor-phase and fish tissue PCB concentrations globally.

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2 Seasons of Research with/by/as the Keweenaw Bay Indian Community

2.1 Introduction

2.1.1 Positionality

As authors of this manuscript, we are researchers in different institutions and hold diverse roles, an environmental engineering graduate student, an assistant professor in human dimensions, and a natural resources director in upper Michigan. Our individual and collective energy are within food sovereignty research and action yet our approaches to this work are quite distinct. Together, we serve as mentors to each other, sharing our unique experiences and expertise, and contributing to each other's professional and personal growth in partnership research.

I am Emily Shaw, an environmental engineering graduate student, a white settler American scientist, with German and British ancestry, living and working in Northern Michigan. My path as an environmental scientist is grounded in education, teaching Great Lakes ecology at Inland Seas Education Association. My involvement with this work reflects my path to understanding the centrality of relationships and the necessity for stewardship to grow from responsibilities rather than rights to the environment. It is these responsibilities that have shifted my understandings of fish relations, connecting them to dynamic food systems and their disruption by chemical contamination; I recognize anthropogenic contamination as an issue of food sovereignty rather than solely a remediation issue.

I am Valoree "Val" Gagnon, an early career human dimensions assistant professor (she, her, ki, kin), and a naturalized U.S. citizen and Korean adoptee, who lives and works within the homelands of the Ojibwa people. My interdisciplinary expertise is in environmental policy, Indigenous food sovereignty and community-engaged research. I focus on the socio-cultural and -ecological impacts of legacy toxic compounds and the policies intended to address them, particularly on fishing communities. My research, teaching, and service center on elevating Indigenous peoples and knowledge, facilitating equitable research practice and design, and guiding partnerships that prioritize the protection and restoration of land and life in the Great Lakes region.

I am Evelyn Ravindran, a natural resource director of a Tribal Nation in Michigan, and an enrolled member of the Keweenaw Bay Indian Community (KBIC), who lives and works along the shores of Lake Superior. In working for the KBIC for more than three decades, I serve in many capacities for the protection of treaty resources and revitalization of food sovereignty. My main priorities are to share KBIC stewardship and governance practices for Lake Superior and to work in partnership with others for the restoration and protection of relationships between water, air, fisheries, and forests, and many other plant and wildlife communities.

Through time, and our shared work, our experiences have remained a way to relate to each other. Our identities are braided together, maintaining their individual integrity in much the same way that we bridge Indigenous and Western sciences. Throughout this manuscript we use our names and alternate between pronouns to reflect Anishinaabe Ojibwa and settler experiences (Tuck and Yang 2012). As we write, we use our individual as well as collective pronouns (e.g., our community) to describe and illustrate the research process and to denote our ongoing partnership. For two of us, this is not a declaration or claim of Ojibwa citizenship. We deliberately use both Indigenous peoples and Indigenous Nations. Indigenous peoples refers to a group of people who are Indigenous, including Nations, but may or may not be legally or politically recognized as Indigenous Nations. Indigenous Nations refers to groups of Indigenous peoples who are also legally or politically recognized by/as Nations. Both terms are necessary in/for different contexts; both are consequential of settler colonial practices and processes. Finally, we have chosen to use the capitalization convention (i.e., Nation and People) when referring to specific Indigenous peoples or nations. In some instances, we generally refer to Indigenous Nations (which is simultaneously generic and specific) to acknowledge that Indigenous Nations have specific place-based knowledge and history with their lands. This same convention applies to our other relatives, too. For example, we might refer to fish nations or the Walleye Nation.

2.1.2 Background and Purpose

In this section, we begin with background information to establish the socio-political history and identity of the Keweenaw Bay Indian Community. Then, we briefly describe the story on the motivation and purpose for creating Seasons of Research together, including the importance of university-Indigenous community partnerships in research. The section concludes with an overview of the remainder of the paper centering Seasons of Research.

2.1.2.1 The Keweenaw Bay Indian Community Lake Superior Band of Chippewa Indians

The Keweenaw Bay Indian Community Lake Superior Band of Chippewa Indians is the successor in interest of the L'Anse and Ontonagon Bands of Lake Superior Chippewa Indians, and signatories to two treaties of peace with the United States (Treaty with the Chippewa 1842, 1854). KBIC is the oldest federally recognized tribe in the State of Michigan (1936), and they retain the largest land base. Comprising large areas of forested land, and diverse aquatic and terrestrial plants and wildlife, the region has vast lake and river systems with more than one hundred sixty tributaries and seventy miles of southern Lake Superior shoreline (Sweat and Rheaume 1998). The KBIC are part of the Anishinaabe, meaning "original person" (Benton-Benai 1988), one of the largest groups of Indigenous peoples on Turtle Island (present-day Americas) with nearly 150 different bands living throughout their homeland in present-day United States and Canada (Inawe Mazina'igan Map Project 2021). The Anishinaabe are known as the Three Fires Council, composed of three tribes known by various names: 1) Chippewa, Ojibway, Ojibwe, or Ojibwa, 2) Ottawa or Odawa and 3) Potawatomi or Bodewadomi (Gagnon et al. 2020). All are related to the Anishinaabe, the larger group of Indigenous people who migrated from the Atlantic shores of North America and began settling in the Great Lakes region before 1000 AD.

The disconnections, and many times direct separation, between Indigenous nations, Indigenous peoples, and Indigenous landscapes often share a common history: exploitation, dispossession, and extraction of Indigenous lands and livelihoods because of processes and practices inherent in/to colonialism and settler colonialism, capitalism, and industrialization (Simpson 2017, Whyte 2018). These relationships continue to be strongly influenced by settler colonial frameworks, particularly in law and policy (Gilio-Whitaker 2019) as well as the sciences (Liboiron 2021) that shape and reshape Indigenous lands and experiences (Hoover 2017). These themes are also central to the disconnected relations and lands and the stories of being KBIC Ojibwa. (For more on the socio-political history and present of the KBIC, see Gagnon 2016.) However, as many scholars have shown, research relationships are also being shaped and reshaped by Indigenous peoples, their priorities, and the Indigenous sciences. (For more on KBIC research partnerships and priorities, see Gagnon et al. 2017 and Gagnon et al. 2018)

2.1.2.2 Seasons of Research

Seasons of Research was created for/by researchers engaged in partnership within the Keweenaw Bay Indian Community and Michigan Technological University. The process for creating guidance began in 2018 with the goal to develop an overarching guide to inform expectations for external partnerships with KBIC. This was in response to an increase in external inquiries to KBIC to do research, participate in programs, and/or collaborate in projects. In June 2018, the first guidance draft was shared with the KBIC Tribal Council by staff and faculty from KBIC Natural Resources Department, KBIC Health System, Keweenaw Bay Ojibwa Community College, and Michigan Tech. As we shared the long list of potential partnership considerations, the Tribal Council requested that we 1) conduct a comprehensive review of existing research policies and protocols by other Tribal nations, and 2) share our recommendations for/with KBIC.

To honor the Council's request, we began gathering information on the existing plans and strategies that guide university and Indigenous community's research partnerships. Although we had good intentions to complete this work in a timely manner, the research was frequently sidelined for other priorities. Fortunately, about midway through, a graduate student Emily joined the effort, made the work the foundation of their dissertation, and assumed the lead for completing the exploratory research, as well as its review and synthesis. Seasons of Research is the result of this work, informed by an extensive review of the scholarship and policies (regional, national, and international) pertaining to research guidance, tribal codes, the Indigenous Science Declaration, and the United Nations Declaration on the Rights of Indigenous Peoples. Early on, we decided to refrain from providing a checklist of institutional requirements or anything that resembled formalized institutional policy. Instead, our intentions were to provide holistic guidance and specify support that enriches KBIC efforts to protect and restore land and life.

Ojibwa communities, like the Keweenaw Bay Indian Community, have an important role in protecting and restoring Basin ecosystems, particularly because Ojibwa knowledge and practices have been sustained in the region for millennia. In his testimony to the United States Congress in 2017, former KBIC President Schwartz shared that in the beginning, the Ojibwa "accepted a responsibility to protect and sustain the natural resources that provide for the lifeways of our people" (Schwartz 2017:3). The challenges that Indigenous peoples in North America (Tribal, First, and Métis Nations) are facing are many, including changes in seasonal weather patterns, increase in extreme weather events, habitat degradation, pollution, and toxic contamination, and loss of native plant, fish, and animal relatives (e.g., Wildlife Stewardship Plan 2014:74). These challenges can be exacerbated by Indigenous peoples' limited capacity (e.g., funds and staff) and the ability to attain and retain needed expertise, restricted socio-political recognition and landbase, and the lack of knowledge by others that make decisions that affect their everyday lives. Indigenous peoples must address ongoing threats while simultaneously revitalizing obligations to land and life and recovering and sharing the knowledge needed to do so. University-Indigenous community partnerships, when these relations are genuine and equitable, can begin to ameliorate some of these challenges. Indeed, many Western scientists within the region, and beyond, are requesting and taking direction from local knowledge holders and their observations (e.g., walleye fishing in the Portage Waterway and copper and mercury contamination; Kozich et al. 2020; Kerfoot et al. 2020; Perlinger et al. 2018).

Seasons of Research "with/by/as" the Keweenaw Bay Indian Community draws directly from the scholarship of Māori scholar Nan Wehipeihana who illustrates a model and strategy for Indigenous-led evaluation (2019). In *Increasing Cultural Competence in Support of Indigenous-Led Evaluation*, Wehipeihana describes evaluator positionality and community relations in terms of "to/for/with/by/as," (2019:379). Situated into four (4) quadrants (x-axis as decision-making, y-axis as community impacts), the preposition demonstrates the nature of the relationship *between* the 'evaluator' and the community; *to/for/with/by/as* determines the partnership result as variations of harm (imposed relations) escalating to positive outcomes (autonomous relations) for the partners and the region as a whole. We discovered Wehipeihana's model particularly useful for also understanding positionality and relations within research partnerships. Research *to/for* no longer suffices; research *with/by/as* is the foundation to engage in equitable partnerships.

In short, the purpose for doing research and/or being researchers can be different between/for researchers and Indigenous communities (Absolon 2011; Holifield et al. 2009; Nadasdy 2004; Whyte 2017). It is also true that Indigenous ontologies, often revealed in Indigenous languages, can be incommensurable (Schelly et al. 2021). For example, Anishinaabemowin (the Anishinaabe language) reflects the philosophy of learning that knowledge comes to us from people, but it is also shared with us by characteristics and beings within the land and interactions between life and livelihoods (Noodin 2018; Noodin 2019). In Seasons of Research, partnerships are crucial to our relationships with the lands and waters, and especially for **honoring responsibilities** to all living beings who call this place home and have done so since time immemorial. Partnerships are known as **reciprocal teaching and learning through shared experiences** with one another, an accountability that fosters **progress towards shared goals**. As with all guiding principles, knowledge is living, open to adaptations, revisions, and other modifications that may arise in response to changes in the landscape, the community as well as research partners.

2.2 Foundations for research and being researchers

with/by/as the Keweenaw Bay Indian Community

This section is a review of the scholarship that provides foundations for research and being researchers with/by/as the Keweenaw Bay Indian Community. First, we summarize work that asserts the practice of Indigenous self-determination includes recognizing autonomy as a key component of research with/by/as Indigenous peoples. Next, we draw on a body of scholarship in Indigenous knowledge systems to emphasize the relational nature of knowing as interconnected with cycles of time, place, and being for different Indigenous peoples. The final section explores equity in research partnerships between Indigenous and Western sciences. As a practice often articulated as bridging knowledge systems, we highlight current scholarship that draws attention to the necessary acknowledgement for the plurality and integrity of different ways of knowing.

2.2.1 Practicing self-determination includes research autonomy with/by/as Indigenous peoples.

Research autonomy with/by/as Indigenous peoples is the ability to determine one's own research priorities and approaches, and the capacity to design and practice research in appropriate and meaningful ways. Importantly, Indigenous research autonomy is centered in Indigenous knowledge systems and philosophies, and simultaneously, respects the autonomy of others. Throughout Turtle Island (present-day North America), long before European contact and influence, Indigenous knowledge and philosophy informed systems of governance (Whyte 2017) and diplomacy (Simpson 2017), resulting in thriving, interconnected Indigenous Nations that engaged in regenerative trade, farming, arts, recreation, and ceremony across the landscape (Dunbar-Ortiz 2015, Witgen 2011). Indigenous knowledge systems have and continue to empower Indigenous Nations since time immemorial. Therefore, current and future revitalization of Indigenous self-determination practices asserts autonomy in research.

Indigenous peoples have been the subject of, and subjected to, vast research injustice. Further, inequitable research practices and prejudiced systemic approaches remain in place at the time of this writing. However, many scholars are actively working to redefine research relations with Indigenous peoples. Much of this work originated with Māori scholar Linda Tuhiwai Smith (Ngāti Awa and Ngāti Porou) in 1999, who uprooted and transitioned the conversation at the intersection of research and Indigenous peoples with the publication of *Decolonizing Methodologies* thereby shifting research to center on reclaiming Indigenous histories and futures.

Legacies of research to/for Indigenous communities have almost wholly been extractive and harmful. Often research has been conducted to document what soon would become 'extinct' (Geniusz 2009), to co-opt and/or appropriate knowledge (Harding 2015), or to illustrate Indigenous peoples' deficiencies, what Palawa scholar Maggie Walter describes as *5D data - difference, disparity, disadvantage, dysfunction, and deprivation* (Walter 2016). Walter and other scholars assert the need for Indigenous data sovereignty practices, the "right of Native nations to govern the collection, ownership, and application of its own data" (Rainie et al. 2017:1; Carroll et al. 2020; Wehipeihana 2019). The exclusion of

Indigenous Nations and limitations on Indigenous self-determination, particularly by those who make decisions that affect the everyday lives and livelihoods of Indigenous peoples and environments, continue to perpetuate mistrust in the academic sciences, their researchers, and in particular, the systems by which they work (Liboiron 2021).

Many Indigenous Nations are engaging in work to determine community-centered expectations and priorities for research partnerships that also reflect community values. Collaborations between Tribal Nations and outside researchers are distinct and thus not replicable. We cannot develop universal frameworks to guide such partnerships because knowledge and relationships are situated in unique places and people groups. In 2012, Anna Harding and partner researchers published a material and data-sharing agreement between the Confederated Tribes of the Umatilla Indian Reservation (CTUIR) and Oregon State University (OSU) (2012). To our knowledge, it was the first such agreement between a Tribal Nation and an academic institution. Inspired by Harding, Harper, Stone, O'Neill, Berger, Harris, and Donotuto (2012), and motivated by our desire for transparency and sharing knowledge across Turtle Island and abroad, we have added a few resources prior to and several more since the publication (See Table 2-1). These collections, Harding et al. (2012) and Table 1, informed the process for creating Research Guidance with/by/as the Keweenaw Bay Indian Community.

Table 2-1. Resources for building, strengthening, and sustaining equitable research partnerships with/by/as Indigenous communities. Listed in reverse chronological order, this collection is intended as a continuation from Harding et al. 2012.

Source	Title	Primary Focus and Contributions
Nunavut Research Institute 1988	Nunavut's Scientists Act (1988)	Provides the research application process to receive a license, required to conduct social, health, land, or water-based research in the Nunavut settlement area
AIATSIS (The Australian Institute of Aboriginal and Torres Strait Islander Studies) 2000	Guidelines for Ethical Research in Indigenous Studies	Outlines three principles for ethical research: consultation, negotiation, and mutual understanding, respect, recognition, and involvement, and benefits, outcomes, and agreement https://www.wipo.int/export/sites/www/tk/en/databases/creative_heritage/docs/aiatsis_ethical_research.pdf
Association of Canadian Universities for Northern Studies 2003	Ethical Principles for the conduct of research in the North	Outlines 20 general principles to guide research, such as community consultation, mutual respect, enhancing local benefit, accountability, informed consent, on-going explanations, research summaries in local language, giving credit, and prioritizing greater consideration for risks and cultural value over the contribution to knowledge

		https://acuns.ca/wp-content/uploads/2010/09/EthicsEnglishmarch2003.pdf
Dehcho First Nation 2004	Traditional Knowledge Research Protocol	Assists Deh Cho First Nation in negotiating terms and conditions for the use of Traditional Knowledge in external research studies and industrial development, outlines a policy for Deh Cho to evaluate proposed projects, and articulates researcher steps to follow with Deh Cho https://dehcho.org/docs/traditionalknowledgeprotocol.pdf
Ho-Chunk Nation 2005	Health and Safety Code-Tribal Research Code	Establishes the Nation's research priorities, including the full research application, terms of research, and IRB process to be considered to conduct research with Ho-Chunk Nation https://ho-chunknation.com/wp-content/uploads/2019/10/3HCC3-Tribal-Research-Code-05.05.05.pdf
Inuit Tapiriit Kanatami and Nunavut Research Institute 2007	Negotiating Research Relationships with Inuit Communities: A Guide for Researchers	Provides practical advice for researchers related to relationship-building and communication through the stages of research -- project design, data collection, and analysis – with Inuit Communities https://www.itk.ca/wp-content/uploads/2016/07/Negotitiating-Research-Relationships-Researchers-Guide_0.pdf
Tipene-Matua et al. 2009	Old Ways of Having New Conversations- Basing qualitative research with Tikanga Māori	Formalizes Tikanga Māori (Māori traditions) into rituals of first encounter, as a part of the research process to create a setting that is conducive to *by Māori, for Māori* research http://www.communityresearch.org.nz/wp-content/uploads/formidable/M%C4%81oriProtocols.pdf

The First Nations Information Governance Centre. Ownership, Control, Access and Possession (OCAP™) (webpage) 2010 Last accessed Jan 28 2022	First Nations Principles of OCAP	Ties the First Nations Principles of OCAP (ownership, control, access, and possession of cultural knowledge, data, and information) rights and responsibilities to self-determination, the preservation of histories, and future development https://fnigc.ca/what-we-do/ocap-and-information-governance/
Indigenous Geography (webpage) 2010 Last accessed Mar 16 2022	Research Ethics: A Source Guide to Conducting Research with Indigenous Peoples	A collection of literature and guidelines for conducting research with Indigenous Peoples. http://www.indigenousgeography.net/ethics.shtm
Harding, Harper, Stone, O'Neill, Berger, Harris, and Donatuto 2012	Conducting Research with Tribal Communities: Sovereignty, Ethics, and Data-Sharing Issues	Delineates an institutional-community research agreement that includes considerations for project scope and collaborators, and material and data collection types; outlines potential constraints such as material and data use, data access and security, community risks and benefits, and mutual review processes; and includes a table of partnership research resources https://ehp.niehs.nih.gov/doi/10.1289/ehp.1103904
NCAI Policy Research Center and MSU Center for Native Health Partnerships 2012	Walk Softly and Listen Carefully: Building Research Relationships with Tribal Communities	Contributes five core values for working with Tribes: Indigenous knowledge is valid and valued, culture is a part of research, stewardship includes interpreting and understanding data and research, tribal sovereignty for research and data, and research must benefit Native people https://www.ncai.org/attachments/PolicyPaper_SpMCHTcjxRRjMEjDnPmesENPzjHTwhOlOWxlWOIWdSrykJuQggG_NCAI-WalkSoftly.pdf
Yukon Research Centre 2013	Protocols and Principles for Conducting Research with Yukon First Nations	Describes research best practices aligned with Yukon First Nations interests as five principles and protocols: ethics, accountability, participatory approach, intellectual property rights, and research outcomes https://achh.ca/wp-content/uploads/2018/07/Protocol_YukonFN.pdf

Climate and Traditional Knowledges Workgroup (CTKW) 2014	Guidelines for Considering Traditional Knowledges in Climate Change Initiatives	Explains eight guidelines for considering traditional knowledges (TKs) in climate change initiatives: understand key concepts related to TKs, recognize indigenous peoples right to not participate, understand and communicate risks, establish institutional interface, provide training for agency staff, provide specific directions for ensuring TKs are protected, recognize the role of multiple knowledge systems, develop grant review guidelines that recognize value and are protective. http://climatetkw.wordpress.com/
First Nations Development Institute 2015	Research Policy	Illustrates research protocols that acknowledge and affirm Native nation's rights to control their data, including, informed consent, voluntary participation, data ownership, protection of identity, and the right to review before publication https://www.firstnations.org/wp-content/uploads/2019/04/First_Nations_Research_Policy_2016.pdf
Wilkinson, Dumontier, [...], and Mons 2016	The FAIR Guiding Principles for scientific data management and stewardship	Details FAIR Data Principles (Findable, Accessible, Interoperable, Reusable) for scientific data management and stewardship with focus on the infrastructure of data reusability and increasing the ability of automated data searches https://www.nature.com/articles/sdata201618
CLEAR Lab; Max Liboiron (webpage) 2016 Last accessed Jan 28 2022	CLEAR's guidelines for research with Indigenous groups	Governs the practices of CLEAR (Civic Laboratory for Environmental Action Research) with a commitment to good relations to land and partnerships with Indigenous groups, emphasis on research invitations, knowledge co-creation, and data sovereignty https://civiclaboratory.nl/2016/09/28/guidelines-for-research-with-indigenous-peoples/
Great Lakes Indian Fish and Wildlife Commission 2016	Guidelines for Conducting Traditional Ecological Knowledge Interviews	Specifies protocols for asking Anishinaabe elders and/or knowledge holders to share traditional ecological knowledge stories and expertise in an interview, provides guidelines for handling interview audio and transcripts, and offers a sample interview question structure

		https://glifwc.org/ClimateChange/GLIFWC%20TEK%20Interview%20Guidelines.pdf
Northwest Indian College (NWIC) 2017	Indigenous Research Policy	Informs NWIC faculty, staff, and external researchers how research is conducted, and specifies responsibilities related to cultural grounding, ownership, control, access, and possession of data, informed consent, and gratitude https://www.nwic.edu/wp-content/uploads/2017/03/Indigenous-Research-Policy.pdf
Inuit Tapiriit Kanatami 2018	National Inuit Strategy on Research	Supports the Inuit Nunangat strategic plan for research: advancing Inuit research priorities, enhancing the ethical conduct of research, aligning funding with Inuit research priorities, ensuring Inuit access, ownership, and control over data and information, and building capacity https://www.itk.ca/national-strategy-on-research-launched/
Gentelet, Basile, and Gros-Louis Mchugh 2018	Toolbox of Research Principles in an Aboriginal Context: ethics, respect, fairness, reciprocity, collaboration, and culture	Delineates an Aboriginal Context research toolbox including memorandum of understanding (MOUs), protocols, guidelines, and worldwide open data sources; includes resources https://centredoc.cssspnql.com/cgi-bin/koha/opac-detail.pl?biblionumber=1308&query_desc=kw%2Cwrdl%3A%20toolbox
Wehipeihana 2019	Increasing Cultural Competence in Support of Indigenous-Led Evaluation: A Necessary Step toward Indigenous-Led Evaluation	Illustrates a model and strategy for Indigenous-led evaluation and describes researcher positionality and community relations to/for/with/by/as researchers https://doi.org/10.3138/cjpe.68444
National Park Service (webpage) 2019. Last accessed Jan 28 2022	Tribal Research Policies, Processes, and Protocols	Includes policies, processes, and protocols related to traditional ecological knowledge (TEK) for many Tribal Nations within the United States https://www.nps.gov/subjects/tek/tribal-policies-processes-and-protocols.htm

Memorial University (webpage) 2020 Last accessed Jan 28 2022	Research Impacting Indigenous Groups Policy and Procedures	Specifies policy that requires researchers to seek relationships, involvement, and approval from Indigenous groups prior to the University approval https://www.mun.ca/research/Indigenous/consent.php
Carroll, Garba, Figueroa-Rodríguez, Holbrook, Lovett, Materechera, Parsons, Raseroka, Rodriguez-Lonebear, Rowe, Sara, Walker, Anderson, and Hudson 2020	The CARE Principles for Indigenous Data Governance	Describes CARE principles (Collective Benefit, Authority to Control, Responsibility, and Ethics) for Indigenous data and the collective benefit and autonomy for/of Indigenous peoples, including secondary data; intended to complement the FAIR guiding principles https://www.gida-global.org/care
Whyte 2020	Sciences of Consent: Indigenous Knowledge, Governance Value, and Responsibility	Explains the research context within the Indigenous philosophy of science which prioritizes consent as an ongoing system of governance, responsibility, and accountability
Poitra, Kolonich, Mitchell, Proctor, Baier, and LaPensée 2021	Reciprocal Research: A Guidebook to Centering Community in Partnerships with Indigenous Nations	Supports practices for growing research partnerships, including communicating needs, outcomes, and goals in a consistent, transparent, and respectful manner, ensuring projects are representative of community values and goals, and furthering research with practical applications that are based on community understandings of their own needs; includes five 5 scenarios, reflections, and resources https://www.canr.msu.edu/resources/reciprocal-research-guidebook-partnerships-indigenous-nations
Kitasoo/Xai'xais Stewardship Authority 2021	Informing First Nations Stewardship with Applied Research: key questions to inform an equitably beneficial and engaged research process	Outlines phases for researchers and First Nation stewardship staff to engage in an equitably beneficial research process, including initial engagement, delineating commitments, methods, data analysis and results, and reciprocity and benefits https://klemtu.com/research-guide/
University of British Columbia (webpage) 2021 Last accessed Jan 28 2022	Indigenous Research Methodologies	Serves as a guide to Indigenous methodologies, and provides research resources and examples in practice https://guides.library.ubc.ca/IndigResearch

United States Caucus of the Traditional Ecological Knowledge Task Team Annex 10 Science Subcommittee 2021	Guidance Document on Traditional Ecological Knowledge Pursuant to the Great Lakes Water Quality Agreement	Provides guidance and support for working with Indigenous nations and knowledge holders to aid in the protection of, and respect for, the Great Lakes and their ecosystems as part of the Great Lakes Water Quality Agreement responsibilities https://www.bia.gov/sites/bia.gov/files/assets/bia/wstreg/Guidance_Document_on_TEK_Pursuant_to_the_Great_Lakes_Water_Quality_Agreement.pdf
Gagnon, Ravindran, and Shaw 2021	Guidance for Research Partners with/by/as the Keweenaw Bay Indian Community	Illustrates research as an iterative process of seasons, including relationship building, planning and prioritization, knowledge exchange, and synthesis and analysis; provides partnership guidance in principles of respect, reciprocity, responsibility, and reverence; and shares resources http://nrd.kbic-nsn.gov/sites/default/files/KBIC_Rsch_Guide_TriFold_2021.pdf
Unama'ki College of Cape Breton University (webpage) n.d. Last accessed Aug 31 2021	Mi'kmaw Ethics Watch (MEW)	Specifies application and research proposal review process by the Watch committee to conduct research with and/or among Mi'kmaw people https://www.cbu.ca/indigenous-affairs/mikmaw-ethics-watch/

2.2.2 Indigenous knowledge systems are relational, interconnected with cycles of time, place, and being.

It has been well established that Indigenous knowledge systems are rooted in and exist as dynamic land-based relationships. Like the land, knowledge is interconnected to all beings, climate and seasons, and the practices of the people who inhabit them and have done so since time immemorial. Specifically, knowledge is connected across a specific land base and a peoples' history to it (Basso 1996; Coulthard and Simpson 2016; Geniusz 2009). Scholars have articulated Indigenous knowledge relationships in diverse ways (i.e., Basso 1996; Berkes 1999; Cote 2010; Kawagley 2006). In *Sacred Ecology* (1999), Fikret Berkes describes traditional ecological knowledge "as a knowledge-practice-belief complex" (17), and the theory of grounded normativity asserts that knowledge is generated through people- and place-based practices (Coulthard 2014). Knowledge is a state of being, enacted through belief and value-informed practices such as honorable harvesting, food and medicine practices, sharing stories, and engaging in ceremonies - these practices are pragmatic for maintaining good relations in everyday life (Kimmerer 2021). In *Look to the Mountain: An Ecology of Indigenous Knowledge*, Tewa scholar Gregory Cajete (Santa Clara Pueblo) explains that "... knowledge gained from first-hand experience in the world is transmitted or explored through ritual, ceremony, art, and appropriate technology. Knowledge gained through these vehicles is then used in everyday living. Education, in this context, becomes education for life's sake (Cajete 1994:25)". In this way, education for *life's sake* does not prioritize the discovery of knowledge solely as the material or cognitive collection of information, but instead, embodies knowledge as a set of ethical and applied practices for sustaining life. Knowledge is lived and education is experienced, and as such, new knowledge is being continuously generated and applied (Berkes 1999). Knowledge is flexible and adaptive to place and the people's relationships to all that exists in place, and because it is lived, it becomes shared across generations, creating a community's memory that is passed through generations.

Practicing Anishinaabe knowledge calls for a broad accounting of and respect for a constellation of relationships in which all things and of all time are interrelated and interdependent (Johnston 1976; Kimmerer 2003; Whyte 2017). Anishinaabe knowledge is alive and present within the lands and all other beings (Absolon 2011), including the seasons, the years, the days, and many recurring events (Johnston 2003). However, as the youngest and most pitiful beings, the Earth's land, forests, rivers, and all other beings are humans' teachers (Bell 2013; Cordova 2007). Human pitifulness is borne from our dependence on all others: trees offer themselves so that we can build homes; and wind helps us to pollinate our foods. An illustrative example comes from Anishinaabe Ojibway scholars Martin Reinhardt and Traci Maday in *Interdisciplinary Manual for American Indian Inclusion* (2005:7): "[F]rom an Anishinaabe Ojibway perspective on education, Mother Earth is the original and primary teacher and classroom." They explain that the English word "education" is most closely related to the Ojibwa term *kinomaage* which literally translates to "the Earth, it shows us the way." Anishinaabe and Haudenosaunee scholar Vanessa Watts (2013) emphasizes the intentionality of all beings, not simply human beings, and that the intentions of each being facilitates intimate relationships rooted in specific lands and waters and winds. Humans, through interactions with the land, careful observation and deep-listening, can learn from plants and animals as they share their teachings, their *gifts* (Kimmerer 2016, 2020), with humans and other beings. In *As We Have Always Done*, Anishinaabe scholar Leanne Simpson (2017) describes these relations as *Nishinaabeg internationalism,* emphasizing the diplomacy to be practiced in relations with nations of many kinds, including plant and tree nations, fish nations, and other wildlife nations as well. Potawatomi scholar Kyle Powys Whyte in "What Do Indigenous Knowledges Do for Indigenous Peoples?" explains that Indigenous knowledge systems comprise the governance and identity of Indigenous peoples which contributes to the continuance of Indigenous Nations and the resurgence of their members (2017). In short, the beliefs and values associated with Indigenous knowledge systems cannot be separated from the people who practice them or the lands they are a part of.

In many Indigenous communities, the shared responsibilities of practicing knowledge and restoring relationships are being actively reclaimed (Geniusz 2009; Kimmerer 2016; LaDuke 2005; Todd 2017). In Anishinaabeg communities, the people's long-time, reciprocal obligation with *Gichi Manidoo* (the Creator) and all orders of creation is being articulated as the people's First Treaty. Also known as Sacred Law, the Great Laws of Nature, and the Original Instructions, the First Treaty obligates all created from rock, water, fire, and wind - the physical world of sun, stars, moon and earth, plant beings, animal beings, and human beings - to care for one another and support one another's autonomy (Johnston 1976). Anishinaabe scholar Nicholas Reo (2019) terms these kincentric obligations as *relational accountability* in acknowledging one's more-than-human network of and obligations to relations both within and outside of research partnerships. The guiding principles for sustaining good relationships and being a good relative are, importantly, included in the landmark 2017 Indigenous Science Declaration, which at the time of this writing, has almost two thousand (2,000) signatories by Indigenous scientists and allies (Kimmerer et al. 2017). Originally presented at the March for Science, this letter endorsed collective action (e.g., organized marches and speakers across the U.S.) for science and simultaneously called for recognition of other ways of knowing. These examples, and many others, elucidate that relational and interconnected Indigenous knowledge systems are critical to everyday life and living, and this includes research and being researchers with/by/as Indigenous peoples.

2.2.3 Bridging knowledge systems requires acknowledging the plurality and integrity of different ways of knowing.

To engage in bridging knowledge systems, one must first acknowledge that there are many distinct knowledge systems, including Indigenous sciences (Simpson 2000; Kimmerer, LaPier, Nelson, and Whyte 2017), and recognize that each way of knowing is integral on its own and viewed equally among knowledge systems (Berkes 2017; Cajete 2000). Like others, we use Cree scholar Willie Ermine's (Sturgeon Lake First Nation) "ethical space of[/for] engagement" theoretical framework to illustrate partnership work between Western and Indigenous sciences (2007). We have added "for" engagement to illustrate that we view the concept of "bridging" as more than theory, but also a place of and for practice. Informed by Robert Poole's work on deep subjectivity (1972), Ermine explains that ethical space is "formed when two societies, with disparate worldviews, are poised to engage each other," (193). As Western scientists and Indigenous people may have different research priorities and approaches, it is important to be deliberate about the distinct ethical values that will inform the research practices in partnership with one another.

We have come to use the concept of a bridge to illustrate the space and *place* for ethical engagement. In our research, we envision the bridge as the place(s) for the co-construction of knowledge where everyone brings their own research toolbox. Using the bridge signifies that people must physically come together, bringing their different philosophies, including community members and professionals who work across and from different disciplines and institutions (Gagnon et al. 2016; Wilson 2005). Further, bridging knowledge systems avoids assimilative and supplementary treatment for/of Indigenous knowledge (Reid et al. 2021; Whyte 2017). Also described as "two-eyed seeing" (Bartlett et al. 2012), building and maintaining the bridge requires different intellects, methods and approaches, and various expertise and skill sets (Mantyka-Pringle et al. 2017; Reid et al. 2021). Finally, doing this work together is a process which, over time, can facilitate the development of shared expectations and vocabularies, shared visions and goals, and the creation of ethics in common for joint action and justice (Wilson 2005).

It is true that Western and Indigenous sciences have different philosophies and values, and it is also true that each has unique strengths that can be complementary to one another, which also strengthens knowledge and understanding between differences (Ausubel 2008; Deloria Jr. and Wildcat 2001; Kimmerer 2016; Mantyka-Pringle et al. 2017; Reid et al. 2021; GLWQA TEK 2021). Western science has enhanced our attention and abilities to see, particularly physical materials and strengthening cognitive intellect (Kimmerer 2019; GLWQA TEK 2021). Just think about ways of seeing and what we have learned from the microscope to the telescope, the development of models and modeling complex systems, and the places that various sensor equipment and autonomous vehicles can venture to see and monitor. Indigenous science enhances our attention and abilities to listen, particularly relational and emotional intellect, contributing to our human intuitive ways of knowing (Kimmerer 2019; GLWQA TEK 2021). Think about various ways of listening and what we have learned from other beings, processes, and interactions in natural systems. Both sciences enhance our abilities to learn, but Indigenous science asks us to prioritize listening so that we might be better relatives and support one another’s autonomy.

In summary, it is critical to recognize that the bridge is not a divider, but a pathway. The engagement is not intended to be in opposition of each other but complementary. Engagement activity is not for consultation or confrontation but for interaction and cooperation. Importantly, the purpose is not to integrate knowledge differences but to maintain integrity and distinctness in ways of knowing. And, as a reminder, the ethical space theory must be accompanied by an ethical place in practice.

2.3 Guidance for research partnerships

The Guidance for Research Partnerships with/by/as the Keweenaw Bay Indian Community is an assertion of self-determination and research autonomy to reclaim our history, restore relationships, and revitalize our future. In this section, we describe the shared responsibilities and reciprocal expectations associated with collaborative, participatory and community-engaged research: relationship building, planning and prioritization, knowledge exchange, and synthesis and application. Strengthening research partnerships is a priority because we know that working together contributes to the resiliency of our shared communities, landscapes, and future. As illustrated in (Figure 2-1), we chose the Medicine Wheel as the research guidance model to reflect Anishinaabe Ojibwa understandings and worldview (Johnston 1976). The Medicine Wheel is an interconnected system of teachings and teachers relating to seasons, directions, elements, and the cyclical nature of life (Johnston 2003). Beginning in the East and moving clockwise, it aims for balance between and among time, space, and all beings, a balance that is sustained when we **ask for permission and receive consent** (Whyte 2020).

Informed by the Indigenous Science Declaration (Kimmerer, LaPier, Nelson, and Whyte 2017), respect, reciprocity, responsibility, and reverence illustrate our intentionality as researchers throughout the seasons. Situated deliberately around the Medicine Wheel, these characteristics demonstrate a commitment to respectful, reciprocal, responsible, and reverent interaction and being. Research partnerships with/by/as the Community **demonstrate respect** for each other's differences, **honor reciprocity** for each other’s actions, **exemplify responsibility** for individual, organizational and community commitments, and **express reverence** for shared lands, waters, and all living beings. Respect, reciprocity, responsibility, and reverence guide our thoughts and actions in research partnerships as they do in everyday life.

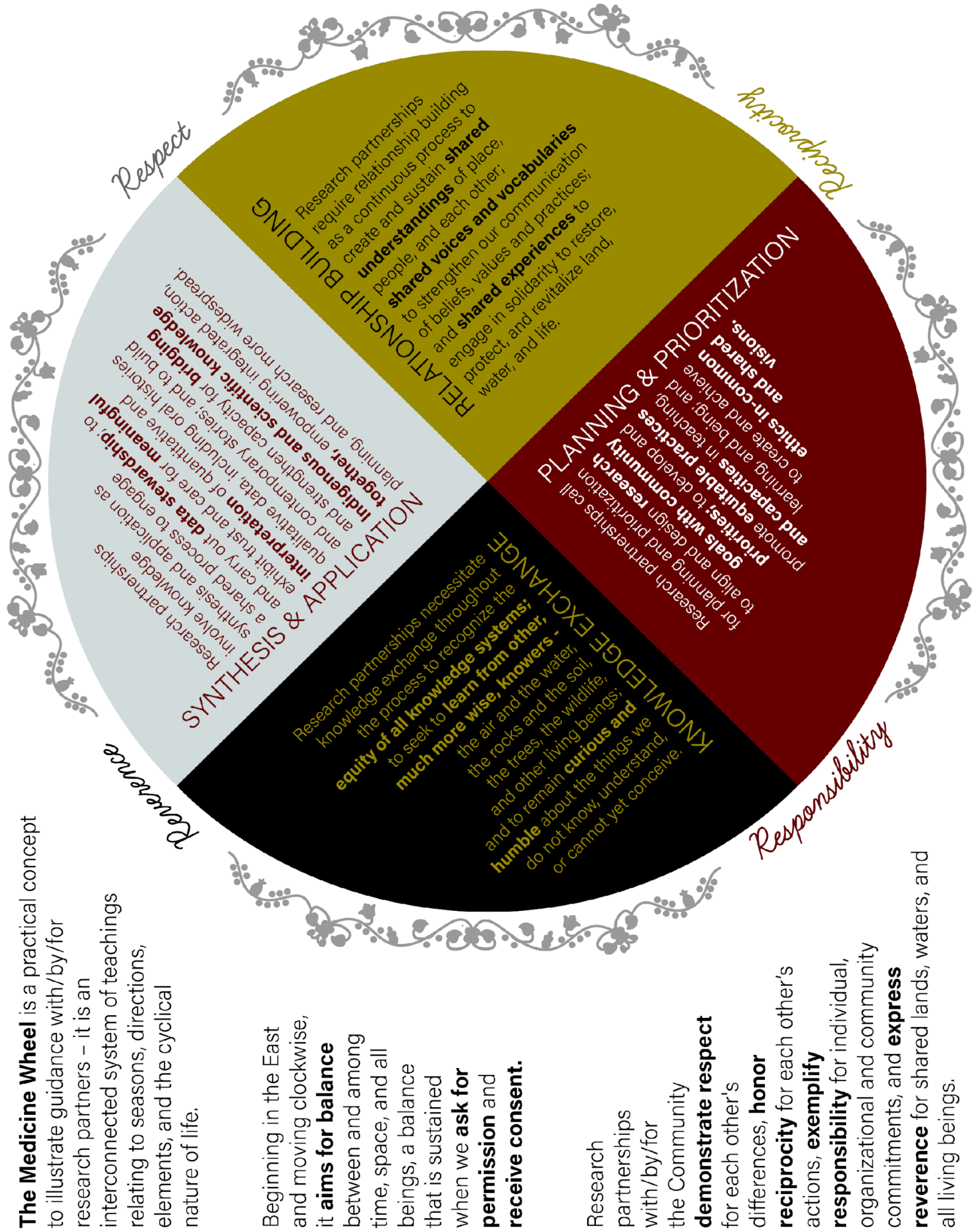

Figure 2-1. This figure illustrates Guidance for Research Partnerships with/by/as the Keweenaw Bay Indian Community. Used with permission from the Keweenaw Bay Indian Community Natural Resources Department.

2.3.1 Relationship building

Research partnerships require relationship building as a continuous process to create and sustain **shared understandings** of place, people, and each other; **shared voices and vocabularies** to strengthen our communication of beliefs, values, and practices; and **shared experiences** to engage in solidarity to restore, protect, and revitalize land, water, and life. Critical to the development of shared understandings is the foundational acknowledgement that colonialism is endemic to the Americas, and that the lived experiences of all places, peoples, and relations are a part of this interconnected story (Brayboy 2006). Partnerships must be built upon the understanding that Indigenous peoples are the original people and knowledge holders of these lands. Our shared voices and vocabularies need to assert, in unison, that Indigenous peoples retain inherent rights and responsibilities, and Indigenous place-based beliefs, values, and practices have sustained relationships across the landscape since time immemorial.

Building relationships in a genuine way necessitates shared experiences that are sustained throughout various stages of the research process. Some shared experiences may or may not directly be related to research, but instead, are directly related to building relationships. This simple truth cannot be overstated. To build relationships, research or otherwise, one must invest actual time and effort with each other. Relationships are work, and as such, require a commitment by those involved to share time together, participating in activities and engaging in dialogue, and in short, to be accountable to one another (Reo 2019). The strength of one's relationships reflects one's commitment, a commitment that does not correspond to a project's end date. Relationship building, we assert, is an on-going practice to grow intellectually and socially, as a partnership and as an individual, in ways that transcend research and inform one's identity and place in the world.

2.3.2 Planning and prioritization

Research partnerships call for planning and prioritization to align and design **research goals with community priorities**; to develop and promote **equitable practices and capacities** in teaching, learning, and being; and to create and achieve **ethics in-common and shared visions**. Critical to aligning research goals with community priorities is the recognition that research is a practice of sovereignty (Tuck 2009). Orienting research goals in this way is to prioritize restoration, revitalization, and protection, ultimately contributing to Indigenous Nations' capacity to rebuild relationships to the land, water, and life. Individual and community healing centers on reconnecting to place and reclaiming the knowledge and practice necessary to do so. Aligning with community priorities requires two very practical activities: do your homework and listen. In most cases, there is an abundance of learning resources that already exist--don't expect community members to teach you what you can learn by doing your homework. Prioritize listening to understand and not listening to respond. Think about and be open to different community understandings of problems and solutions. Remember, it is not the community's responsibility to serve as the site for your research outreach and education plans.

To facilitate equitable practices and capacities, research partnerships must share leadership, decision making authority, and when applicable and available, research funds (Walter

2018). This necessitates early involvement and planning to ensure intentional inclusion rather than addendums to existing plans (Gagnon et al. 2017). Important to equity in research is the understanding that each research partner will be both a teacher and learner (Reid et al. 2020), and that the research plan and budget reflects and supports each partner in both roles. This shift is deliberate, and as such, Indigenous people need not be assigned as research human subjects but rather as human researchers. As a part of the process of enacting equity, the intention is also to realize the ethical values that the researchers have in common which informs the shared visions for the future, the research process and expectations, and importantly, guides interactions with the research foci as well as with one another. Shared ethics are crucial to building the research foundation, co-learning throughout, and strengthening the research relationship (Ermine 2007). Ethics and vision sustain the partners' commitment to achieve research and relationship expectations. Planning and prioritization, we assert, is a process of affirming equity, in research partnerships and across personal and professional boundaries, to rebuild relationships between university and community partners.

2.3.3 Knowledge exchange

Research partnerships necessitate knowledge exchange throughout the process to recognize the **equity of all knowledge systems**; to seek to **learn from other, much wiser, knowers** - the air and the water, the rocks and the soil, the trees, the wildlife, and other living beings; and to remain **curious and humble** about the things we do not know, understand, or cannot yet conceive. To practice equitable knowledge exchange is to first recognize that Western science is simply one system of knowing, albeit young, diverse, and contentious, and as such need not claim to be the source of authority (Harding 2015). Many systems of knowledge are practiced and have been passed through the generations since time immemorial. We must also acknowledge that all human learning originates *from* the world that surrounds us, and that wisdom is not solely a human feature but belongs to many (Kimmerer 2019, 2021). Learning *about* the natural world, and its many features, interactions, and beings, can be argued as a relatively new practice. As researchers, we must periodically ask ourselves what we can learn from the plant and tree beings, from the processes between the air and water, and from climatic shifts and the transformations between seasons. We cannot assume to know; we need to listen--what are their intentions, what is their work, and what accumulated wisdom are they trying to share with us?

Learning from others, particularly from more-than-human beings, requires lifelong curiosity from a position of humility (Deloria Jr. and Wildcat 2001). As humans we experience the world with a set of senses that are particularly confined, and others have gifts that we as humans do not (Kimmerer 2009, 2020). Knowledge exchange, we assert, is an iterative process to refine and transform our understandings of the world and our relational interactions with others, all others, so that we might learn to be better residents and relatives in our communities, societies, and of the Earth.

2.3.4 Synthesis and application

Research partnerships involve knowledge synthesis and application as a shared process to engage and carry out **data stewardship**; to exhibit trust and care for **meaningful**

interpretation of quantitative and qualitative data, including oral histories and contemporary stories; and to build and strengthen capacity for **bridging Indigenous and scientific knowledge together**, empowering integrated action, planning, and research more widespread. Critical to the practice of data stewardship, to being data stewards, is the recognition that Indigenous peoples have and retain procedures and protocols associated with caring for many different forms of knowledge (Whyte 2017). Being good data stewards will not always align with federal regulations, thus it is critical to establish a data stewardship plan from the forefront. Part of this responsibility is to also discern and be respectful of everyday knowledge, belonging to everyone, and guarded or sacred knowledge, belonging to particular knowledge holders (Geniusz 2009). Put simply, some knowledge does not belong to/in research. As a strong word of advice, researchers need not seek out guarded knowledge.

Historically, knowledge and research findings have not only been extracted and misused, but they have also been sorely misrepresented, sowing distrust between academic researchers and Indigenous peoples (Absolon 2011). Thus, shared decision-making authority is crucial throughout the stages of data analysis and synthesis. In data interpretation, emphasis is focused on scientifically sound, legally defensible results, but partnership research must also be culturally sound and defensible. This means that the data interpretation, and its process, must be meaningful to all research partners, including Indigenous partners. Meaningful interpretation strengthens the ethical space, place, and partners of engagement, and when conducted respectfully, contributes to building capacity for partnership research, for bridging Indigenous and Western sciences together, in the future. Meaningful interpretation is the foundation for integrated action and planning. Synthesis and application, we assert, are the practices that determine, and/or ensure, the continuation of seasons in seasons of research.

In borrowing the wisdom of the natural seasonal cycle, this section has shared partnership guidance as seasons of research. In the physical world, and in research partnerships, seasons move forward in time and return us to where we are, building on the histories that have come before. To create a foundation for equity in future generations, each seasonal and cyclical iteration must be bound by thoughts and actions that **demonstrate respect and reciprocity** for diverse ways of knowing and being, and **embody responsibility and reverence** for timeless commitments to land, water, and life (Kirkness and Barnhardt 1991; Kimmerer et al. 2017). Such actions contribute to socio-ecological equity in the real world as well as equity in research partnerships; these practices are inseparable as professionals and as everyday citizens of Earth.

2.4 Practicing seasons of research

Extending from more than a decade-long partnership between Val and Evelyn, this section provides a reflection of my, Emily's, experiences to engage in seasons of research as a doctoral research practice. I demonstrate my seasons of research through a project in progress that seeks to elucidate patterns in mixture toxicity (e.g., toxicity that results from exposure to multiple contaminants simultaneously) of fish tissue to inform food sovereignty work and rebuild relationships with fish. In sharing these experiences through writing, I have come to realize the many ways that research seasons overlap and interconnect. Often, one can be engaged with multiple seasons simultaneously. Overall, practicing seasons of research is a multidirectional commitment and system of accountability to the land and communities of life.

As a foundation, I discovered that my grounding in Indigenous scholarship allowed me to cultivate and center my research on **building relationships** with the land, waters, and beings. As a settler scholar, I had much to unlearn and relearn. Beginning with the literature, I built intellectual relationships with scholars and came to understand that knowledge comes from sacred relationships and their grounded nature means they are non-transferrable (Watts 2013; Kimmerer 2013). I realized that without place-based relationships for myself, I could not transfer others' teachings as my own learning. This motivated me to build relationships with the lands, and KBIC and community members. Moving north from lower Michigan, I recognized many plant and animal relatives, but I did not know their names. Learning names is the first gesture of respect (Kimmerer 2016). So, I ventured to learn who they are. With my plant ID app on my phone and my guidebooks in hand, my joy for walking in the woods transformed into an adventure to know the plants and animals who lived here, too. Simultaneously, in relationship building with the KBIC, I first attended a Food Sovereignty Lunch and Learn in the summer of 2016. That afternoon, I learned about a community-guided research project that centered on the question, *when can we eat the fish?* (ASEP Project 2020). This event was an introduction to my understanding of how fish-human relationships are affected by chemical contamination (Gagnon et al. 2017; Todd 2014, 2017). I began to see 'anthropogenic stressors' differently, shifting in terms of impacts to socio-cultural practices in Ojibwa food systems.

Aligning my research goals with the Community's needs has grown from intentional project **planning and prioritization.** Since the Lunch and Learn, and while I was in my master's program, I was simultaneously building relationships with KBIC partners to establish a foundation for doing a dissertation project in partnership. In doing so, I have participated in a number of community events to build and strengthen my relationships within KBIC: I became a Lake Superior water protector and engaged as a water walker; I have spent hours at the KBIC Debweyendan Indigenous Gardens (DIGs) planting, weeding, listening, and harvesting; I accepted invitations to share research talks at Tribal Water Day and staff meetings, and attended an assortment of other events that aligned with my personal and professional interests. Building relationships is an on-going practice and as such, it includes the expectation to show up, share expertise, and work together. I could not have expected to create and sustain shared understandings, voices, and vocabularies without these shared experiences. And through building relationships, we are also contributing to each other's capacities to do shared work together in the present and future.

Engaging in seasons of relationship building and planning and prioritization helped me to recognize the distinction between 'research informed by' and 'research in partnership with' (Minkler 2005; LaVeaux and Christopher 2009). However, this wasn't always easy; it was an especially complicated process because I was transitioning from being a science 'educator' to being a graduate 'student.' I have long recognized the creative capacity of science and research; for years I had used it as a tool in hopes of inspiring Great Lakes stewardship in others. My experiences in stewardship and outreach served me well, especially for my transition to do indiginist research and to be an indiginist researcher. Indiginist research, as described by Opaskwayak Cree scholar Shawn Wilson and White Settler American scholar Margaret Hughes in *Research and Reconciliation* (2019:7-8), must be interdisciplinary and represents a transformation through accountability and responsibility to Indigenous philosophies and laws. For me, this process has been a transformation of my mindset; it has redefined my understanding of being a scientist.

My official transition to the season of planning and prioritization with KBIC began on a wintry December afternoon a few months after I defended my master's thesis. I drove an hour south to the L'Anse Indian Reservation to share my thesis results with KBIC Natural Resources Department staff in Pequaming. At a cramped conference table, we considered ways we might use Lower Michigan river-fish-PCB results for a dissertation research study related to mixture toxicity. Our dialogue revealed significant overlap in our visions for the future of fish-human relationships. As a fishing community, KBIC's concern about both PCB and mercury accumulations in fish, particularly in inland water bodies frequently harvested by their members, provided the trajectory for dissertation research on mixture toxicity. With their consent to move forward, I could work on a research proposal and return to share more about what I was learning along the way.

Learning to orient my work for the sake of rebuilding relationships rather than protecting resources meant that I had to invest time in learning from Indigenous knowledge holders in the Great Lakes basin. To do this, I volunteered to help organize and facilitate speaker events at my university, and I also attended many seminars by Indigenous speakers. Building a foundation to support and sustain **knowledge exchange** undoubtedly comes from building these genuine relationships. For this reason, in my experiences, the seasons of relationship building and knowledge exchange are deeply interconnected. Early on, these exchanges were intellectually personal and part of a rattling process of unlearning and relearning. Reflecting on the work of Indigenous scholars challenged my existing understandings of science and knowledge. Connecting Indigenous philosophies to my Western science knowledge required that I recognize the necessity for weaving and bridging rather than integrating. I was responsible for unlearning knowledge hierarchies and recognizing that integrating knowledge reinforces them; instead, weaving and bridging maintains their distinct values and priorities.

Recognizing my role as a caretaker, I began to understand knowledge exchange as a system of accountability and reciprocity (Kimmerer 2015; Todd 2017). Critical to caring for our relationships and facilitating equitable knowledge exchange is careful listening. Lingering over coffee and snacks during Tribal Water Day in 2019, I listened to conversations with KBIC staff and commercial and recreational fishermen that helped to expand my understanding of the significance of repairing fish-human relationships. Ogaa (walleye) is not just an important food source. Citizens of the many fish nations sacrifice themselves so that their human relatives have sustenance (Hoover 2013; Todd 2014). For this gift, humans express gratitude to fish by tending to their habitats and harvesting only what they need. Realizing how chemical contamination disrupts many reciprocal exchanges, the gravity of expecting some people to harvest less to avoid contamination struck me. I came to understand chemical contamination as a food sovereignty issue. My responsibility, as a research partner, is to work towards the elimination of fish consumption advisories. At the Debweyendan Indigenous Gardens (DIGs) workdays, I also listen to bees, butterflies, and other pollinators. I noticed their affection for bee balm (*Monarda fistulosa*) and so I planted them near my vegetable garden at home. This year, my zucchini and tomatoes are thriving, returning their gratitude to me! Practicing listening in this way has reconceptualized my understanding of who is an expert; I no longer overlook the wisdom shared by pollinators and bee balm.

Engaging in research partnerships promotes **synthesis and application** in ways that embrace knowledge as everyday practice and a series of relationships that sustain daily life. This brings me to the present moment reflecting and looking inward. Although I have not yet conducted analysis on mixture toxicity, I find myself wrapped within a season of reflexive and action-oriented work. My memory travels to the Guidance review process with the KBIC Tribal Council. Amid a worldwide pandemic, KBIC had a slew of priorities that definitely superseded our research. Sometimes, it felt selfish to take their time and consume their focus and efforts on wordsmithing Seasons of Research. And, on one occasion, some Council members expressed concerns about the lack of explanation on “Consultation” and “consultation,” (See BOR 2012:5-6 for more information about “Big C” and “Little c”). They did not wish to encourage outsiders to use the Guide as an alternative to Consultation with the KBIC. And they especially did not want to open the floodgates for researchers to request KBIC partnerships. In response, we incorporated revisions to address their concerns and ensure the purpose and intention of the guidance reflected KBIC self-determination and sovereignty. The KBIC Tribal Council approved the Guidance for Research Partnerships with/by/as the Keweenaw Bay Indian Community in April 2021.

It is also worth noting that early on, and for an extended time, we called the north season ‘analysis and synthesis.’ While it accurately represents a Western science approach (i.e., a project end date), it conveyed a terminus that was inauthentic to what I/we wanted our partnership to be. Renaming the season ‘synthesis and application’ emphasizes the importance of action; it compels us to act on what we have learned, tying together theory and practice (Brayboy 2006:440). ‘Application’ also seamlessly connects a partnership pathway for another cyclical iteration through the seasons; for our partnership, we will soon begin a seasonal round of research focused on mixture toxicity, continuing our work to rebuild fish-human relationships. In doing so, I aim to embody all the lessons imparted to me in co-creating guidance as seasons of research with/by/as the Keweenaw Bay Indian Community.

Research conducted in these ways changes you - the questions you ask, the answers you hear (and where you hear them), and the conclusions you make, and especially, the actions you do or do not consider or take. I’m brought back to insights shared by Anishinaabe scholar Kathleen E. Absolon (Minogiizhigokwe) in *Kaandossiwin: How We Come to Know* (2011) about research. Absolon describes “re-search,” as “journeys of learning, being, and doing…,” (10). And although I did not fully comprehend initially, I can now realize the transformations she speaks of, of what we/I know, and of who we/I are, as part of an Anishinaabe re-search practice and process. I have come to know, and will continue to grow, re-search as an indiginist researcher.

2.5 A prelude to the future

Seasons of research with/by/as the Keweenaw Bay Indian Community, and the foundational scholars and scholarship it is built upon, is an acknowledgement of a changing landscape in partnership research with/by/as Indigenous peoples. The guidance shared here is a research practice in respect, reciprocity, responsibility, and reverence intended to support, revitalize, and protect the autonomy of others. Research equity is simply one part of a necessary transformation. These changes are also part of a larger, much longer, and more complex context of resistance and survivance by Indigenous peoples (Ketchum 2021; Daigle 2019; Vizenor 2008) as well as other marginalized populations and their allies.

This leads us to close with a hopeful prelude - the current landscape of equitable partnership research, we believe, is an introduction of more significant transformations to come. Like many of you, we have been thinking a lot on the future of bridging knowledge systems in science, governance, and education. As a society, our consciousness is changing. It feels like we are in the midst of moments that are transforming shared beliefs and values, and what we consider to be 'normal.' This is also true in the sciences and research landscape. The concept and application of bridging knowledge systems and expertise, for example, has broadened its reach across groups and institutions - the Wildlife Society (Learn 2020), NASA (Native Skywatchers 2021), and many academic disciplines. Importantly, "bridging" was a primary theme for a conference in May 2021, the International Association for Great Lakes Research (IAGLR) (IAGLR 2021). This was especially significant because in the IAGLR 2020 annual meeting, there was one (1) paper in the last session of the five (5) day conference that was inclusive of Traditional Ecological Knowledge. This year, a session called "bridging knowledge systems" spanned three (3) days of the conference by forty-one (41) participant presenters.

Also of significance, we are witnessing a rise in court cases concerning the legal personhood of ecological systems and other beings, leveraging the precedent set by the legal personhood granted to corporations. The citizens of Ohio are aiming for the implementation of legal personhood for Lake Erie (Chiasson 2019; Daley 2019); Ojibwe citizens are exerting legal efforts on behalf of manoomin (wild rice) rights (LaDuke 2019; Pember 2021); and the legal personhood for Magpie River in Quebec Canada became official in the spring of 2021 (Townsend et al. 2021). This is taking place in other U.S. states (e.g., Florida and California) as well as in other parts of the world (e.g., India, Ecuador, and New Zealand).

We would also like to note a recent American precedent: Debra Haaland, a member of the Pueblo of Laguna, was sworn in as the 54th Secretary of the Department of the Interior on March 18, 2021. This means that in the entire history of the U.S., it has only been a matter of months and days that a descendent of the original peoples, stewards, and knowledge keepers has overseen its lands, species, natural resources, and Indian peoples. More importantly, at the time of this writing, less than one year has passed since an Indigenous person is leading the Cabinet agency that oversees the government-to-government relations between American Indian Nations and the United States. Also, in early September 2021, Bryan Newland (Ojibwe), a Bay Mills Indian Community citizen, joined Haaland as the Assistant Secretary for Indian Affairs, Department of Interior (DOI 2021). Although these precedents give us great cause to celebrate, it also gives us great pause of the tremendous tasks that remain in front of us. We have much work to do.

These, and others, are the transformations taking place that will open new doors and provide new pathways for novel inquiries and approaches concerning equitable partnerships with/by/as Indigenous communities. Seasons of research guidance offers an opportunity to build and sustain a new era of discoveries in research as well as for life's sake.

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3 Combined toxicity – a framework for quantifying risk from exposure to contaminant mixtures in fish

3.1 Introduction

Toxicity from exposure to chemical mixtures has long been recognized as a concern (e.g., Rall, 1974); defining terms and building consensus on what are and are not reasonable assumptions continues to be complicated. For many decades the U.S. Environmental Protection Agency (USEPA) guidelines pertaining to chemical mixtures have attempted to provide unifying focus areas and terminology, but these efforts are thought to have been largely ineffective (Sprinkle & Payne-Sturges, 2021). Here, mixture toxicity refers to chemical mixtures with toxic effects on the same physiological system (e.g., the reproductive system) while combined toxicity refers to chemical mixtures with toxic effects that act on separate physiological systems. *Adverse outcome pathways* (AOPs; Ankley et al. 2010) require an initiating molecular event but not a clearly defined toxic pathway. These reasonable pathways towards toxic effects offer a useful framework for expanding toxic risk to consider mixture and combined toxicity in new ways.

Despite a lack of consensus, frameworks for mixture and combined toxicity have been established and both are recognized as important considerations in cumulative risk assessments. In science and in policy, there is a general acceptance of the hazards of particular environmental chemical mixtures; examples include cigarettes (Borgerding & Klus, 2005), PM2.5 (National Ambient Air Quality Standards, NAAQS), and toxic equivalencies (TEQs) for dioxins, furans, and dioxin-like (DL) PCBs (van den Berg et al., 2006). Both cigarettes and PM2.5 are studied and regulated with consideration for their combined effects (e.g., Li, 2016). Toxic equivalencies (TEQs) for dioxins, furans, and DL-PCBs consider mixture toxicity; that is, the shared aryl hydrocarbon receptor effects. This work expands the chemical mixture hazard list by including exposure to persistent legacy compounds via fish consumption.

Recognized as trans-boundary contaminants (Perlinger et al., 2016) that are toxic to humans, persistent legacy contaminants, specifically, polychlorinated dibenzodioxins (dioxins; PCDDs), polychlorinated dibenzofurans (furans; PCDFs), polychlorinated biphenyl (PCB) compounds, and mercury (Hg)), are the focus of this study. Regulated under multiple international agreements based on protecting human health and the environment (e.g., Basel Convention, Rotterdam Convention, Stockholm Convention, and Minamata Convention), these five compound classes have environmental distributions all of which are tied to human activity. Globally, PCBs, PCDDs, PCDFs, and Hg are ubiquitous with elevated concentrations in remote areas including Antarctica and the artic as well as Africa where PCB usage was quite low (Friedman & Selin, 2016; Gioia et al., 2014; Montone et al., 2003). Environmental fate and transport of these compounds are controlled by physico-chemical properties (i.e., K_{ow}) that result in latitudinal patterns of volatilization and deposition that leave temperate (e.g., the Great Lakes basin) and arctic regions especially vulnerable (Wania & MacKay, 1996). Heavily forested and/or wetland-abundant areas have an increased capacity to transform deposited inorganic mercury (Hg) into methylmercury (MeHg), the toxic form (Perlinger et al., 2018). Despite decades of study and remediation, environmental concentrations of these individual contaminants, particularly in fish tissue, continue to be sufficient to cause harm. Better understandings of the hazards of legacy organic pollutants and mercury might improve implementation of international regulations (e.g., the Stockholm Convention; Wang et al., 2022).

Exposure to chemical contamination occurs via four pathways – dermal contact, inhalation, placental transfer, and ingestion. Environmental and social conditions can strongly influence the particulars of chemical exposure and pathways. For example, regions with large wetland and/or forested areas can have higher methylmercury concentrations (Depew et al., 2013). Social conditions that result in low-income households living in older parts of cities may also result in higher exposure to lead because soil lead concentrations tend to be elevated in older parts of cities, relative to newer areas, because of previously acceptable uses of lead. Humans, especially children, and other animals can be exposed by consuming contaminated soils or by inhaling resuspended lead (e.g., Laidlaw et al., 2012; Mielkel & Reagan, 1998). Finally, legacy contaminants such as those regulated under the Stockholm Convention (e.g., PCBs, dioxins, furans, and Hg) are abundant in fish tissue and lake sediments. Exposure occurs primarily through fish or shellfish consumption, which places fish-reliant populations with high consumption rates at a greater health risk.

Here, we refer to combined toxicity as the dissimilar toxic effects (i.e., toxic effects appearing in different physiological systems) that result from exposure to a chemical mixture. This is exemplified in this article by Framework 2 which sums the risks for all chemicals in a mixture based on the most sensitive human health endpoint known for each chemical even if those endpoints involve different physiological systems. In contrast, mixture toxicity refers to similar (within a single physiological system) effects from exposure to a chemical mixture (e.g., all harmful effects on the reproductive system). These definitions differ from previous usage of mixture and combined toxicity as synonymous. This definition does not require a common molecular pathway (e.g., Ah receptor) for either mixture or combined toxicity. Recent literature shows an expansion of existing toxicity frameworks and priorities (e.g., Ankley et al. 2010, LaLone et al. 2017); we posit that this expansion seems not only well-founded but also represents more realistically the hazards associated with exposure to chemical mixtures. In the proof of concept presented in this paper, we consider a simplified exposure scenario of an environmentally relevant chemical mixture. We focus on a single exposure pathway (ingestion) of a single food source (fish) that represents the primary exposure pathway for these contaminants.

There are five lines of evidence that we used to justify aggregating dissimilar outcomes to represent a combined toxic risk that is distinctly different from mixture toxic risk. First, a combined toxicity framework acknowledges that the human body is a collection of interconnected systems. Multiple organs and systems are connected through genes, proteins, and other macro-molecules and are known as body axes (e.g., the Hypothalamic-Pituitary-Gonadal axis and the Gut-Brain axis). Molecular connections act between body systems, and these connections increase the likelihood that dissimilar effects will co-occur (i.e., that more than one physiological system will be impacted). For example, the aryl hydrocarbon (Ah) receptor is an important immune regulator as well as a participant in the gut-brain axis (Barroso et al., 2020; Rothhammer & Quintana, 2019); thus, dioxins, furans and DL-PCBs have immune-system effects. Hg and non-dioxin-like (NDL) PCBs are recognized as neurologic toxicants (Klocke & Lein, 2020; Sitarek & Gralewicz, 2009).

Second, even assuming a common molecular pathway, dissimilar effects still occur. For example, NDL-PCBs have been shown to disrupt dopaminergic and GABAergic signaling in zebrafish (Brun et al., 2021). As neurotransmitters, dopamine and GABA are clinically important in a variety of diseases (e.g., Parkinson, schizophrenia, autism, depression, seizure disorders, etc.). Toxicological testing often considers the functions of biomolecules and hormones within narrow bounds and, therefore, does not fully identify all pathways of toxic outcomes.

Third, the adverse outcome pathway (AOP) framework de-emphasizes the necessity of delineating a complete toxic pathway (Ankley et al., 2010). Following exposure to a toxicant there is some molecular initiating event that leads to organ, organism, and population effects. Previous work has applied the AOP framework to demonstrate many pathways leading to the same toxic effect (e.g., bee colony collapse; LaLone et al., 2017). Similarly, we expand this framework to include multiple compounds that do not share a molecular event but do cause either similar or dissimiliar toxic effects at the organism level.

Fourth, cumulative risk is clearly defined as "the combined risks from aggregate exposures to multiple agents or stressors" (p. xvii, U.S. EPA, 2003), but frameworks that quantify risks associated with exposure to chemical mixtures have not been established. Both policy and science contribute to the on-going problem of ill-defined toxicity frameworks (Sprinkle & Payne-Sturges, 2021). This practice results in humans continuing to be exposed to, for example, aggregated air pollution mixtures that are unsafe (Younes et al., 2021), and the acceptance of 10,000s of cancer cases by agency ignorance of cumulative risk with respect to drinking water (Stoiber et al., 2019).

Finally, hazard quotients and indices are an accepted framework for quantifying hazards associated with exposure to a particular compound at a particular concentration (Gandhi et al., 2017; Pico et al., 2019; Srivastava et al., 2014), whereas in reality, chemicals often occur as mixtures in a range of differing concentrations. Additive toxicity is accepted as the most probable outcome of exposure (EC, 2012; Cedergreen 2014; Martin et al. 2021). The U.S. EPA also advocates for additive hazard quotients unless there is strong evidence to the contrary (USEPA, 2000). By extending existing frameworks and conceptualizations for the toxicity of chemical mixtures, this work embodies the precautionary principle by more realistically representing the chemical risk associated with fish consumption.

Fish consumption is the primary exposure pathway for the contaminants considered in this study, and thus people and communities that rely on fish – those unable and/or unwilling to limit consumption – are at a greater risk. For the Anishinaabe Ojibwa peoples in the Great Lakes basin (e.g., the Keweenaw Bay Indian Community Lake Superior Band of Chippewa; KBIC) fish consumption is particularly important because they are a fishing community with generations of knowledge tied to practices of harvesting and consuming fish. Such practices build and maintain relationships with the land (Nankervis & Hindelang, 2014). These place-based knowledge systems are reciprocal and non-transferrable and can be understood as *Anishinaabe gikendassowin* (Anishinaabe knowledge; Cajete, 1994; Geniusz, 2009; Watts, 2013; Simpson, 2017).

Regulatory agencies worldwide have established reference doses (RfDs) to protect humans from the toxic effects that can result from exposure to chemical compounds. For legacy compounds RfDs exist for multiple body systems, but do not exist for all systems (Table 3-S1). Some examples include, PCBs affect the immune system (MDCH, 2012), reproductive system (EFSA, 2015; JECFA, 2016), and the liver (JECFA, 2016); dioxins and furans affect the reproductive system (EFSA 2015); and mercury affects the reproductive system (EFSA 2015) and the excretory system (Office of Science and Technology, 2001). Current RfDs were revised from previous RfDs, which does demonstrate an existing willingness and ability to redefine acceptable effects.

Responding to a recent call for more theoretical and creative toxicity work (Martin et al. 2021), we consider combined and mixture toxicity from a theoretical perspective, discussing our models within this theoretical context. We quantify mixture and combined toxicity for humans based on simultaneous exposure to PCBs, dioxins, furans, and Hg via fish consumption. The primary hypothesis of this work is that combined toxicity more accurately represents risk than does individual contaminant or mixture toxicity. Our research questions ask, *Why should we quantify the mixture and combined toxicity of exposure to PCBs, dioxins, furans, and mercury?*, *What is the variation in relative and absolute contributions to combined toxicity between fish composite types and across ecoregions?* and, *What constitutes an effective strategy for quantifying mixture and combined toxicity of chemical mixtures?* Understanding the nuances of toxicity defined by different frameworks will advance the science of mixture toxicity by providing a comparison of toxicity frameworks for legacy contaminants.

3.2 Methods

As part of an on-going partnership with the Keweenaw Bay Indian Community Lake Superior Band of Chippewa Indians (KBIC), this work is guided by a Tribal Council approved *Guidance for Research Partnerships* that, "describes responsibilities and expectations associated with collaborative, participatory, and community-engaged research" (Gagnon et al., 2021; Shaw et al., 2022). With various formal and informal processes for obtaining KBIC consent for partnership research, the type of consent is dependent upon the kind of research and KBIC department(s) with whom the researchers aim to partner. Consent for this research was requested by Emily Shaw, following two years of relationship building, and granted by KBIC in a discussion with staff from the KBIC's Natural Resources Department. Only then, with ongoing KBIC guidance and direction, was the project proposal written, defended, and research begun. Other university-affiliated authors have more than 30 years of research partnership experience with KBIC. We present these research methods to reiterate the non-negotiable status of consent and permission as a part of Indigenous-university research partnerships (Liboiron, 2021; Whyte, 2020).

More details about the National Lake Fish Tissue Survey (NLFTS) can be found in previous publications (Olsen et al., 2009; Stahl et al., 2009) but pertinent details are summarized here. Fish samples were collected from 500 waterbodies between 1999-2002. Fish samples were classified as either bottom-dweller or predator; five fish were included in each composite. Fish composites (i.e., bottom-dweller or predator) included fish that were the same species and of similar size. Length and weight were measured for individual fish and percent lipids was quantified for each composite. Here we calculated the average length and weight for each composite. The QA/QC procedures included duplicates, but these were removed for this analysis (n = 122) leaving 884 samples for analysis. Five samples were removed because they did not have information about the fish species, length, lipid, or waterbody name. Samples that were missing weight data (n = 2) or lipids data (n = 15) were not excluded.

This work reclassified lakes as either lake or reservoir due to limited differences between the original classification of other water body types (Leanne Stahl, April 2021, personal communication). Originally, samples were collected from waterbodies identified and classified by the U.S. EPA's River Reach File Version 3 (Robert Horn & Grayman, 1993). Twenty-three and twenty-four waterbodies were classified as other and NA, respectively. These 47 water bodies were re-classified based on the site name. For example, Yellow Flowage was classified as a reservoir.

While the original study quantified 268 persistent, bioaccumulative, and toxic chemicals (Stahl et al. 2009), this work focused on five compound classes: polychlorinated biphenyl compounds (PCBs, both dioxin-like (DL) and non-dioxin-like (NDL)), dioxins, furans, and mercury (Hg). PCB concentrations were quantified by isotope dilution high-resolution gas chromatography/mass spectrometry (Method 1668, revision A). Dioxins and furans were quantified by isotope dilution high-resolution GC/MS (Method 1613, revision B). Mercury concentrations were quantified by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry (Method 1631, revision B).

All concentrations were considered valid; any samples that were below the detection limit were considered zero. Hg and NDL-PCBs were detected in 100% of fish samples. Dioxins and furans detections were sparser; 230 fish composites without any dioxins (215 predator samples (44%) and 15 bottom samples (4%)). There were 139 fish composites without any furans (125 predators (26%) and 14 bottom dwellers (4%)). For DL-PCBs only one predator sample did not have any detectable DL-PCBs, and we assumed that this will not affect the results. Because of the large percentage of non-detects, the assumption of zero for these concentrations may have affected the results for dioxins and furans. However, the conservative assumption that many zero concentrations exist is realistic since these compounds are less abundant compared to Hg and PCBs.

Statistical analyses and calculations were conducted using R statistical software (R Development Core Team 2007). While length, weight, lipid, and hazard indices (HI) were approximately log-normally distributed, we used non-parametric statistics to account for skewness and minor irregularities. Finally, ArcMap 10.8.1 was used for mapping. Data were projected into NAD_1983_2011_Contiguous_USA_Albers with nutrient regions aggregated from U.S. EPA (USEPA, n.d.). Watershed data, including land use and land cover data, for each sample lake was obtained from LakeCat (Hill et al. 2018). Lakes were matched based on a shared identifier. Distance to urban area was calculated in ArcMap and any sample location within an urban area was assigned a distance of 1 meter.

Three toxicity frameworks were used to evaluate combined and mixture toxicity for five compound classes. Each framework aggregated reference doses (RfDs) for each compound class based on a specific criterion. Framework 1 used the most recently available RfD considering different endpoints; thus, this framework is for combined toxicity. Framework 2 used the most sensitive RfD considering different endpoints for each of the compound classes and also represents combined toxicity. Framework 3, representing mixture toxicity, used a shared reproductive RfD. Despite Framework 3 being classified as mixture toxicity, these associated reproductive endpoints do not consider the same endpoint; the RfD for NDL-PCBs is protective of decreased birth weight; DL-PCBs, PCDDs, and PCDF RfDs protect against decreased sperm count and pre- and post-natal effects; the Hg RfD protects fetal neurodevelopment.

To quantify toxicity (i.e., toxic risk), measured fish tissue concentrations were normalized to threshold concentrations to establish unitless descriptors of toxicity called hazard quotients (HQs; Equation 3-1). Concentrations of PCDDs, PCDFs, and DL-PCBs were converted to toxic equivalencies (van den Berg et. al., 2006). Summed together, the individual HQs create a hazard index to represent mixture or combined toxicity based on concentration addition (i.e., additive toxicity; Equation 3-2). The threshold concentrations were calculated in Equation 3-3 based on reference doses (RfDs) for specific toxic endpoints (Table 3-1), assumed adult human body weight (70 kg), and the EPA-defined subsistence fish consumption rate (143.3 g/day; Equation 3-3; USEPA, 2011).

$$hazard\ quotient\ (HQ) = \frac{measured\ concentration}{threshold\ concentration} \quad \text{Equation [3-1]}$$

$$hazard\ index\ (HI) = \sum HQ \quad \text{Equation [3-2]}$$

$$threshold\ concentration = \frac{RfD \times human\ body\ weight}{fish\ consumption\ rate} \quad \text{Equation [3-3]}$$

Table 3-1. References doses (RfDs) used to calculate the threshold concentrations for the three mixture toxicity frameworks considered in this work. Additional RfDs are available in Table 3-S1.

Compound Class	Framework 1 Most Recent RfD (combined toxicity)	Framework 2 Most Sensitive RfD (combined toxicity)	Framework 3 Shared Reprod. RfD (mixture toxicity)
Non-dioxin-like PCBs (NDL PCBs)	7 ug/kg-day liver toxicity (WHO 2016)	0.02 ug/kg-day immunologic toxicity (MDCH 2012)	0.07 ug/kg-day reprod. toxicity (MDCH 2012)
Dioxin-like PCBs (DL PCBs)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reprod. toxicity (EFSA 2018)
Polychlorinated dibenzodioxins (PCDDs)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reprod. toxicity (EFSA 2018)
Polychlorinated dibenzofurans (PCDFs)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reproductive toxicity (EFSA 2018)	2.86E-7 ug TEQ/kg-day reprod. toxicity (EFSA 2018)
Mercury (MeHg)	0.19 ug/kg-day reproductive toxicity (EFSA 2015)	0.05 ug/kg-day urinary toxicity (USEPA - OoW 2001)	0.19 ug/kg-day reprod. toxicity (EFSA 2015)

3.3 Results

In general, results across the three toxicity frameworks are similar. For all five compound classes there are significant differences between the fish composites; bottom-dwellers have greater toxicity from the hydrophobic compounds (dioxins, furans, dioxin-like PCBs, and non-dioxin-like PCBs) while predators have greater toxicity from mercury ($P < 0.001$; **Error! Reference source not found.** and **Error! Reference source not found.**). Regional differences exist for individual and combined toxicity for both fish composites ($P < 0.001$). Bottom-dwellers were significantly longer, heavier, and fattier compared to predator samples ($P < 0.001$; Table 3-3), leading to higher contaminant concentrations and higher HQs and HIs. Summary statistics and cumulative density curves for contaminant concentrations are available elsewhere (Stahl et al. 2009). Given regional variation in growth rates and ecosystems, we evaluated these differences, for both types of fish composites, among the EPA's nutrient regions. For predator samples, there were no differences between length and weight among the nutrient regions ($P \geq 0.10$), but there were significant differences in lipid content ($P = 6.21 \times 10^{-13}$). For bottom-dwellers there were significant differences among the regions for all three fish characteristics ($P < 0.001$).

For Framework 1, a combined toxicity framework that considers toxic effects based on the most recent reference dose (RfD), 819 samples (93%) exceed the threshold for safe fish consumption by subsistence fish consumers. For bottom-dwellers, 369 samples (94%) and 450 predator samples (93%) had HI > 1. For each compound class, there are significant differences in HQs between the two types of fish composites (bottom-dwellers and predators; $P < 0.001$;Table 3-S2). For predators, toxicity (HQs) for all five compound classes varies among nutrient regions ($P < 0.001$) as does combined toxicity (HI, $P = 1.08 \times 10^{-6}$). For bottom-dwellers, toxicity (HQs) for all five compound classes varies among nutrient regions ($P < 0.001$) as does combined toxicity (HI, $P = 1.34 \times 10^{-12}$).

For Framework 2, a combined toxicity framework that considers the most sensitive toxic effects, 875 samples (99%) exceeded the threshold for safe fish consumption by subsistence fish consumers. For bottom-dwellers, 391 samples (99%) and 484 predator samples (100%) had HI > 1. For each compound class HQ there are significant differences between the two types fish composites ($P < 0.001$; **Error! Reference source not found.**). However, there is no statistical difference between the total combined toxicity HI for the two fish categories ($P = 0.73$). For predators, toxicity for all five compound classes varies among nutrient regions (HQs, $P < 0.001$) as does combined toxicity (HI, $P = 2.70 \times 10^{-8}$). For bottom-dwellers, toxicity for all five compound classes varies among nutrient regions (HQ, $P < 0.001$) as does combined toxicity (HI, $P = 7.75 \times 10^{-11}$).

For Framework 3, a mixture toxicity framework that considered a common reproductive health endpoint, 823 samples (94%) exceeded the threshold for safe fish consumption by subsistence consumers. For bottom-dwellers, 371 samples (94%) and 452 predator samples (93%) had HI > 1. For each compound class HQ, there are significant differences between the two types of fish composites ($P < 0.001$;Table 3-S3). For predators, hazard quotients for all five composite classes vary among nutrient regions ($P < 0.001$), and combined toxicity also varies ($P = 8.87 \times 10^{-7}$). For bottom-dwellers, toxicity for all five composite classes varies among nutrient regions ($P < 0.001$), and combined toxicity also varies among regions ($P = 1.49 \times 10^{-12}$).

Table 3-2. Median values for toxicity (HQ for compound classes, HI for combined toxicity) based on the most sensitive endpoint (i.e., combined toxicity). Differences between the fish composites were evaluated using Mann-Whitney non-parametric t-tests and P-values are reported for each comparison.

	NDL PCB Tox.	DL PCB Tox.	Dioxin Tox.	Furan Tox.	Mercury Tox.	Comb. Tox.

Predator Samples (N = 484)	0.31	0.07	0.007	0.05	9.93	12.31
Bottom-dweller Samples (N = 393)	1.67	0.61	2.11	0.54	2.98	12.29
P-value	2.2 E-16	2.2 E-16	2.2 E-16	2.2 E-16	2.2 E-16	0.73

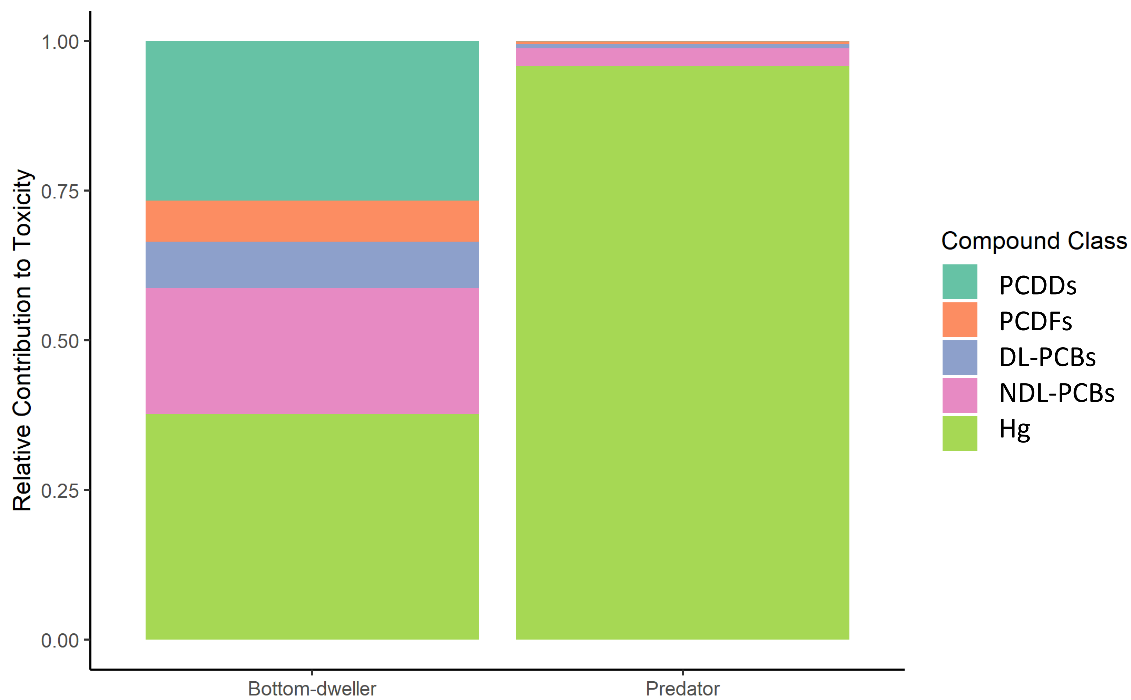

Figure 3-1. Relative toxic contributions for Framework 2 which is based on the most sensitive endpoint. Fractions are calculated based on median hazard indices (HIs), the sum of the hazard quotients of each of the five compound classes. Data used here are from the U.S. Environmental Protection Agency (USEPA)'s National Lake Fish Tissue Survey (NLFTS; 1998 - 2000).

Table 3-3. Median values for length, weight, and lipid for predator and bottom-dweller samples. Differences between the fish composites were evaluated using Mann-Whitney non-parametric t-tests and P-values are reported for each comparison.

	Length (cm)	Weight (g)	Lipid (%)
Predator Samples (N = 484)	370	583	0.77
Bottom-dweller Samples	434	938	5.12

(N = 393)			
P-value	< 2.2 E-16	< 2.2 E-16	< 2.2 E-16

Sample locations were projected into a national coordinate system to evaluate spatial patterns. Visual inspection of the hazard indices (HIs) indicated that for predator samples, all samples east of the Mississippi River exceeded one. While many samples west of the Mississippi River exceeded one (99%), there were two samples with HIs < 1. For bottom dweller samples, there were three samples with HI < 1; one sample was located east of the Mississippi River and two samples were located west of the Mississippi River.

Hazard indices were summarized for each fish composite in each nutrient region and demonstrate regional differences in the toxic contributions from individual compound classes (P < 0.001; Kruskal Wallis). For predator samples, mercury is the dominant contaminant (>50%) across the entire contiguous U.S. except for the Glaciated Dairy Region, which includes parts of Minnesota, Wisconsin, and Michigan, and in the Texas-Louisiana Coastal and Mississippi Alluvial Plains, which extend north into parts of Arkansas and Missouri (Figure 3-2). In both latter regions, mercury contributes only approximately 25% of toxicity. Rather than lower mercury concentrations, this difference is likely due to very high dioxin-like PCB concentrations. For example, in Michigan, Torch Lake is located within the Glaciated Dairy Region and has very high dioxin-like PCB HQs relative to neighboring lakes. Dioxin-like PCBs dominate toxicity in the Dairy Region and dioxins are the most abundant contaminant in the Coastal and Alluvial Plains. For bottom-dwellers, the toxic contribution is less uniform among nutrient regions, but in combination dioxins and dioxin-like PCBs consistently contribute more than 50% of toxicity (Figure 3-S6). Mercury makes a relatively minor contribution in most regions but is more abundant in the Xeric West and Western Forested Mountains regions.

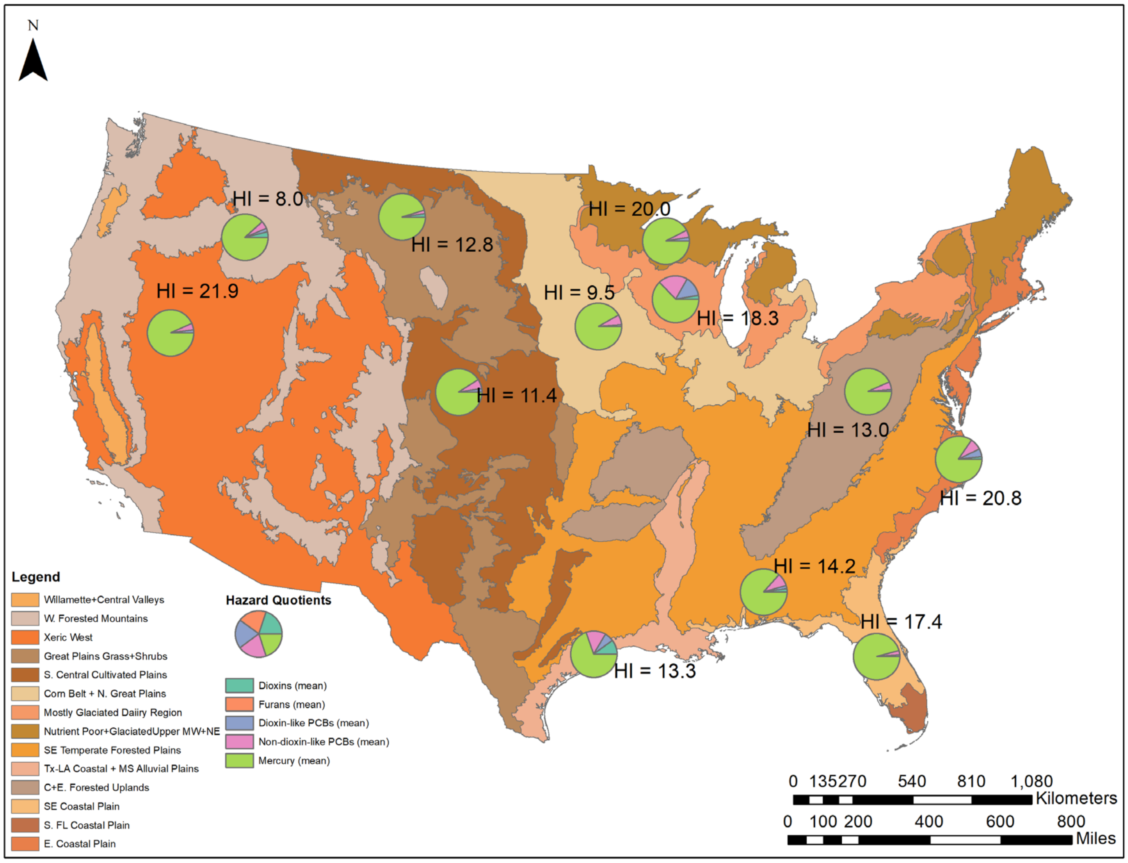

Figure 3-2. Mean hazard indices (HIs) of predator samples are summarized for EPA-defined nutrient regions. For predator samples, Hg contributes most toxicity across nutrient regions except for two regions where dioxins and dioxin-like PCBs are more important. Data used here are from the U.S. Environmental Protection Agency (USEPA)'s National Lake Fish Tissue Survey (NLFTS; 1998 - 2000).

3.4 Discussion

Establishing fish consumption guidelines based on individual contaminant concentrations ignores the reality of exposure to chemical mixtures and is not aligned with accepted frameworks for cumulative risk. As an alternative, we propose quantifying combined toxicity based on the most sensitive endpoint for each component in the chemical mixture according to Framework 2. Such an approach is more protective of human health and demonstrates the magnitude of contaminant reductions necessary to reduce combined toxicity (i.e., hazard indices) according to Equation 3.

Our results support adopting combined toxicity protective of the most sensitive endpoint as the alternative framework in three ways. First, more than 90% of fish samples exceed the safe threshold (HI = 1) for all three frameworks. Mercury is the most abundant contaminant in predator samples and about 40% of predator samples are mis-identified as safe based on mercury concentrations ($HQ_{Hg} < 1$ and $HI \geq 1$). For bottom-dwellers, PCBs and TEQs are more abundant; more than 90% of samples are mis-identified as safe based on PCB concentrations ($HQ_{PCB} < 1$ and $HI \geq 1$). With respect to TEQs, 66-97% of bottom-dwellers were mis-identified as safe. Second, establishing combined toxicity guidelines requires a nuanced approach because the relative importance of each compound class varies among the three toxicity frameworks considered here. Third, combined toxicity more accurately represents cumulative risk from fish consumption and enables individuals to make better informed decisions. For predators, most fish (99%) with $HQ_{mercury} > 1$ also have $HI > 1$; however, fish consumption guidelines (FCGs) based only on mercury concentrations are underestimating cumulative risk and preclude informed decision making. Lastly, regional differences between magnitude and composition of mixture and combined toxicity likely reflect either regional emission patterns or landscape factors that influence the transport and transformation of contaminants. Such variation emphasizes the need for localized strategies for describing and defining allowable toxicity and managing exposure to contaminants.

3.4.1 Why is combined toxicity a more appropriate representation of risk?

To better protect human health and the environment from the adverse effects due to toxic exposure, toxicity frameworks should be redefined. What constitutes an acceptable level of risk needs to be thoughtfully, and analytically, reconsidered. Within the reality of chemical mixtures, sole consideration by regulatory agencies of individual contaminants and their individual effects – on the environment and on people – ignores the combined effects of exposure and passes this ignorance on to others. Cumulative risk, that is, the aggregated risks from many stressors, has evolved as an important component of environmental risk assessments (Goldstein & Goldstein, 2002; EPA, 2003) and is an important consideration for environmental justice. Despite decades of acceptance of the cumulative risk framework, its application continues to be hindered by policies and management decisions related to chemical mixtures (Sprinkle & Payne-Sturges, 2021).

Societal values attributed to the environment (e.g., water and fish) are necessary considerations for defining acceptable levels of contamination. An important question becomes whose values are being considered? What is valued, and in what ways, determines the acceptable levels of risk. Indigenous scholars have long articulated values in terms of a collection of practices and relationships that are specific to place (Cajete, 1994; Simpson, 2017; Watts, 2013). For many Tribes (e.g., the Keweenaw Bay Indian Community), harvesting and consuming traditional foods are valued practices that are shared across generations (Nankervis & Hindelang, 2014). Subsistence practices maintain land-based relationships that emphasize valuation systems that also prioritize culture (Bélisle et al., 2021; van Horne et al., 2021). For many Tribes, safely harvesting and consuming fish is an assertion of food sovereignty that maintains their relationship (i.e., knowledge) with their lands; contaminated fish limit the sovereignty of whole communities in the present as well as for future generations (Hoover, 2017).

Similarities between Framework 1 (an illustrative combined toxicity framework) and Framework 3 (a mixture toxicity framework) emphasize the potential for combined and mixture toxicity frameworks to represent risk reasonably and more appropriately (Figure 3-S1 and Figure 3-S2). Framework 1 (combined toxicity) and Framework 3 (mixture toxicity) produce HQs that are remarkably similar in terms of toxic contribution from the five compound classes and the magnitude of contamination (Table 3-S3 and Table 3-S3, respectively). The similarity is likely the result of RfDs being the same for all compound classes except for NDL-PCBs. The most recent NDL-PCB RfD is protective of liver toxicity rather than reproductive toxicity and is two orders of magnitude higher than the reproductive RfD. As a result, NDL-PCB toxicity accounts for about 10% of total mixture toxicity and <1% of combined toxicity.

Our results also point to why combined toxicity is a more appropriate strategy than hazard quotients based on single compounds. Consistently, across all three toxicity frameworks, the majority (>90%) of fish samples had a hazard index (HI) greater than 1, the threshold for safe fish consumption (Figures 4, 5, 6 and Tables 3, 4, 5). These results emphasize conclusions from Stahl et al. (2009) that the contaminants are "widely distributed" and indicate that fish tissue concentrations are sufficient to threaten human health, especially for subsistence fish consumers. Further, 72% of predator samples and 33% of bottom-dweller samples would be considered safe to consume with respect to total PCB contamination but have an HI > 1. With respect to Hg contamination, less than 10% of samples had an HI > $HQ_{mercury}$. While single contaminant fish consumption guidelines accurately identify "unsafe" predator samples, it is important to acknowledge that the HI estimate here is limited and does not consider other chemicals known to be present in fish tissue (e.g., PFAS – Hoa et al., 2022; pesticides – Zhou et al., 2018). Therefore, these HIs are likely to underestimate the total combined toxicity.

Based on evidence presented here, combined toxicity more accurately represents risk, especially for PCB contamination. Presently, the planetary boundary for novel entities has been exceeded, in part because the risks associated with exposure to the majority of the 100,000s of chemicals that are present in the environment cannot be known (Persson et al., 2022). The time for managing individual contaminants has passed; it is time to consider chemical mixtures and their combined and mixture toxicity effects on humans and the environment.

3.4.2 What is the variation in relative and absolute contributions to combined toxicity between fish composite types and across ecoregions?

Contaminant burdens vary between bottom-dwellers and predators. Bottom-dwellers are significantly more contaminated than predator samples ($P < 0.001$). Also, the composition of the toxicity varies between the two composites. This difference is relatively unsurprising since bottom-dwellers include many species of carp, suckers, and catfish; benthivores spend much of their life in the lake bottom consuming contaminated sediment as a byproduct of messy eating. For all three toxicity frameworks, but especially the most sensitive (Framework 2), mercury contributes most of the toxicity (> 50%) for predator samples but less than 25% of toxicity for bottom-dwellers. Likely, this difference is because greater lipid content for bottom-dwellers leads to a greater body burden for the hydrophobic compounds.

With this framework, we can explore regional differences that emphasize the importance of localized strategies for risk assessment (Buell et al. 2021). Across the U.S., HIs exceed 1 but the toxic contribution of the five compound classes varies (Figure 3-2 and Figure 3-S6). Of the waterbodies with HI < 1, most of those waterbodies are west of the Mississippi River. These waterbodies tend to be further away from the primary sources (i.e., urban areas) of the contaminants considered in this study. Some literature shows that eutrophic systems have lower contaminant concentrations because of biodilution (Chen & Folt, 2005). Although U.S. land area is divided into nutrient regions, the intra-region variability is high and in-lake measurements are needed to evaluate this relationship at the national scale, as demonstrated here.

Regional differences in contamination exist for both composite types ($P < 0.001$). While not extensively evaluated, understanding regional patterns of contamination is necessary for developing localized strategies to mitigate risks associated with fish consumption (e.g., federal and state fish consumption guidelines). Figure 3-2 shows us that Hg is the dominant contaminant in predator samples in most regions across the U.S. Although Hg guidelines alone identify the predator samples that are unsafe, as demonstrated in Figure 3-S7, Hg alone significantly underestimates the actual risk. This underestimation is problematic because 1) accurate guidelines are important so that individuals can make informed decisions about frequency and fish size for safe fish consumption, and 2) policies aimed at reducing risk must be informed by the magnitude of reduction needed.

Contaminant sources and landscape characteristics that influence contaminant cycling and accumulation at multiple scales and might explain the variability of magnitude and composition of toxicity across the U.S. Urban areas are known sources of PCBs, dioxins, and furans (Hafner & Hites, 2003) and we see significant negative relationships between bottom-dweller combined toxicity and distance from urban areas ($P< 0.001$; Figure 3-S8). Hazard indices for bottom-dwellers are higher in regions along the east coast. For both predators and bottom-dwellers, HIs tend to be higher east of the Mississippi River, the most populous part of the U.S. Coal-fired power plants are known sources of Hg, but previous studies have concluded there are not strong relationships between Hg deposition and fish tissue concentrations, largely because the presence of landscape characteristics associated with methylation are more important (e.g., forested land; Depew, 2009). We see a strong relationship between forested landcover area and Hg concentrations in fish (Figure 3-S9).

3.4.3 How can combined toxicity be used to quantify risk more appropriately?

To judge the merits of quantifying combined toxicity as an element of cumulative risk, it becomes necessary to articulate a framework that appropriately protects health. Importantly, how we determine acceptable risk is strongly influenced by our worldview. In *Pollution is Colonialism* (2021), Métis author Max Liboiron describes current settler philosophies related to our access to the environment as being grounded in a colonial logic of *permission to pollute*; humans have a right to the environment so long as we pollute below an established threshold. The central assumption is that pollution below the threshold poses no risk. It is not a stretch to extend this logic to xenobiotic chemical contamination. Our current strategies of managing emissions and exposure to primarily single contaminants ignores chemical and social realities – chemical transformations can create toxic reaction products (Fernando et al., 2018; Grimm et al., 2015) and/or potentiate other chemicals and social transformations modify existing relationships between humans, plants, and animals (Murphy, 2017).

Defining combined and mixture toxicity as separate elements of cumulative risk makes the distinction between toxicity that results from dissimilar effects from simultaneous exposure to contaminants (combined toxicity) and toxicity that results from similar effects from simultaneous exposure to contaminants (mixture toxicity). Limited by regulatory reference doses (RfDs) that identify and quantify toxic effects, we considered multiple molecular initiating events that lead to dissimilar toxic effects and cause harm to different parts of the same system (e.g., the human body or a bee colony; LaLone et al. 2017). That is, we can consider combined toxicity. Recently, others have stated that additive toxicity is reasonably protective given scientific uncertainty (Gandhi et al., 2017); we propose additive combined toxicity protective of the most sensitive endpoint as equally reasonable given the scientific uncertainty surrounding combined and mixture toxicity and the paucity of established contaminant reference doses. We believe this to be the necessary strategy for managing toxicity from exposure to chemical mixtures more holistically.

Moving beyond the theoretical considerations for how to quantify combined toxicity, we discuss our results as a practical framework for quantifying combined toxicity. Previous risk assessment work shows that safely protecting Tribal Nations also benefits non-Indigenous communities (Buell et al., 2020). Given that fish consumption continues despite an awareness of chemical contamination (O'Neill, 2007; Savadatti et al., 2019; Wattigney et al., 2019), it is important to consider the most abundant contaminants at environmentally relevant concentrations. Here we meet both criteria by using monitoring data.

Managing toxicity based on the combined toxicity of the most sensitive RfD is a reasonable framework that holistically considers the value associated with fish consumption. Hazard indices, like the ones used here are relatively simple means to estimate the mixture and combined toxicity associated with the simultaneous exposure of contaminants. Such frameworks are commonly used to represent the hazards associated with contamination via fish consumption (e.g., Gandhi et al. 2017).

Effective strategies for quantifying cumulative risk necessitate accurate representations of toxicity. Results from this study reveal that compared to individual contaminant toxicity, combined toxicity more accurately represents the magnitude of risk. This finding is an important contribution to the field because to date toxicology narrowly focuses on mixture effects (Martin et al. 2021). Continuing this path marginally advances scientific understanding as risk assessment cannot keep pace with the risks associated with 100,000s of chemicals and their mixtures (Persson et al. 2022).

3.5 Conclusion

Combined toxicity is a valid framework that more nearly represents risk and could be used to establish fish consumption guidelines (FCGs). Currently, FCGs as a risk mitigation strategy underestimate actual hazards associated with fish consumption. More than 90% of fish samples considered here exceed the safe threshold (HI = 1); based on individual contaminant concentrations, most fish samples are also mis-identified as safe. Using the most sensitive endpoint for components of a chemical mixture rather than the current strategy that expects a shared toxic pathway is more protective and more acceptably protects human health and the environment.

Magnitude of contamination and variation in relative importance of chemical classes vary between different fish types and across ecoregions. Such variation underscores the importance of local research partnerships, particularly when quantifying risk associated with environmental exposures. After all, values and priorities can also vary regionally but on-going relationships between researchers and community promote shared priorities and can lead to more meaningful interpretations and definitions of acceptable risk (Buell et al. 2021; Shaw et al. 2022).

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3.7 Chapter 3 supplementary information

Table 3-S1. Reference doses (RfDs) for chemical compounds considered in this work. Values in *italics* indicate no adverse effects levels (NOAELS), lowest observable adverse effect level (LOAELs), or tolerable daily intakes (TDIs) which can be used to estimate RfDs. For PCBs, studies sometimes consider individual congeners, groups of congeners, or Aroclors; that detail is specified in each cell.

PCBs	**DL-PCBs/ PCDDs/PCDFs**	**MeHg**
36 ug PCB_{28}/kg-day liver toxicity (Chu et al. 1996a in JECFA 2016)	75,000 / (mg/kg-day) cancer slope factor (MDCH 2013)	*1.3 ug/kg-day neuro development (ATSDR 1999)*
42 ug PCB_{128}/kg-day liver toxicity (Lecavalier et al. 1997 in JEFCA 2016)	2.3 pg TEQ/kg-day (JECFA 2001 in EFSA 2018)	*1.6 ug/kg-day fetal development (Health Canada 2007)*
34 ug PCB_{153}/kg-day liver toxicity (Chu et al. 1996b in JEFCA 2016)	2 pg TEQ/kg-day (SCF 2001)	0.1 ug/kg-day cardiovascular+ neurologic effects (MDCH 2009)
10 ug PCB_{153}/kg-day liver and thyroid effects (NTP 2006a)	1.0 pg TCDD/kg-day (ATSDR 2012)	0.014 ug/kg-day cardiovascular events (MDCH 2009)
2E-5 mg A1254/kg-day (CA EPA 2008)	*0.001 ug 23478 PeCDF/kg-day thymic effects (ATSDR 1994)*	0.1 ug/kg-day neurologic effects (MDCH 2009)
0.07 ug A1016/kg-day reduced birth weight (EPA 1996B)	*0.01ug/kg-day intermediate duration oral MRL (ATSDR 1994)*	*0.19 ug/kg-day prenatal neurodevelopment (EFSA 2015)*
0.02 ug A1254/kg-day immunological effects (EPA 1996C)	*200 pg TCDD/kg-day hemolytic complement activity (ATSDR 1998)*	*0.1 ug/kg-day (US NRC in EFSA 2015*)
2.0 / (mg/kg-day) cancer slope factor (EPA 1997A)	*20 pg TCDD/kg-day Thymus weight (ATSDR 1998)*	*0.23 ug/kg-day cardiovascular effects (JECFA 2007)*
1.7 ug/kg-day neurologic effects (MDCH 2012)	*1 pg/kg-day altered social interactions (ATSDR 1998)*	0.02 ug/kg-day no adverse effects (USEPA 1995)

		0.05 ug/kg-day 0.01 ug/kg-day 0.025 ug/kg-day Cognitive and neurologic effects (Rice, 1996; Gilbert and Grant-Webster 1995)
		0.3 ug/kg-day central nervous system effects (USEPA 1985)
		0.3 ug/kg-day neurologic effects (ATSDR 1999)
		0.05 mg/kg-day kidney damage (Munro et al. 1980)

Table 3-S2. Median HQs based on the most recent endpoint (i.e., combined toxicity; Framework 1). Differences between the fish composites were evaluated using Mann-Whitney non-parametric t-tests with P-values reported for each fish composite comparison.

	NDL-PCB Tox.	DL-PCB Tox.	Dioxins Tox.	Furans Tox.	Mercury Tox.	Comb. Tox.
Predator Samples (N = 484)	0.0009	0.07	0.007	0.05	2.61	3.49
Bottom-dweller Samples (N = 393)	0.005	0.61	2.11	0.54	0.78	6.15
P-value	2.2E-16	2.2E-16	2.2E-16	2.2E-16	2.2E-16	2.4E-13

Table 3-S3. Median HQs based on the shared reproductive endpoint (i.e., mixture toxicity; Framework 3). Differences between the fish composites were evaluated using Mann-Whitney non-parametric t-tests with P-values reported for each fish composite comparison.

	NDL-PCB Toxicity	DL-PCB Toxicity	Dioxins Toxicity	Furans Toxicity	Mercury Toxicity	Mixture Toxicity
Predator Samples (N = 484)	0.09	0.07	0.01	0.05	2.61	3.65

Bottom-dweller Samples (N = 393)	0.48	0.61	2.10	0.54	0.78	6.65
P-value	2.2E-16	2.2E-16	2.2E-16	2.2E-16	2.2E-16	6.6E-15

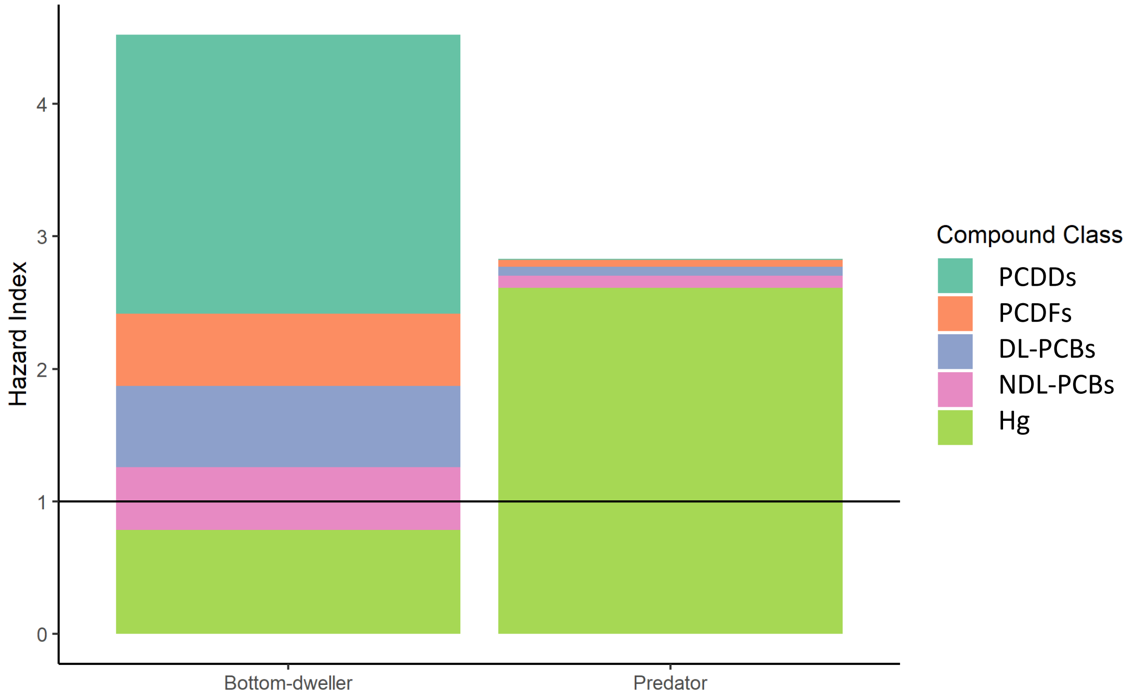

Figure 3-S1. Median hazard indices for framework 3 which is based on the shared reproductive endpoint. Median hazard quotients for each compound class are summed together.

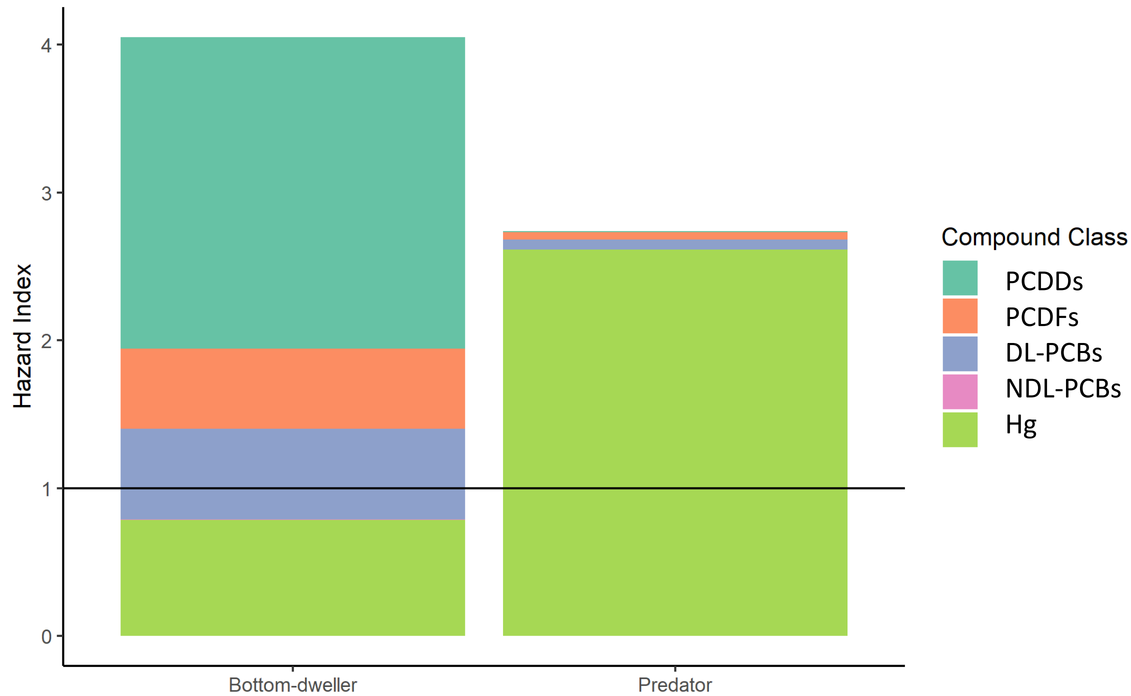

Figure 3-S2. Median hazard indices for framework 1 which is based on the most recently available endpoint. Median hazard quotients for each compound class are summed together

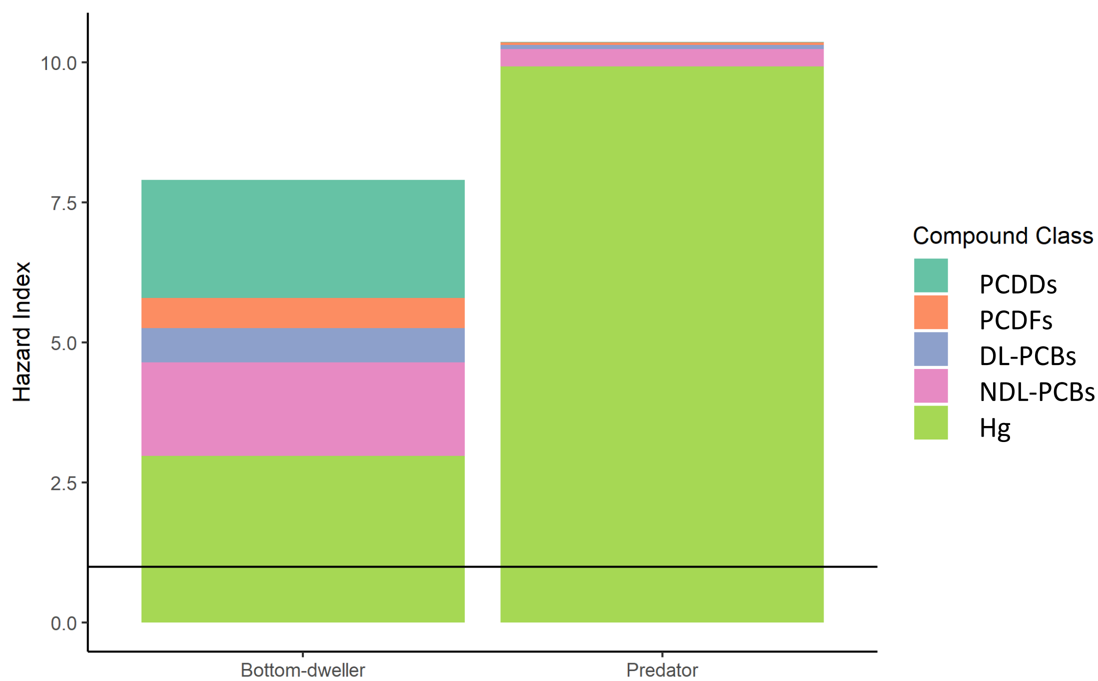

Figure 3-S3. Median hazard indices for framework 2 which is based on the most sensitive endpoint. Median hazard quotients for each compound class are summed together.

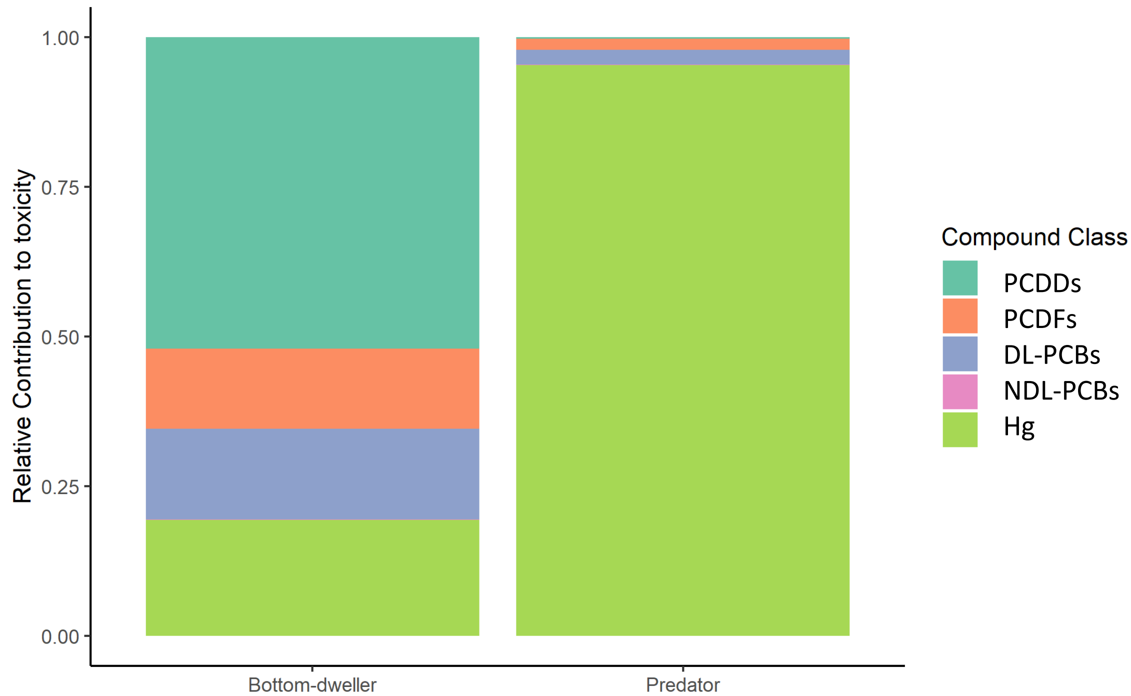

Figure 3-S4. Relative toxic contribution for framework 1 which is based on the most recent endpoint. Fractions are calculated based on median hazard indices (HIs), the sum of the hazard quotients for each of the five compound classes.

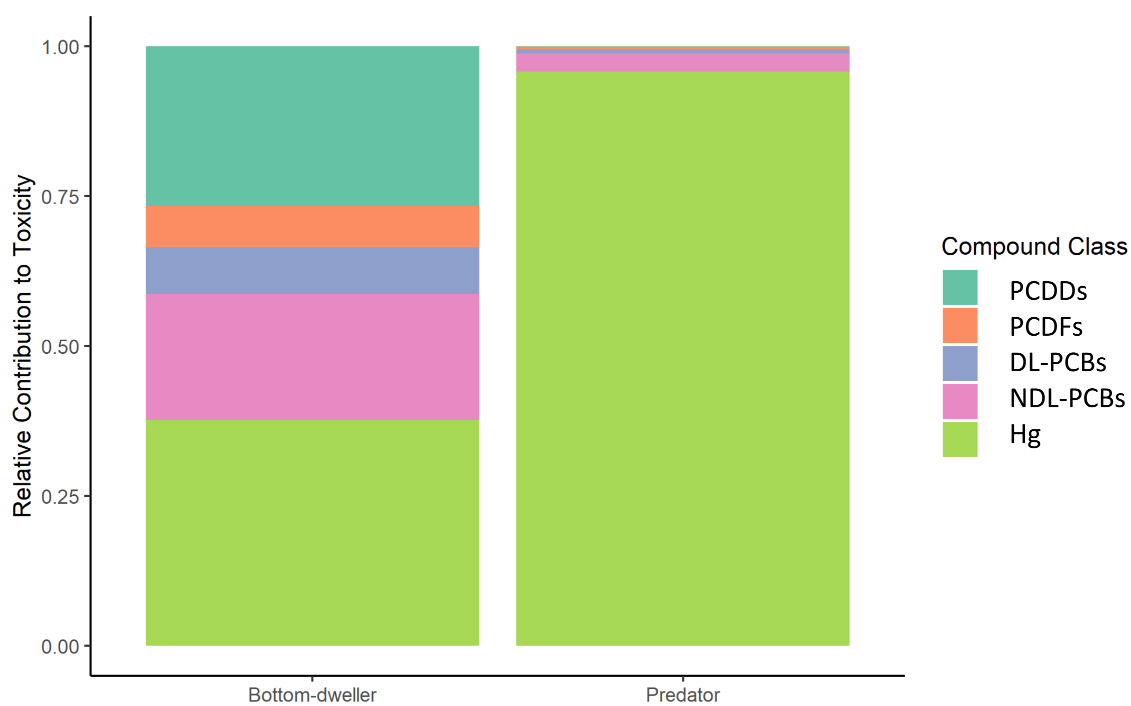

Figure 3-S5. Relative toxic contribution for framework 3 which is based on the shared reproductive endpoint. Fractions are calculated based on median hazard indices (HIs), the sum of the hazard quotients for each of the five compound classes.

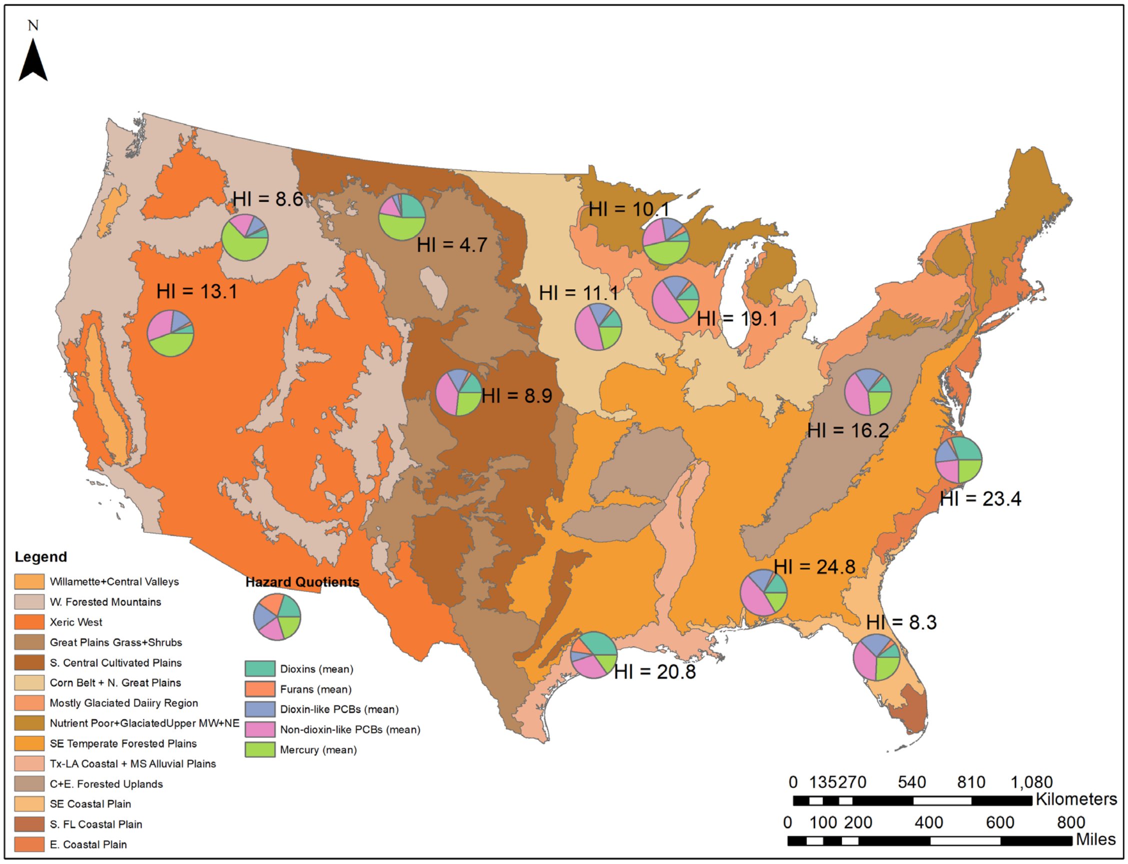

Figure 3-S6. Mean hazard indices (HIs) of bottom-dweller samples are summarized for EPA-defined nutrient regions. Non-dioxin-like PCBs are an important source of toxic risk for bottom-dwellers across ecoregions.

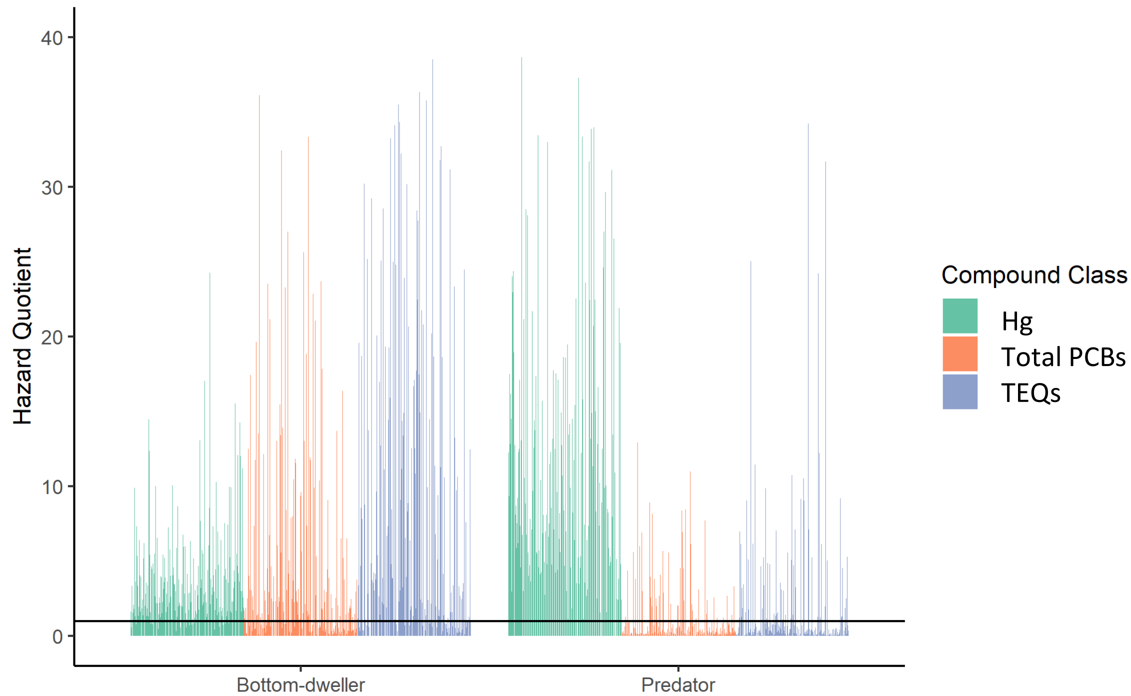

Figure 3-S7. Hazard quotients (HQs) for Hg, total PCBs, and toxic equivalencies (TEQs; PCDDs, PCDFs, and DL-PCBs). The black line represents a HQ = 1.

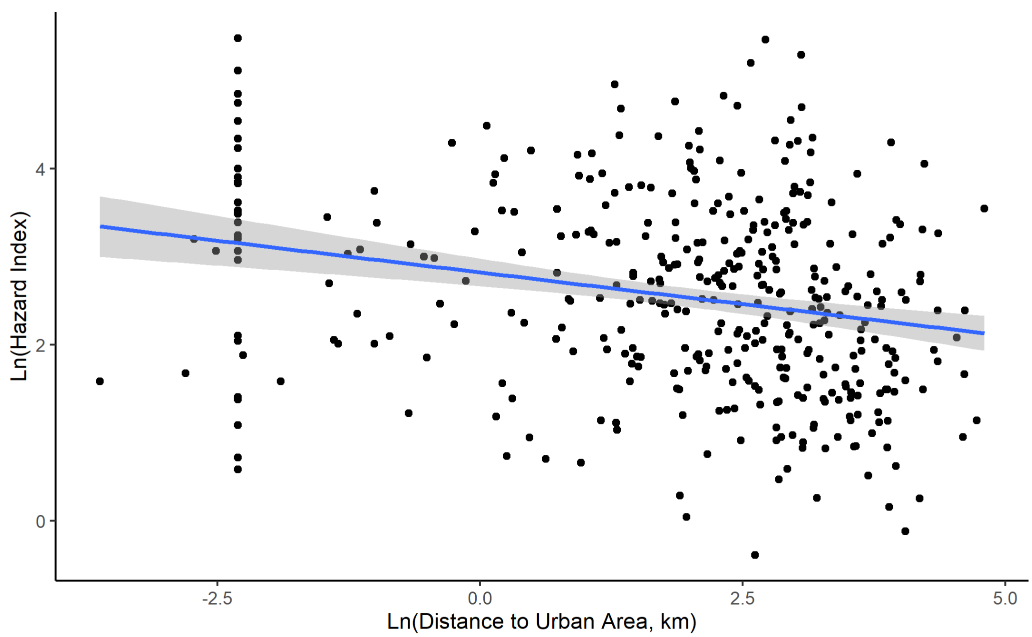

Figure 3-S8. Regression for the hazard index (HI) as a function of the sample lake's distance to an urban area for bottom-dwellers (rho = -0.247, P = 4.3 x 10^{-8}). Distance to urban area was calculated in ArcMap.

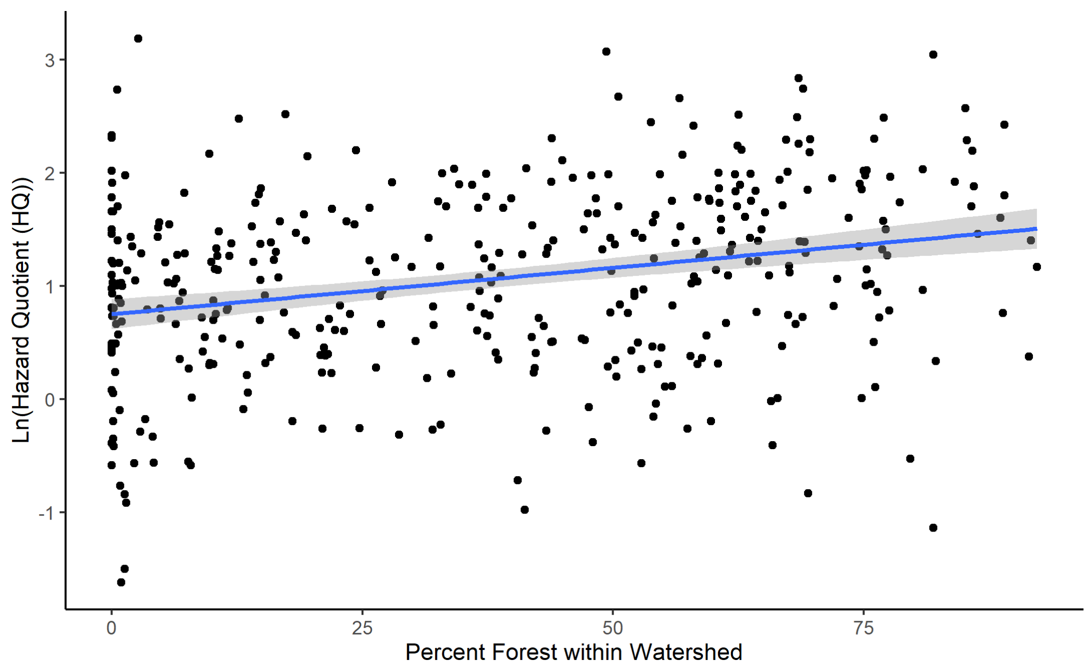

Figure 3-S9. Regression for the mercury hazard quotient (HQ_{Hg}) as a function of percent forest land within the watershed for bottom-dwellers (rho = 0.28; P = 1.9 x 10^{-8}). Percent forest within the watershed is from the U.S. EPA's LakeCat database (Hill et al., 2018).

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4 Local remediation can decrease local concentrations of ASEPs – a policy analysis of PCB concentrations in the Great Lakes basin

4.1 Introduction

Even though U.S. production of PCBs ended in 1993 (Breivik 2002), PCBs are difficult to break down and continue to be present in concentrations that present a hazard to human health and the environment. Globally, about 1.3 million tonnes of polychlorinated biphenyl compounds (PCBs) were produced since 1929, with more than 50% originating in the United States; most U.S.-based PCBs were manufactured by Monsanto (Breivik et al., 2002; Melymuk et al., 2022). The 209 PCB congeners were produced and sold as chemical mixtures. PCBs were used in many industries with different mixtures used for different purposes (e.g., plasticizers, electrical fluid, carbon-less copy paper, etc.). Based on their chemical structure and the resulting toxicity, the 209 congeners can be organized into two groups: dioxin-like and non-dioxin-like PCBs.

While PCBs were initially heralded as a major technological advance, the legacy of PCBs is associated with past and on-going issues of environmental injustice that extend through space and time. In the environment, the transport and transformation of PCBs is largely controlled by biogeochemical features like the movement of air masses, organic matter sequestration, and food web structures. Physical and chemical properties of a compound affect movement among different environmental compartments (i.e., phase partitioning; Hawker & Connell, 1988; Sarna et al., 1984) facilitating selective transport and leads to global dispersion, often concentrating PCBs at high latitudes (e.g., the Great Lakes basin; Perlinger et al., 2016; Wania & MacKay, 1996). Due to these patterns of volatilization and deposition the spatial extent of potential harm is greatly expanded.

Long environmental half-lives, ranging from tens to hundreds of years, extend the injustice from PCB exposure through time. Although PCB production and use facilities were sited based on proximity to certain demographic groups (e.g., Spears 2014), these sources continue to be re-mobilized. Through time, PCB risks are passed on to younger generations through off-loading via placental transfer and breast milk (Dekoning & Karmaus, 2000).

Fish consumption, varying greatly among different populations, remains the primary exposure pathway for humans to PCBs. For many Tribal Nations, including the Anishinaabe whose homelands are within the Great Lakes basin, harvesting and consuming fish is an important cultural practice. However, current risk-management strategies center on fish consumption guidelines (FCGs) that recommend a limited number of meals per month based on contaminant concentrations within an area (e.g., Michigan Eat Safe Fish Guides). Even the U.S. EPA's more protective fish consumption rates for subsistence fish consumers (142.5 g/day) is far below the KBIC's desired fish consumption rate of 242 g/day (Asher Consulting, 2016). A primary shortcoming of FCGs is the assumption that populations have an interest in and ability to limit consumption. Yet previous work shows that fish consumption often continues in the presence of contamination because people are unable and/or unwilling to limit their consumption (O'Neill, 2007; Wattigney et al., 2019). Therefore, particularly in the interest of environmental justice, it becomes necessary to consider limiting toxic exposure via alternative strategies such as site cleanup and/or remediation that directly limit and eliminate contamination sources.

To assess the effectiveness of alternative strategies focused on environmental PCB sources, this research sought to quantify the effect of Canada's 2008 PCB Regulation (SOR 2008-273) on Great Lakes atmospheric and fish tissue PCB concentrations, employing a difference-in-difference (DID) approach. To our knowledge this is the first paper to quantitatively evaluate the effect of a PCB reduction policy stemming from the Stockholm Convention. This is particularly important because the Convention seeks environmentally sound PCB management by 2028, with an elimination of in-use PCBs by 2025 (Melymuk 2022; http://www.pops.int/TheConvention/Overview/tabid/3351/Default.aspx). Using atmospheric and fish tissue monitoring data from within the United States and Canadian boundaries, samples were grouped based on their sample location; U.S. stations are unimpacted by Canadian policy and Canadian stations are presumed to be impacted. Primary results of this research found evidence that Canada's PCB regulations have had a statistically significant effect on decreasing Great Lakes atmospheric PCB concentrations. Additionally, there is evidence of a significant decrease in lake trout fish tissue. However, results related to the PCB levels in fish tissue are heterogeneous among the different lakes.

The rest of the paper is structured as follows: first we present the international policy context for PCBs and briefly describe the sample locations. Next, we describe the research methods and data used in the analysis, including the difference-in-difference (DID) model for the two types of PCB data we have (i.e., vapor-phase and fish tissue). In section 4.4 we present results from the analyses and in 4.5 and 4.6 share an interpretation of the results. Finally, we end with conclusions and policy implications.

4.2 Background and context

Rather than quantify the economic impacts of Canada's PCB Regulations policy, we quantify the environmental impacts of the policy, specifically the efficacy of the policy in achieving the goals originally laid out in the Stockholm Convention: protection of human health and the environment. With the over-arching goal of rebuilding fish-human relationships, this approach is aligned with Indigenous knowledge and values as a strategy for environmental management (Jacobs et al. 2022). Often difference-in-difference (DID) analysis is used to quantify the economic impacts of policy action. The presence of chemical contamination has negative economic effects while site-remediation can increase property value (Braden et al., 2011; Savchenko & Braden, 2019; Gardner, 2022). Within Indigenous Knowledge Systems, practices that maintain place-based relationships are central and an important component of environmental valuation and prioritization. By evaluating policy efficacy within the framework of contaminant reductions we are emphasizing the significance of contaminant reductions from a non-monetary valuation framework. Valuing ecosystems based on landscape practices and a sense of place align well with Indigenous philosophies and prioritize human health and the environment (Bélisle et al., 2021; Donatuto et al., 2020; Poe et al., 2016).

To protect human health and the environment, the movement, trade, and use of PCBs are addressed in the Basel Convention, the Rotterdam Convention, and the Stockholm Convention. Most directly relevant to this analysis is the Stockholm Convention which entered into force in 2004 with provisions to eliminate the production and use of PCBs and safely manage stockpiles and wastes containing PCBs. With 150 other countries, both the U.S. and Canada signed the Convention. Of the two countries, only Canada has ratified and taken significant action to meet the goals outlined in the Convention (Melymuk et al. 2022).

Pursuant to the Canadian Environmental Protection Act of 1999, Canada accelerated the phase out and elimination of PCB-containing material in concentrations greater than 50 mg/kg under the SOR/2008-273 (PCB Regulations 2008). As a signatory to the Stockholm Convention, this legislation is aligned with article one which outlines the objective for the convention, "… to protect human health and the environment from persistent organic pollutants" (Article 1, p. 6; Stockholm Convention).

Ironically, the geographically specific Great Lakes Water Quality Agreement establishes a commitment by both Canada and the United States to restore and protect the waters of the Great Lakes from chemical contamination (GLWQA 1972). In 2012, the GLWQA amendments established the *Chemicals of Mutual Concern Annex* with a commitment to "… contribute to the achievement of the General and Specific Objectives of this Agreement by protecting human health and the environment through cooperative and coordinated measures to reduce the anthropogenic release of chemicals of mutual concern into the Waters of the Great Lakes" (Annex 3). PCBs are one of the chemicals listed.

Basin-wide, trends demonstrate that vapor-phase and fish tissue PCB concentrations are decreasing. In the vapor-phase, PCBs show a half-life of about 15 years with trends that are homogeneous across sample sites (Salamova et al., 2015). Recently these trends have, in part, been attributed to the Stockholm Convention (Hites, 2019). In fish tissue, trends show improvement but are heterogeneous across sample sites (McGoldrick & Murphy, 2016; Hites & Holsen, 2019; Urban et al., 2020). Using the DID analysis, we can quantify the magnitude of the policy impact instead of making inferences about policy efficacy.

Different lakes and bays in the Great Lakes watershed vary greatly with respect to nutrient concentrations, food web structure, and contaminant sources, and these factors can influence the concentration and accumulation of PCBs in the food web (e.g., Abma et al., 2015; Chen & Folt, 2005; Venier et al., 2014). Due to their large surface area and low temperatures, the upper Great Lakes (Superior, Huron, Michigan) represent major sinks for semi-volatile compounds (Guo et al., 2018). Lakewide Action and Management Plans (LAMPs) for the five Great Lakes confirm that contaminant concentrations have declined but the rate of decline has slowed, and fish consumption guidelines remain necessary (*Lake Erie Lakewide Action and Management Plan*, n.d.; *Lake Huron Lakewide Action and Management Plan*, n.d.; *Lake Michigan Lakewide Action and Management Plan*, n.d.; *Lake*

Ontario Lakewide Action and Management Plan, 2018; *Lake Superior Lakewide Action and Management Plan*, 2016).

Here our primary objective is to evaluate the effect of Canada's SOR 2008-273 policy in reducing atmospheric and fish tissue PCB concentrations in the Great Lakes basin. Using a difference-in-difference analysis, we test two hypotheses: 1) There is a significant decrease in vapor-phase PCB concentrations that is the result of the policy, and 2) There is no significant decrease in fish tissue PCB concentrations because of the policy. In our conclusions and policy implications section we answer one question related to our hypotheses: *Can local regulation of ASEPs significantly reduce local exposures to these contaminants, and, if so, in what environmental reservoirs?* Here we describe the data and models used to test these hypotheses, present our results, and discuss plausible explanations for the describe the data and models used to test these hypotheses, present our results, and discuss plausible explanations for the unequal effects of policy on vapor-phase (atmospheric) and fish tissue PCB concentrations.

4.3 Methods

As part of an on-going partnership with the Keweenaw Bay Indian Community Lake Superior Band of Chippewa Indians (KBIC), this work is guided by a Tribal Council approved *Guidance for Research Partnerships* that, "describes responsibilities and expectations associated with collaborative, participatory, and community-engaged research" (Gagnon et al., 2021; Shaw et al., 2022). Consent for this research was requested by Emily Shaw, following two years of relationship building, and granted during a meeting with KBIC's Natural Resources Department staff. Only then, with ongoing KBIC guidance and direction, was the project proposal written, defended, and research begun. We present these research methods to reiterate the non-negotiable status of consent and permission as a part of Indigenous-university research partnerships (Liboiron, 2021; Whyte, 2020).

4.3.1 Software and Data

Statistical analyses were completed in R (R Core Team, 2022). Specifically, event study regressions and difference-in-difference (DID) analyses (Berge, 2018) were conducted using vapor-phase and fish tissue total PCB concentrations from Canada and the United States. Sample locations differed for vapor-phase PCBs and fish tissue PCBs. There were 7 atmospheric and 16 fish sample locations within the Laurentian Great Lakes basin, respectively (Figure 4-1). Sample sites were coded as being in the U.S. or Canada based on the sample location, and the sample period was defined as 1990 – 2020. Sites were excluded from the analysis if the sample period did not include at least three years prior to and at least three years post-policy implementation (2008). The inter-quartile range (IQR) is the difference between the first and third quartiles of the data and defines outliers. In this work, outliers were defined as outside ±1.5*IQR of the median and excluded from the analysis on a site-by-site basis.

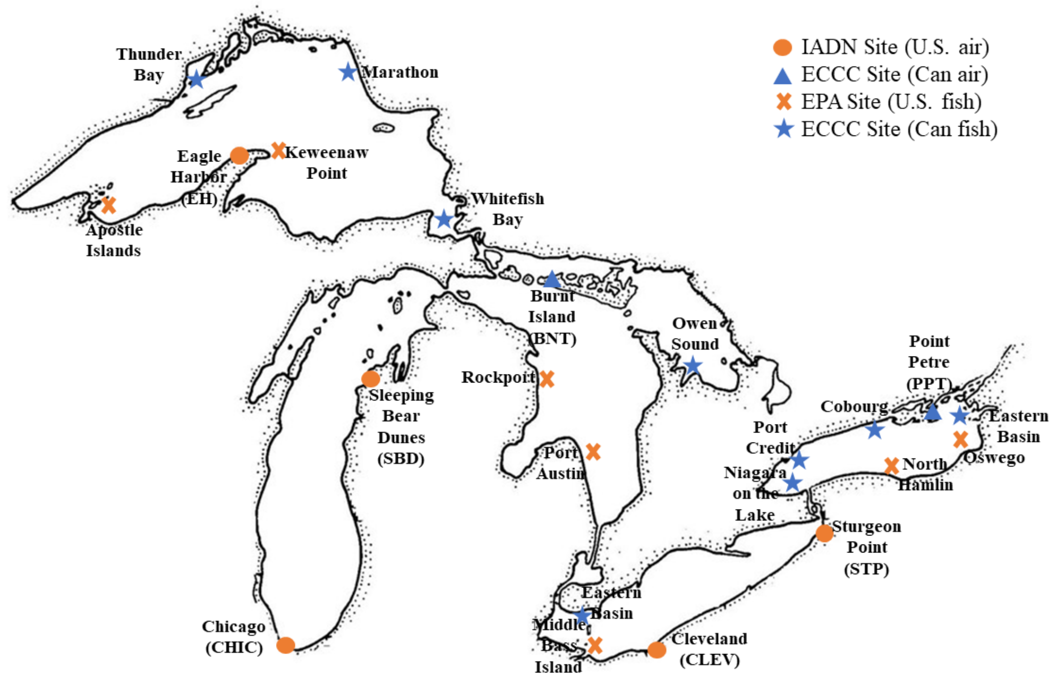

Figure 4-1. A map of vapor-phase and fish tissue sample locations. Vapor-phase samples were collected at seven sample location by the Integrated Atmospheric Deposition Network (IADN) in the U.S. and Environment Climate Change Canada (ECCC) in Canada. Lake trout and walleye samples were collected at 16 locations by the U.S. Environmental Protection Agency (USEPA) and ECCC. Walleye samples were collected at two sites in Lake Erie. At the other 14 sample locations lake trout were collected. Image courtesy of NOAA's Great Lakes Environmental Research Laboratory (GLERL) and is licensed under the Creative Commons.

The atmospheric data used in this study were collected by long term monitoring programs of Canada and the U.S. Atmospheric PCB concentrations are measured by the Integrated Atmospheric Deposition Network (IADN) and Environment Climate Change Canada (ECCC) at five U.S. and two Canadian sample locations. Canada quantifies vapor-phase total PCBs based on 78 congeners and IADN quantifies total PCBs based on 58 congeners. Individual samples were excluded from the analysis if the summed total PCB concentration was 0 or the concentration was considered an outlier (outside 1.5*IQR). In total, 122 vapor samples were removed; 34 samples were removed because their sum was 0 (24 samples from the Burnt Island site and 10 samples from the Point Petre site, both in Canada) and 88 outliers were removed (< 6% of samples at each site). In 2011, a second sampler was placed at Point Petre. Because the sample concentrations were summed, for the Point Petre site only, we calculated the mean PCB concentrations after 2011 and included those values in our analysis.

Fish tissue data used in this study were collected by long term monitoring programs of Canada and the U.S. Whole fish PCB concentrations are measured by the U.S. Environmental Protection Agency (USEPA) and Environment Climate Change Canada

(ECCC) at six U.S. and eight Canadian sample locations throughout the Laurentian Great Lakes basin. Lake trout and walleye are collected and analyzed for PCBs, the former in Lakes Superior, Michigan, Huron, and Ontario and the latter in Lake Erie. In Lake Erie, walleye samples were collected prior to 2011 and lake trout after 2011. To minimize confounding factors, sample locations were only included if one fish species was collected across the sampling period; this excluded the Eastern Basin (Canadian fish sample location) and Dunkirk (U.S. fish sampling location) in Lake Erie. Canada quantifies total PCBs as Aroclor 1254 and the U.S. quantifies total PCBs based on 142 congeners. In total, 183 samples were excluded from analysis; 4 samples were excluded based on lab codes (3 samples from the Apostle Islands, Lake Superior, and 1 sample from Middle Bass Island, Lake Erie), 31 outliers were removed (≤ 6% of samples at each site), and 152 samples without age data were excluded (138 lake trout samples (1.7%) and 14 walleye samples (2.9%)). Included in the analyses were 566 samples that were below the detection or quantification limit with values assigned, by different labs, as 50, 60, 70, 80, 90, or 100 ug/kg.

4.3.2 Difference-in-difference (DID) analysis

Difference-in-difference (DID) analysis is a quasi-experimental technique that quantifies a policy effect on a specific characteristic (e.g., fish tissue PCB concentrations) at treatment (Canadian sites influenced by policy SOR 2008-273) and control (U.S.) sites. Total PCB concentrations were normalized to minimize ecosystem differences among sample sites. Vapor-phase PCB data were temperature normalized by difference ($C_{temp\text{-}norm} = C_{measured} - C_{medianT}$ [) and fish tissue PCB data were age-normalized by ratio ($C_{age\text{-}norm} = (C_{measured} / A_{fish}) \times A_{median}$ [Equation 4-2).

$$C_{temp\text{-}norm} = C_{measured} - C_{medianT} \quad \text{[Equation 4-1]}$$

- $C_{measured}$ is the measured PCB concentration
- $C_{medianT}$ is the estimated PCB concentration at the site's median temperature based on site-specific regressions of Ln[PCB] vs. 1/Temp (K)

$$C_{age\text{-}norm} = (C_{measured} / A_{fish}) \times A_{median} \quad \text{[Equation 4-2]}$$

- $C_{measured}$ is the measured PCB concentration
- A_{fish} is the fish age
- A_{median} is the median for age for a specific site-year pair

Data suitability was evaluated using two techniques: a qualitative assessment of normalized concentration trends at each sample site as a function of year and an event study regression that calculates DID coefficients for each year. For the event study, DID coefficients (i.e., policy effects) prior to policy implementation are expected to be zero. Data were considered suitable if three of the five years immediately prior to policy implementation had policy effects coefficients of zero.

For air data, a Great Lakes basin wide DID analysis was based on $Y_{i,t} = \beta_0 + \beta_1 A_i + \beta_2 T_t + \beta_3 DID_{i,t} + \varepsilon_{i,t}$ [Equation 4-3 and for fish tissue data, a Great Lakes basin wide DID analysis was based on $Y_{i,t} = \beta_0 + \beta_1 A_i + \beta_2 T_t + \beta_3 DID_{i,} + \beta_4 L_{i,t} + \varepsilon_{i,t}$ [Equation 4-4. In the simplest form, a 2x2 DID analysis organizes the data across two sets of binary variables (i.e., fixed effects) and calculates an interaction term (DID) as the product of the site (*A*) and time (*T*) effects. Therefore, $DID_{i,t}$ is only equal to one at treatment (Canadian) sites after the policy is in effect. With respect to the coefficients, data are organized as either a control (U.S.; A = 0) or treatment (Canada; A = 1) and as either pre-policy (Year < 2008; T = 0) or post-policy (Year ≥ 2008; T = 1). Taking the difference between the four regression equations leaves β_3, the DID coefficient which represents the policy effect (Table 4-1). To evaluate the validity of the basin wide DID analyses, subsets of data were analyzed. For air data, a paired analysis between Point Petre (treatment, Canada) and Sturgeon Point (control, U.S.) was done. For fish tissue data, a greater number of sample locations facilitated lake-specific analyses for lake trout (including Lakes Superior, Huron, Ontario) and walleye (Lake Erie).

Table 4-1. Using coefficients from Equation 4-3 and Equation 4-4, this table shows how the two-way fixed effects (i.e., 2x2 DID) model describes site and time in binary terms. Given that *DID* is the interaction of site and time, *DID* = 1 only at treatment sites in the post-policy period. The difference between the rows and columns shows that β_3, the policy effect, is only present when DID = 1.

	Control (U.S.) A = 0	Treatment (Canada) A = 1
Pre-Policy (Year < 2008) T = 0	β_0	$\beta_0 + \beta_1$
Post-Policy (Year ≥ 2008) T = 1	$\beta_0 + \beta_2$	$\beta_0 + \beta_1 + \beta_2 + \beta_3$

$Y_{i,t} = \beta_0 + \beta_1 A_i + \beta_2 T_t + \beta_3 DID_{i,t} + \varepsilon_{i,t}$ [Equation 4-3]

- Where:
- *i* and *t* represent the sample location and years;
- *Y* is the temperature-normalized mean PCB concentration (equation 1);
- β_0 is the regression intercept;
- β_1 is the coefficient for the sample site fixed effect;
- A_i is a binary variable to distinguish between control (0) and treatment (1) locations;
- β_2 is the coefficient for the year fixed effect;
- T_t is a binary variable to distinguish between pre- (0) and post-policy (1) years;
- β_3 is the coefficient for the fixed effect interaction and represents the policy effect;
- $DID_{i,t}$ is the cross-product of A_i and T_t for a given site-time combination;
- ε is the error term.

$Y_{i,t} = \beta_0 + \beta_1 A_i + \beta_2 T_t + \beta_3 DID_{i,} + \beta_4 L_{i,t} + \varepsilon_{i,t}$ [Equation 4-4]

- Where:
- i and t represent the sample location and years;
- Y is the temperature-normalized mean PCB concentration (equation 2);
- β_0 is the regression intercept;
- β_1 is the coefficient for the sample site fixed effect;
- A_i is a binary variable to distinguish between control (0) and treatment (1) locations;
- β_2 is the coefficient for the year fixed effect;
- T_t is a binary variable to distinguish between pre- (0) and post-policy (1) years;
- β_3 is the coefficient for the lipid content;
- $L_{i,t}$ is the lipid content (%);
- B_4 is the coefficient for the fixed effect interaction and represents the policy effect;
- $DID_{i,t}$ is the cross-product of A_i and T_t for a given site-time combination;
- ε is the error term.

4.4 Empirical Results

Presented in greater detail elsewhere (e.g., Hites & Holsen, 2019; Salamova et al., 2015; Venier & Hites, 2010), PCB concentrations at each sample site are summarized below. For vapor-phase PCB concentrations, average non-normalized concentrations ranged from 46 - 1,068 pg/m^3. Chicago had the highest mean concentration (1,068 pg/m^3) followed by Cleveland (484 pg/m^3). Burnt Island had the lowest concentration (46 pg/m^3) with the remote U.S. sample locations being slightly higher (Sleeping Bear Dunes and Eagle Harbor with 82 pg/m^3 and 63 pg/m^3, respectively). Sturgeon Point had higher concentrations compared to Point Petre with 211 pg/m^3 and 95 pg/m^3, respectively. After temperature normalization (Equation 4-1), vapor-phase PCB concentrations were similar across all sample locations (Figure 4-2).

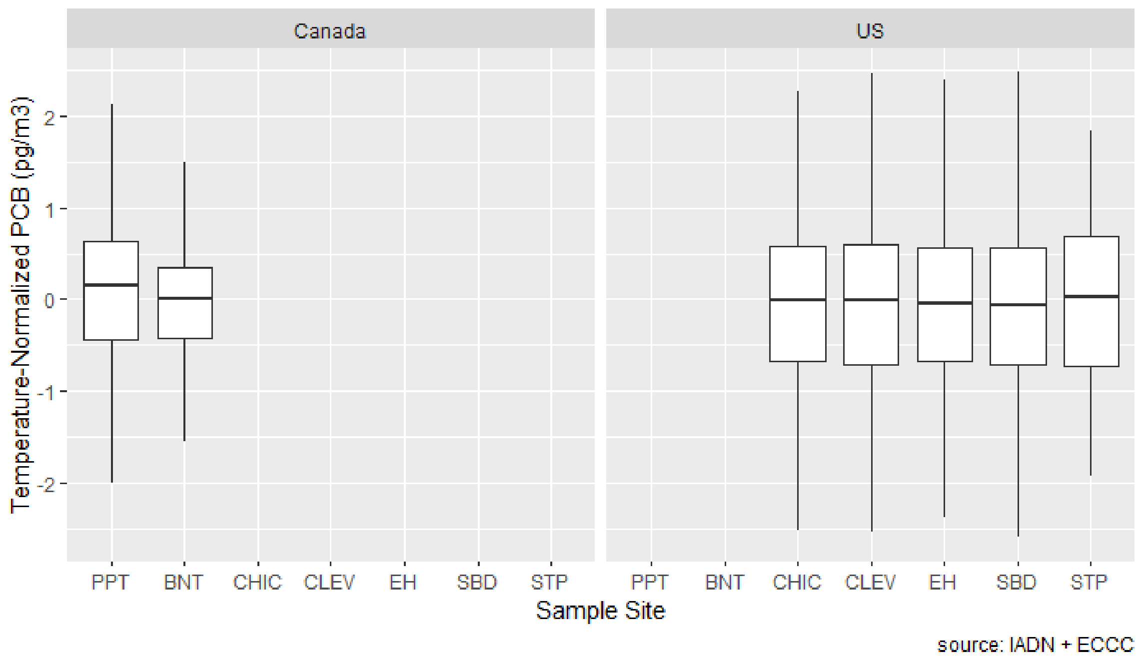

Figure 4-2. Box and whisker plot of vapor-phase PCB concentrations at seven sample locations in the Great Lakes basin. On the left panel, the first two sites are sample locations in the Canada (treatment sites). The five sites on the right are in the U.S. (control sites). Names of sample sites, abbreviations, and locations are in Figure 4-1. The lower and upper boxes correspond to the first and third quartiles. The whiskers extend to 1.5*IQR. Outliers have been removed.

Median lake trout PCB concentrations ranged from 100 – 1,500 ug/kg. Lake trout in Lake Ontario have the highest median lake trout PCB concentrations (545 – 1,500 ug/kg); four of the five highest PCB concentrations are at sites in Canadian waters. Lake trout at Whitefish Bay and Keweenaw Bay (Lake Superior) have the lowest median concentrations (43 - 800 and 50 - 496 ug/kg, respectively). Other sample sites in Lakes Superior, Huron, and Ontario have median concentrations that range from 150 - 1550 ug/kg. Lake trout from the Apostle Islands and Owen Sound (Lake Superior) have similar mean concentrations, 355 and 304 ug/kg, respectively. However, their median concentrations are different, 312 and 150 ug/kg, respectively. At Port Austin, Oswego, Marathon, Rockport, and Thunder Bay (Lake Huron) the PCB concentrations have varied median concentrations (300 - 688 ug/kg). Both Canadian sites (Thunder Bay and Marathon) are lower than the sites in U.S. waters of Lake Huron. With Age-normalized PCB concentrations, median concentrations vary among sample sites but there is much overlap in the range of concentrations among sites for lake trout (Figure 4-3).

In Lake Erie, walleye PCB concentrations range from 191 – 7,100 ug/kg. Median concentrations are higher in the Western Basin (Canada) compared to Middle Bass Island (U.S.), 900 and 515 ug/kg, respectively. Similar to lake trout, median Age-normalized PCB concentrations vary between walleye sample sites, but the range of measurements is similar (Figure 4-4).

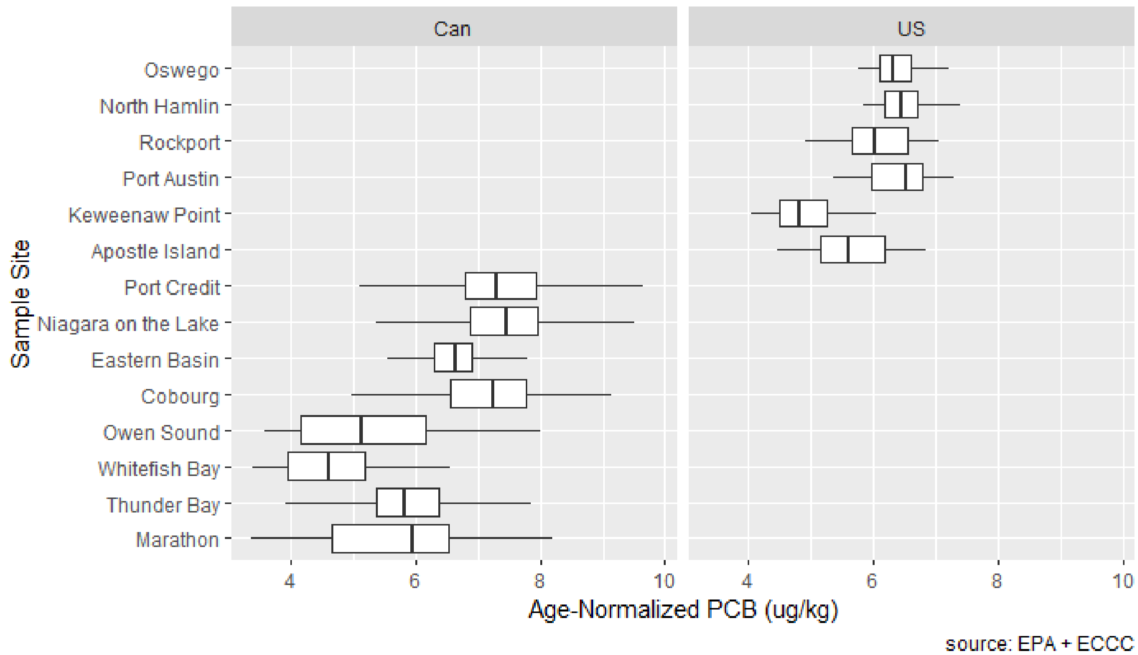

Figure 4-3. Box and whisker plot of lake trout PCB concentrations at 14 sample locations in the Great Lakes basin. On the left, the sample locations in Canada (treatment sites) and on the right are the U.S. (control sites). Names of sample sites, abbreviations, and locations are in Figure 4-1. The left and right boxes correspond to the first and third quartiles. The whiskers extend to 1.5*IQR. Outliers have been removed.

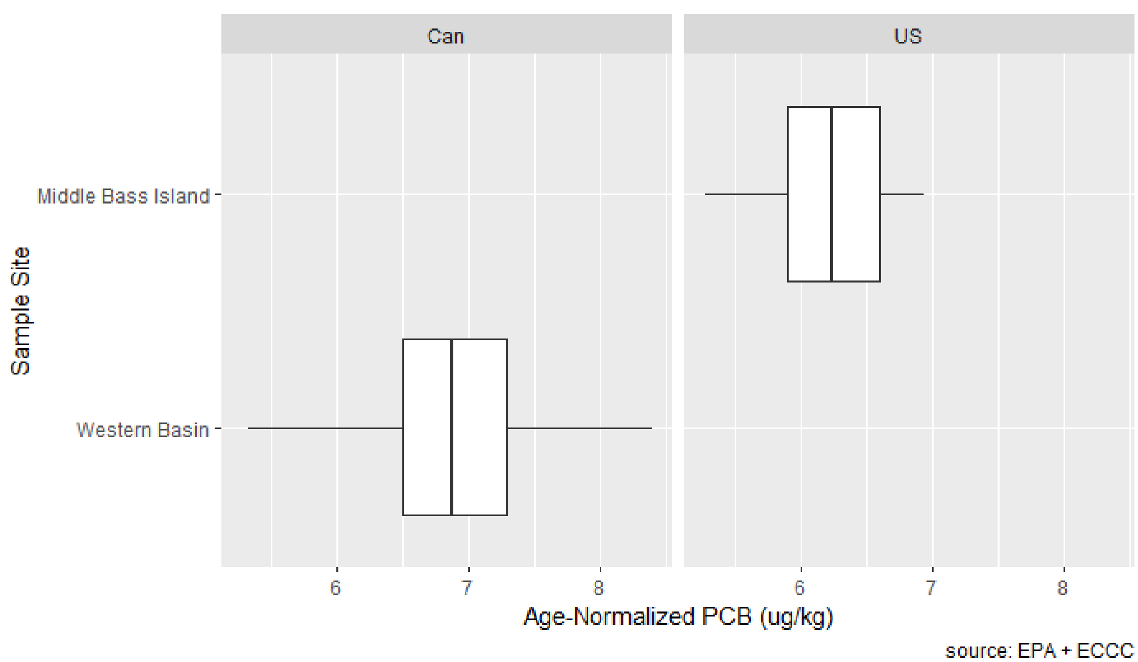

Figure 4-4. Box and whisker plot of walleye PCB concentrations at 2 sample locations in the Lake Erie. On the left, the sample locations in Canada (treatment sites) and on the right are the U.S. (control sites). Names of sample sites, abbreviations, and locations are

in Figure 4-1. The left and right boxes correspond to the first and third quartiles. The whiskers extend to 1.5*IQR. Outliers have been removed.

4.4.1 Vapor-phase difference-in-difference (DID)

Results from the standard difference-in-difference (DID) analysis show significant decreases in basin-wide vapor phase PCB concentrations because of Canada's 2008 PCB Regulations policy (SOR 2008-273; Table 4-2; $P < 0.001$). The event study regression is also referred to as a dynamic DID and weights policy effects (β_3) by year (ie., Time to Treatment) and presents more nuanced results; Figure 4-5 shows two relevant results. First, the Great Lakes basin-wide vapor-phase PCB data is suitable for DID analysis because there is no policy effect (i.e., coefficients are not different from 0) prior to policy implementation. There are five years where the coefficients are different from zero, but only two of the five are within five years of the policy implementation (TTT = 0; 2008). Second, after the policy goes into effect, the weighted coefficients indicate a heterogeneous response meaning that the policy effects are varied following policy implementation. In most years after the policy implementation, there is a significant policy effect (i.e., coefficients do not cross 0) except that sometimes the effect is positive (i.e., an increase in PCB concentrations attributed to the policy) and sometimes the effects are negative (i.e., a decrease in PCB concentrations attributed to the policy). Immediately after the policy went into effect there are five years of a positive or insignificant effect. Between 2013 and 2015 there are significant negative policy effects. From 2016 to 2019 there are significant positive policy effects.

Table 4-2. Standard 2x2 difference-in-difference (DID) results showing the DID estimate (β_3) which represents the impact of Canada's 2008 PCB Regulations Policy (SOR 2008-273) on vapor phase PCB concentrations in the Great Lakes basin. Sample locations are available in Figure 4-1.

Great Lakes Basin-wide Vapor Phase	**Estimate**	**Std. Error**	**P-value**
Intercept (β_0)	0.28	0.019	< 0.001
DID estimate (β_3)	-0.45	0.024	< 0.001
Site Fixed Effects	Yes		
Time Fixed Effects	Yes		

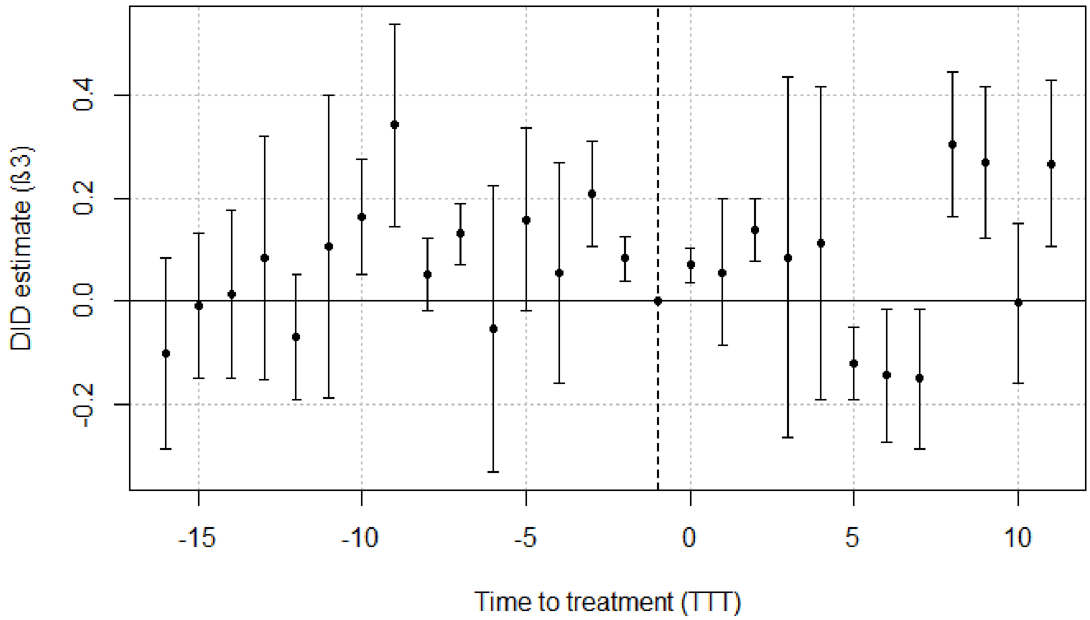

Figure 4-5. Event study regression plot, also referred to as a dynamic DID, for the Great Lakes basin-wide vapor phase PCBs. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.

Paired comparisons between Sturgeon Point (U.S.) and Point Petre (Canada) confirm the basin-wide results; there is a significant overall policy effect, but the impact is limited to 2013 – 2015 (Figure 4-6). In most years, the PCB policy effect (β_3) is not different from zero. However, similar to the basin-wide results, between 2013 and 2015 there are significant negative policy effects. Comparison between these sites offer a realistic representation of policy effect because both sample sites are located near or down-wind of where most PCB contamination exists in Ontario (Figure 4-1; Shu-Yin et al. 2018). Burnt Island, the other Canadian (treatment) site is located near the North Channel in Lake Huron, a much more remote location that is less impacted by Toronto's PCB contamination.

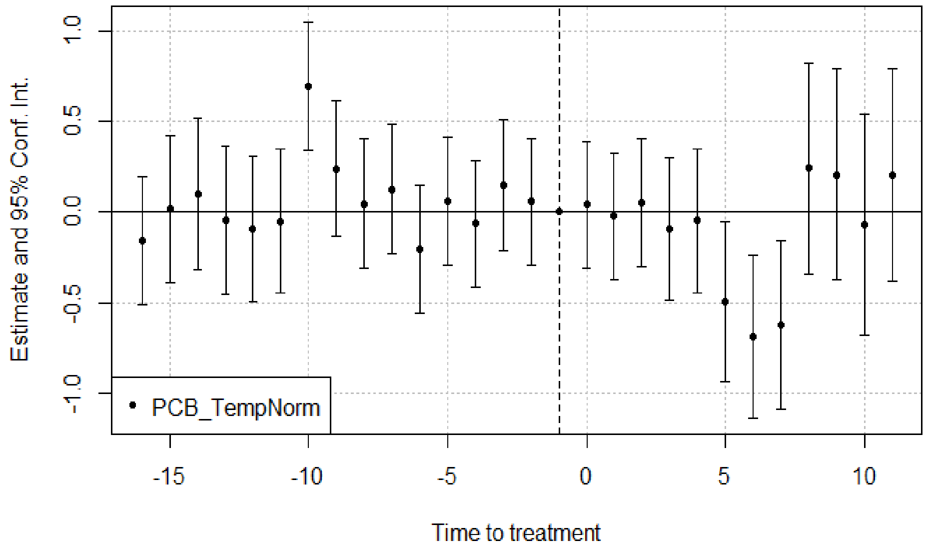

Figure 4-6. Event study regression plot, also referred to as a dynamic DID, for vapor phase PCB concentrations at Point Petre (Canada) and Sturgeon Point (U.S.; Figure 4-1). This site comparison was used as a robustness check to confirm the basin-wide results. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.

4.4.2 Fish tissue difference-in-difference (DID)

Results from the standard difference-in-difference (DID) analysis show significant increases in basin-wide lake trout PCB concentrations because of Canada's 2008 PCB Regulations policy (SOR 2008-273; Table 4-2Table 4-3 $P < 0.001$). The event study regression is also referred to as a dynamic DID and weights policy effects (β_3) by year (i.e., Time to Treatment) and presents more nuanced results; Figure 4-7 shows two relevant results. First, the Great Lakes basin-wide lake trout PCB data is suitable for DID analysis because there is no policy effect (i.e., coefficients are not different from 0) prior to policy implementation. There are six years where the coefficients are different from zero, but only one of the six are within five years of the policy implementation (TTT = 0; 2008). Second, after the policy goes into effect, the weighted coefficients indicate a heterogeneous response meaning that the policy effects are varied following policy implementation (Table S1). In most years after the policy implementation, there is not a significant policy effect. Between 2010 - 2012 there are significant negative policy effects. In the period 2013 - 2016, only 2014 shows significant effects and the policy effect is positive; other years in this period have insignificant policy effects.

Table 4-3. Standard 2x2 difference-in-difference (DID) results showing the DID estimate (β_3) which represents the impact of Canada's 2008 PCB Regulations Policy (SOR 2008-

273) on lake trout PCB concentrations in the Great Lakes basin. Sample locations are available in Figure 4-1.

Basin-wide Lake Trout	Estimate	Std. Error	P-value
Intercept (β_0)	706.3	131.8	8.7×10^{-8}
DID estimate (β_3)	632.9	133.4	2.1×10^{-6}
Site Fixed Effects	Yes		
Time Fixed Effects	Yes		

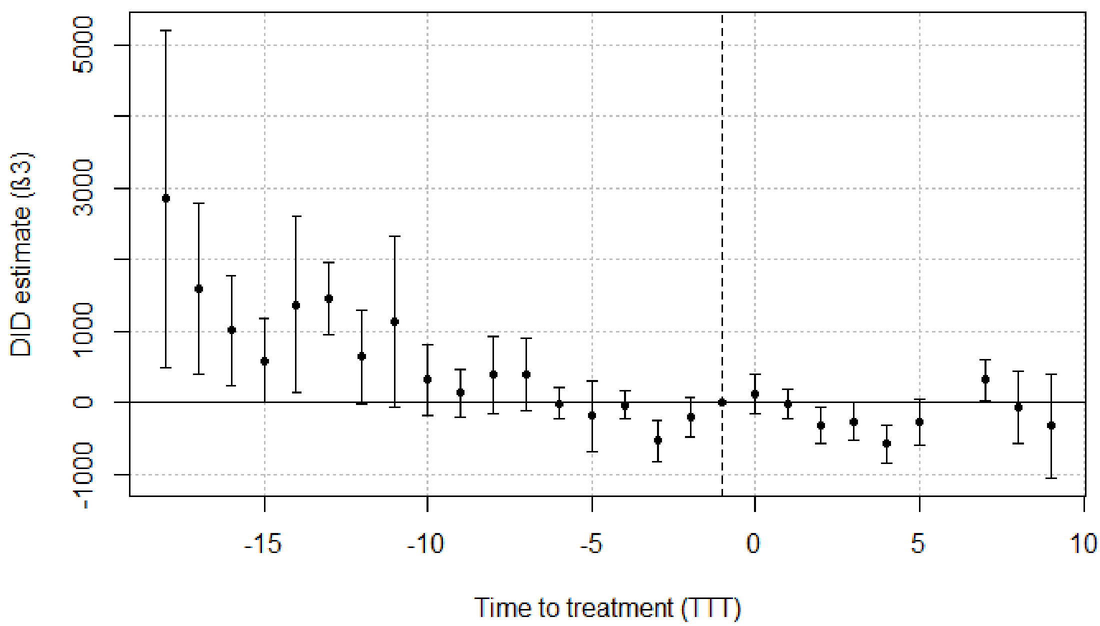

Figure 4-7. Event study regression plot, also referred to as a dynamic DID, for lake trout PCB concentrations within the Great Lakes basin. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.

Compared to basin-wide DID results, lake-specific DID analyses emphasize the heterogeneity of policy effects across space. In Lake Superior, the 2x2 DID shows that there is no significant policy effect reflected in lake trout tissue (Table 4-4; P = 0.876). The event study regression shows two things (Figure 4-8 and Table S7). First, the Lake Superior lake trout PCB data is suitable for DID analysis because there is no policy effect (i.e., coefficients are not different from 0) prior to policy implementation. There are seven years where the coefficients are different from zero, but only two of the seven are within the last five sample events prior to the policy implementation (TTT = 0; 2008). Second, after the policy goes into effect, the weighted coefficients indicate a relatively homogeneous lack of response to the policy. In most years the DID estimate (β_3) is not different from 0.

In Lake Ontario, the 2x2 DID shows that there is a significant policy effect reflected in lake trout tissue (Table 4-5; P = 0.0002). The event study regression shows two things (Figure 4-9 and Table S8). First, the Lake Ontario lake trout PCB data is suitable for DID analysis because there is no policy effect (i.e., coefficients are not different from 0) in most of the 10 years prior to policy implementation. There are four years where the coefficients are different from zero. Second, after the policy goes into effect, the weighted coefficients indicate a response similar to the vapor phase PCB results; there is no policy effect immediately after policy implementation, but there are significant negative effects for three years (2011 – 2013), preceded and followed by insignificant effects.

In Lake Huron (lake trout) and Lake Erie (walleye) the data are not sufficient for the analysis because of significant policy effects (i.e., non-zero DID coefficients) prior to 2008 (Table S5 and Table S6). Since this is prior to policy implementation DID coefficients are expected to be 0. Therefore, these results were not discussed as a part of the lake-specific analysis.

Table 4-4. Standard 2x2 difference-in-difference (DID) results showing the DID estimate (β_3) which represents the impact of Canada's 2008 PCB Regulations Policy (SOR 2008-273) on lake trout PCB concentrations in Lake Superior. Sample locations are available in Figure 4-1.

Lake Superior Lake Trout	**Estimate**	**Std. Error**	**P-value**
Intercept (β_0)	360.19	70.73	4.0×10^{-7}
DID estimate (β_3)	11.21	71.65	0.876
Site Fixed Effects	Yes		
Time Fixed Effects	Yes		

Table 4-5. Standard 2x2 difference-in-difference (DID) results showing the DID estimate (β_3) which represents the impact of Canada's 2008 PCB Regulations Policy (SOR 2008-273) on lake trout PCB concentrations in Lake Ontario. Sample locations are available in Figure 4-1.

Lake Ontario Lake Trout	**Estimate**	**Std. Error**	**P-value**
Intercept (β_0)	922.7	261.5	4.2×10^{-4}
DID estimate (β_3)	967.3	263.0	2.4×10^{-4}

Site Fixed Effects	Yes
Time Fixed Effects	Yes

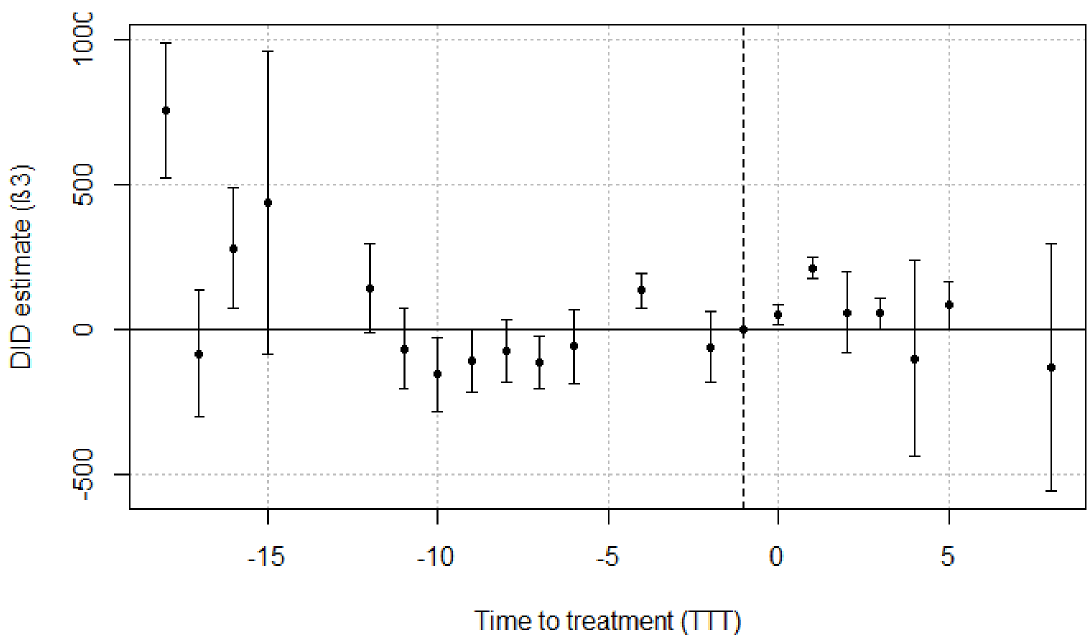

Figure 4-8. Event study regression plot, also referred to as a dynamic DID, for lake trout PCB concentrations in Lake Superior. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.

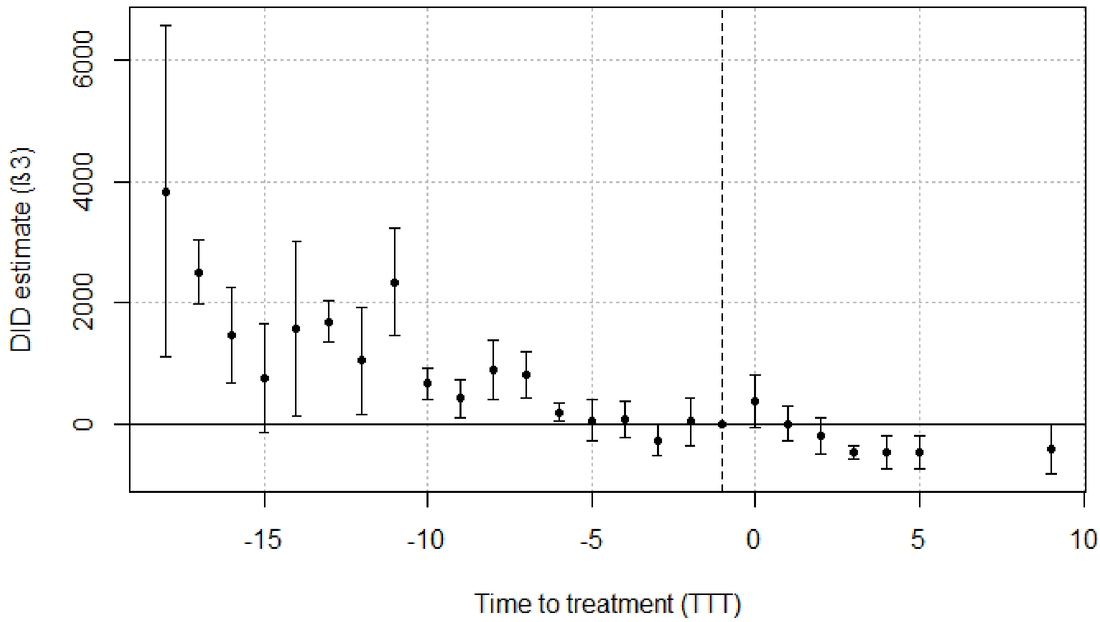

Figure 4-9. Event study regression plot, also referred to as a dynamic DID, for lake trout PCB concentrations in Lake Ontario. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.

4.5 Discussion

Policy evaluation is an important step in the policy process and aligning it with community values can represent a meaningful step towards shared governance of natural resources. Here, difference-in-difference (DID) shows that Canada's 2008 PCB Regulations (SOR-273) has had a significant effect on vapor-phase PCB concentrations and a significant positive effect on lake trout PCB concentrations in the Great Lakes basin. These results demonstrate the positive impact that localized action can have on transboundary pollutants in some environmental reservoirs (i.e., the atmosphere) while also drawing attention to the possibility of unequal policy impacts due to the lack of response in Great Lakes lake trout. Community engagement is necessary for robust policy evaluation.

4.5.1 Can local regulations of ASEPs significantly reduce local exposure to these contaminants, and, if so, in what environmental reservoirs?

Yes, local regulations of atmosphere-surface exchangeable pollutants (ASEPs) can reduce local exposure to contaminants (e.g., PCBs). Results from the standard 2x2 difference-in-difference (DID) analysis show an overall decrease in vapor-phase PCB concentrations that can be attributed to Canada's PCB Regulations (SOR 2008-273; Table 4-2). A comparison to the event study regression shows that the year-to-year policy effect is variable (Figure 4-5). Rather than contradicting the 2x2 DID results, the weighted event study reflects a heterogeneous policy effect. Recent work demonstrates the shortcomings of 2x2 DID analysis with such heterogeneous effects (Roth et al., 2022), but a comparison of basin-wide results with an analysis of Sturgeon Point (control site; U.S.) and Point Petre (treatment site; Canada) show similarities (Figure 4-6) and offer additional evidence that the significant effects are true. These sites were selected for comparison because they are located downwind of most PCB-containing waste in Ontario (Diamond et al. 2010; Shu-Yin et al. 2018) and are therefore most likely to reflect a policy influence. With only two treatment sites (Point Petre and Burnt Island) confirmation across many pairs of sites is impossible.

Trend analysis for Great Lakes PCB concentrations show atmospheric declines (Venier et al., 2014; Salamova et al., 2015), and there is evidence that the rate of decline in 2015 is faster than 1995 (Hites 2019). Results here confirm Hites' conclusion that the difference is the result of the Stockholm Convention; the decline in Great Lakes vapor-phase PCB concentrations can be attributed to Canada's PCB regulation that is aligned with the Stockholm Convention. With over 12,200 tonnes of PCB-containing waste and 32 tonnes of pure PCBs (99% of Canada's PCB inventory; Melymuk et al. 2022) removed, important sources of emissions are no longer released into the atmosphere. With a relatively short residence time, air masses in the Great Lakes basin are replenished in a few days. Therefore, reductions in PCB sources are seen quickly, too. A spatially uneven policy effect could be the result of the downwind/upwind position of the remediated site.

However, standard 2x2 DID results for the basin-wide lake trout PCB concentrations show an overall increase that can be attributed to Canada's PCB Regulations (Table 4-3). A comparison to the event study regression shows that, similar to the vapor-phase data, the year-to-year policy effect is variable (Figure 4-7). Rather than a causal increase in fish tissue PCB concentrations the systematic pattern in DID coefficients likely reflects that the model is currently unable to account for other time-varying factors. Although the fish tissue results are briefly discussed, future work will explore better ways to represent lake trout PCB concentrations.

Since fish integrate PCBs over their lifetime, especially early on, the lack of causative decreases in fish tissue concentrations that can be attributed to Canada's policy is unsurprising given that the median age of the fish studied was 5 (Barber et al., 1991). Further complicating the issue is that in Canada site-specific medians ranged from 4 – 6 years and in the U.S. the median age ranged from 4 – 9 years. Fish age ranged from 1 – 25 years with differences among sample sites. Lake trout PCB concentrations were age-normalized but there is still a correlation between length and PCB concentrations at many sample sites. Since length is often used as an age proxy, these results indicate that the age-normalization technique was not entirely effective. Future work will consider other age-normalization techniques to ensure PCB concentrations that are independent of growth. Another option is to use ln(PCB) concentrations with growth descriptors (i.e., age and/or length) as control variables. Preliminary analysis shows that the former might be an option because the systematic trend in pre-policy DID coefficients is reduced.

In addition to integrating PCBs over their lifetime, many fish species have wide home areas that might minimize the effects of localized remediation (Brooks et al., 2017). Some of the lake trout sample sites have known sediment contamination which might represent a source of PCBs to a localized food web (e.g., Whitefish Bay; Venier et al., 2014; Shaw and Urban 2022). Basin-wide trends in fish tissue PCBs appear to be declining (Bhavsar et al., 2007; Hites & Holsen, 2019; Zhou et al., 2018) but rates of decline have slowed (e.g., Visha et al., 2015) perhaps as a result of methodological differences and temporal that are spatially heterogeneous (Urban et al., 2020; Paterson et al., 2020). Future work will consider site-specific comparisons, similar to the robustness checks for the vapor-phase analysis. This analysis would be valuable because it would facilitate comparisons between sites with similar levels of known background contamination and might clarify relationships between lake trout PCB concentrations and Canada's SOR 2008-273.

Overall, there are two shortcomings that might influence these results. First, static two-way fixed effects models have difficulty handling spatially heterogeneous treatment effects (Roth 2022). Likely the influence is minimal because most of the PCB-containing waste that was removed was within Ontario, the province that borders the Great Lakes basin (Melymuk et al., 2022). Further, the influence of spatially heterogeneous treatment effects is dampened by the presence of only two treatment locations. As mentioned previously, Point Petre is downwind of most of the PCB-containing waste; Burnt Island is upwind. Therefore, we do not expect significant policy effects to be present at Burnt Island. Event study regressions for basin-wide and site-specific analysis were similar (Figure 4-5. Event study regression plot, also referred to as a dynamic DID, for the Great Lakes basin-wide vapor phase PCBs. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals. and Figure 4-6. Event study regression plot, also referred to as a dynamic DID, for vapor phase PCB concentrations at Point Petre (Canada) and Sturgeon Point (U.S.; Figure 4-1). This site comparison was used as a robustness check to confirm the basin-wide results. Negative values for Time to Treatment (x-axis) precede policy implementation and positive values are post policy; Time to Treatment = 0 is 2008. To eliminate collinearities, 2007 (Time to Treatment = -1) is used as a reference. The y-axis is the DID estimate (β_3) with 95% confidence intervals.) which supports the assumption that, in this study, there is minimal influence from spatially heterogeneous treatment effects.

Second, the assumption of no anticipatory effects (that is, no policy action preceded the policy implementation) also might influence the results. However, it is known that material was removed and stored in anticipation of a 2008 policy (Miriam Diamond, personal communication). While staggered policy implementation dates (e.g., anticipatory effects) can be considered in an event study regression, these data are not suitable (Sun & Abraham, 2021). Without details about the timing of waste removal at each location, it is impossible to set up a staggered response. Additionally, with only two treatment locations we were unable to experiment with user-defined staggered responses.

In the presence of anticipatory policy action, it can be reasonably assumed that the removal of PCB-containing waste prior to the policy date would have the effect of minimizing the influence of the policy. Since the vapor-phase DID analysis still shows significant declines as the result of the policy, there is a possibility that the actual policy effects are greater. To confirm these results additional vapor-phase PCB sample sites are needed.

4.6 Conclusions and policy implications

4.6.1 Conclusions

Results of this study offer evidence and hope that local action can have positive and significant outcomes related to transboundary pollutants. The difference-in-difference (DID) results presented here show that Canada's PCB Regulations (SOR 2008-273) had a significant impact on vapor-phase PCB concentrations. There is no corresponding reduction in lake trout PCB concentrations in the Great Lakes basin.

Simultaneously, we caution against exclusively local regulation as the sole policy for transboundary pollution. The global fate and transport of pollutants require international cooperation and action to fully address their hazardous existence in any one locale (Gagnon et al., 2018). As the primary producer and consumer of PCBs, the U.S. holds a unique responsibility for inventorying and disposing of PCB-containing materials. Despite the lack of response in Great Lakes lake trout, it remains possible that the magnitude of PCB-containing waste potentially eliminated by nation-wide reductions within the U.S. might be sufficient to demonstrate causal decreases in fish. Without meaningful reductions in lake trout PCB concentrations, fish consumers and Indigenous Nations within the Great Lakes basin continue to be at a greater health risk from fish consumption. Finally, results here demonstrate that the environmental reservoir considered when defining policy success presents an opportunity for environmental justice/injustice.

4.6.2 Policy implications

Valuing landscapes for the relationships they sustain offers a lens for policy evaluation that extends beyond economic impacts. Adapting to major environmental shifts (e.g., climate change and chemical contamination) can be more effective when Tribal experts are able to lead and support community priorities (Schramm et al., 2020a; Schramm, et al., 2020b). Because of place-based cultural practices, Indigenous, Tribal, and First Nations often value landscapes beyond a monetary framework (e.g., Hoover, 2013; Poe et al., 2016). Results from this work demonstrate the importance of policy evaluation metrics relevant to the primary concern, that might vary among communities and be closely tied to values.

Despite an ability to reduce local exposures to contaminants via outdoor air pollution, this source represents a minor exposure pathway for both adults and children to PCBs (Weitekamp et al., 2021). Both indoor air pollution and dietary ingestion are significantly more important. Indoor air pollution, in schools and workplaces, is an important pathway for both adults and children, and is by far the most important pathway for children (e.g., Bannavti et al., 2021; Young et al., 2021). In adults, dietary intake (e.g., fish consumption) is the primary exposure pathway. Fish consumption varies greatly, with people living in the Great Lakes basin consuming above average amounts of fish (U.S. EPA). Tribal and First Nations within the basin consume fish at even higher rates (e.g., Asher Consulting 2016).

For communities that rely heavily on fish consumption for subsistence and for cultural practices, the policy failed to alleviate the risk associated with exposure via fish consumption. Tribal and First Nations within the Great Lakes basin have place-based knowledge systems that are built and maintained through landscape practices, including harvesting and consuming fish (Cajete, 1994; Watts, 2013). Chemical contamination disrupts these practices (van Horne et al., 2021). Further, many Tribal and First Nations have higher fish consumption rates compared to the national averages in either the United States or Canada (e.g., Asher Consulting LLC 2016) and are therefore at greater risk to PCB exposure.

Transboundary pollution (e.g., PCBs) is often distributed far from the original source in uneven and inequitable patterns. Despite evidence that local regulations can impact local exposure to ASEPs, action at many levels is necessary to protect human health and the environment (Gorman et al., 2016). The U.S. was the primary producer and consumer of PCBs that were used globally. By not taking action to manage PCB stocks using environmentally sound management (ESM) strategies, the U.S. puts the regulatory burden on countries that contributed less PCB waste to the environment. While a complete PCB inventory in the U.S. does not exist, previous work shows that within the Great Lakes basin the U.S. has twice as much PCB-containing waste and pure PCB mass and twice as many sites with PCB-contaminated waste (Diamond et al., 2010; Melymuk et al., 2022). That an improvement in vapor-phase PCB concentrations was clear following a nation-wide policy in Canada lends support to the idea that further PCB stock reductions in the Great Lakes basin could have a greater impact.

With respect to chemical policy, consideration of cumulative risk can capture the landscape practices (e.g., harvesting and consuming fish) that maintain relationships with a place. Afterall, "all policy is health policy" (Hardeman et al., 2020). To effectively engage with Indigenous Peoples, communities must be meaningfully consulted throughout the policy process, especially at the beginning. When outsiders propose policy "solutions" for Indigenous Communities without their input, the effects can be "unruly" (Lea, 2020). That is, unintended but foreseeable consequences can result from a misunderstanding of local conditions. Therefore, environmental policy that considers cumulative risks must focus on health impacts – the prevention of negative impacts and the promotion of positive impacts. However, appropriately quantifying risks is complex as risk can vary greatly between and among populations and communities.

Evaluating policy efficacy, at a minimum, must consider whether the policy effectively addresses its primary objective. For SOR 2008-273, the policy is in response to the Stockholm Convention, a global treaty to protect human health and the environment from persistent environmental chemicals (e.g., PCBs), and has the objective of destroying or properly storing all PCB-containing materials with concentrations greater than 50 mg/kg. Recent work demonstrated that Canada has made major significant progress towards their goal and has successfully eliminated 99% of PCB-containing waste (Melymuk 2022). This secondary evaluation of SOR 2008-273 considers its efficacy with respect to the Stockholm Convention priorities of protecting human health and the environment.

4.7 Chapter 4 bibliography

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4.8 Chapter 4 supplementary information

Table 4-S1. Event study (dynamic difference-in-difference (DID)) regression results for temperature-normalized vapor-phase PCB concentrations in the Great Lakes. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

```
> esttable(ES_vaporTemp)
                             ES_vaporTemp
Dependent Var.:              PCB_TempNorm

(Intercept)              103.4*** (10.11)
Treat x TTT = -16        -0.1015 (0.0940)
Treat x TTT = -15        -0.0095 (0.0713)
Treat x TTT = -14         0.0127 (0.0830)
Treat x TTT = -13         0.0834 (0.1206)
Treat x TTT = -12        -0.0690 (0.0617)
Treat x TTT = -11         0.1056 (0.1499)
Treat x TTT = -10        0.1627* (0.0570)
Treat x TTT = -9         0.3413* (0.1006)
Treat x TTT = -8          0.0522 (0.0357)
Treat x TTT = -7        0.1303** (0.0306)
Treat x TTT = -6         -0.0530 (0.1419)
Treat x TTT = -5          0.1582 (0.0901)
Treat x TTT = -4          0.0555 (0.1094)
Treat x TTT = -3        0.2074** (0.0519)
Treat x TTT = -2         0.0820* (0.0225)
Treat x TTT = 0         0.0691** (0.0173)
Treat x TTT = 1           0.0560 (0.0728)
Treat x TTT = 2         0.1384** (0.0312)
Treat x TTT = 3           0.0849 (0.1790)
Treat x TTT = 4           0.1122 (0.1551)
Treat x TTT = 5         -0.1223* (0.0357)
Treat x TTT = 6         -0.1443. (0.0662)
Treat x TTT = 7         -0.1506. (0.0690)
Treat x TTT = 8         0.3048** (0.0720)
Treat x TTT = 9          0.2691* (0.0753)
Treat x TTT = 10         -0.0042 (0.0788)
Treat x TTT = 11         0.2662* (0.0824)
SiteCLEV               0.1589*** (0.0188)
SiteEH                -0.0738*** (0.0105)
SitePPT                 -0.1488* (0.0560)
SiteSBD               -0.0825*** (0.0088)
SiteSTP               -0.0941*** (0.0121)
SiteBNT               -0.3495*** (0.0514)
Year                  -0.0515*** (0.0050)
__________________    ___________________
S.E.: Clustered                  by: Site
Observations                        4,924
R2                                0.18932
Adj. R2                           0.18368
---
Signif. codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

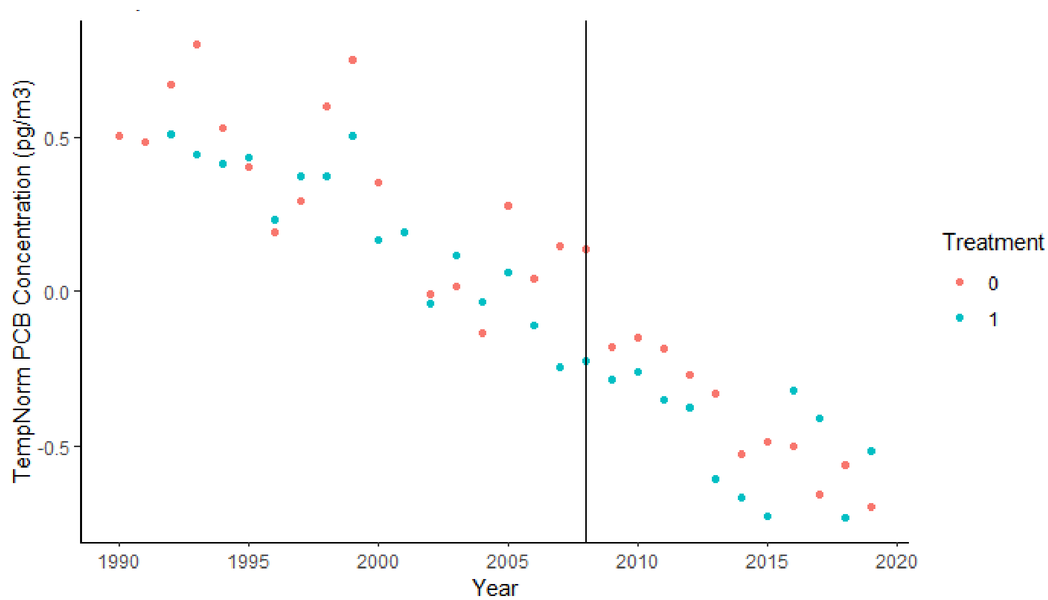

Figure 4-S1. Annual average temperature-normalized vapor-phase PCB concentrations for treatment (1; Canadian) and control (0; U.S.) sample locations. The parallel trend assumption is an important assumption for the standard 2x2 difference-in-difference (DID) analysis.

Table 4-S2. Event study (dynamic difference-in-difference (DID)) regression results for temperature-normalized vapor-phase PCB concentrations at Point Petre and Sturgeon Point (see Figure 4-1 for sample locations). These data were used as a robustness check for the vapor-phase DID. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

	ES_vaporPS
Dependent Var.:	PCB_TempNorm
(Intercept)	103.1*** (3.37e-9)
Treat x TTT = -16	-0.1595*** (2.52e-11)
Treat x TTT = -15	0.0185*** (2.36e-11)
Treat x TTT = -14	0.0985*** (2.19e-11)
Treat x TTT = -13	-0.0423*** (2.02e-11)
Treat x TTT = -12	-0.0930*** (1.85e-11)
Treat x TTT = -11	-0.0502*** (1.68e-11)
Treat x TTT = -10	0.6951*** (1.51e-11)
Treat x TTT = -9	0.2386*** (1.35e-11)
Treat x TTT = -8	0.0468*** (1.18e-11)
Treat x TTT = -7	0.1260*** (1.01e-11)
Treat x TTT = -6	-0.2044*** (8.41e-12)
Treat x TTT = -5	0.0618*** (6.73e-12)
Treat x TTT = -4	-0.0633*** (5.05e-12)
Treat x TTT = -3	0.1513*** (3.37e-12)
Treat x TTT = -2	0.0565*** (1.68e-12)
Treat x TTT = 0	0.0403*** (1.68e-12)
Treat x TTT = 1	-0.0228*** (3.36e-12)
Treat x TTT = 2	0.0546*** (5.05e-12)
Treat x TTT = 3	-0.0963*** (6.73e-12)
Treat x TTT = 4	-0.0459*** (8.41e-12)
Treat x TTT = 5	-0.4976*** (1.01e-11)
Treat x TTT = 6	-0.6855*** (1.18e-11)
Treat x TTT = 7	-0.6251*** (1.35e-11)
Treat x TTT = 8	0.2438*** (1.51e-11)
Treat x TTT = 9	0.2080*** (1.68e-11)
Treat x TTT = 10	-0.0655*** (1.85e-11)
Treat x TTT = 11	0.2048*** (2.02e-11)
SitePPT	0.0551*** (2.71e-12)
Year	-0.0514*** (1.68e-12)
S.E.: Clustered	by: Site
Observations	1,717
R2	0.23068
Adj. R2	0.21745

Table 4-S3. Event study (dynamic difference-in-difference (DID)) regression results for temperature-normalized vapor-phase PCB concentrations in the Great Lakes, excluding Chicago and Cleveland. This test acted as a robustness check for the validity of the vapor-phase DID results. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

	ES_vaporTemp
Dependent Var.:	PCB_TempNorm
(Intercept)	112.3*** (4.499)
Treat x TTT = -16	-0.1852* (0.0542)
Treat x TTT = -15	-0.0699 (0.0329)
Treat x TTT = -14	-0.0210 (0.0450)
Treat x TTT = -13	0.0319 (0.1121)
Treat x TTT = -12	-0.1163* (0.0380)
Treat x TTT = -11	0.0598 (0.1480)
Treat x TTT = -10	0.4721. (0.1787)
Treat x TTT = -9	0.3053* (0.0974)
Treat x TTT = -8	0.0199 (0.0166)
Treat x TTT = -7	0.1029** (0.0143)
Treat x TTT = -6	-0.0765 (0.1449)
Treat x TTT = -5	0.1396 (0.0913)
Treat x TTT = -4	0.0415 (0.1124)
Treat x TTT = -3	0.1987* (0.0530)
Treat x TTT = -2	0.0584*** (0.0057)
Treat x TTT = 0	0.0466*** (0.0024)
Treat x TTT = 1	0.0482 (0.0586)
Treat x TTT = 2	0.0966* (0.0266)
Treat x TTT = 3	0.1005 (0.1830)
Treat x TTT = 4	0.1326 (0.1577)
Treat x TTT = 5	-0.5022** (0.0629)
Treat x TTT = 6	-0.6076*** (0.0453)
Treat x TTT = 7	-0.5425*** (0.0461)
Treat x TTT = 8	0.3311** (0.0471)
Treat x TTT = 9	0.3001** (0.0481)
Treat x TTT = 10	0.0313 (0.0491)
Treat x TTT = 11	0.3063** (0.0503)
SiteEH	0.2919** (0.0415)
SitePPT	0.2371*** (0.0012)
SiteSBD	0.2825** (0.0414)
SiteSTP	0.2191** (0.0415)
Year	-0.0561*** (0.0022)
________________	___________________
S.E.: Clustered	by: Site
Observations	3,909
R2	0.24234
Adj. R2	0.23609

Table 4-S4. Event study (dynamic difference-in-difference (DID)) regression results for age-normalized lake trout PCB concentrations in the Great Lakes basin. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

```
OLS estimation, Dep. Var.: PCB_AgeNorm1
Observations: 5,736
Standard-errors: Clustered (Station)
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
RMSE: 1,007.7   Adj. R2: 0.574701
> esttable(ES_troutAge)
                                                                 ES_troutAge
Dependent Var.:                                                 PCB_AgeNorm1

(Intercept)                                          58,142.5*** (7,217.9)
Lipid_pct                                                  14.60 (8.448)
Treat x TTT = -18                                     2,849.6* (1,206.9)
Treat x TTT = -17                                       1,593.3* (612.1)
Treat x TTT = -16                                       1,008.8* (398.0)
Treat x TTT = -15                                         585.4. (298.7)
Treat x TTT = -14                                       1,373.7* (632.4)
Treat x TTT = -13                                     1,458.9*** (257.7)
Treat x TTT = -12                                         639.7. (336.5)
Treat x TTT = -11                                       1,137.6. (610.4)
Treat x TTT = -10                                          316.7 (249.1)
Treat x TTT = -9                                           140.2 (171.3)
Treat x TTT = -8                                           385.9 (273.9)
Treat x TTT = -7                                           391.8 (261.4)
Treat x TTT = -6                                          -11.99 (109.9)
Treat x TTT = -5                                          -187.7 (254.3)
Treat x TTT = -4                                          -28.77 (105.4)
Treat x TTT = -3                                        -534.0** (142.0)
Treat x TTT = -2                                          -202.8 (138.3)
Treat x TTT = 0                                            121.1 (145.2)
Treat x TTT = 1                                           -25.65 (105.8)
Treat x TTT = 2                                          -315.1* (124.5)
Treat x TTT = 3                                          -258.8. (136.0)
Treat x TTT = 4                                        -579.0*** (130.4)
Treat x TTT = 5                                           -268.7 (164.1)
Treat x TTT = 7                                           320.2. (149.2)
Treat x TTT = 8                                           -64.22 (254.5)
Treat x TTT = 9                                           -323.6 (368.6)
StationLakeOntario,CentralBasin,PortHope-Cobourg,Ontario    1,296.0*** (46.57)
StationLakeOntario,EasternBasin,OswegoNY              1,062.2*** (103.4)
StationLakeOntario,NiagaraontheLake,Ontario           1,651.0*** (54.32)
StationLakeOntario,PortCredit,Ontario                 1,693.9*** (55.80)
StationLakeSuperior,Marathon,Ontario                    741.8*** (99.54)
StationLakeSuperior,WhitefishBay                          -62.71 (83.19)
StationLH-PortAustin                                    843.7*** (150.8)
StationLH-Rockport                                      669.6*** (151.2)
StationLO-NorthHamlin                                   796.1*** (145.8)
StationLO-Oswego                                        756.6*** (147.3)
StationLS-ApostleIsland                                  577.0** (157.8)
StationLS-KeweenawPoint                                   379.9* (155.9)
StationThunderBay-PieIsland                               -118.0 (74.25)
Year                                                    -29.10*** (3.582)
_______________________________________               ____________________

S.E.: Clustered                                                  by: Station
Observations                                                           5,736
R2                                                                   0.57774
Adj. R2                                                              0.57470
```

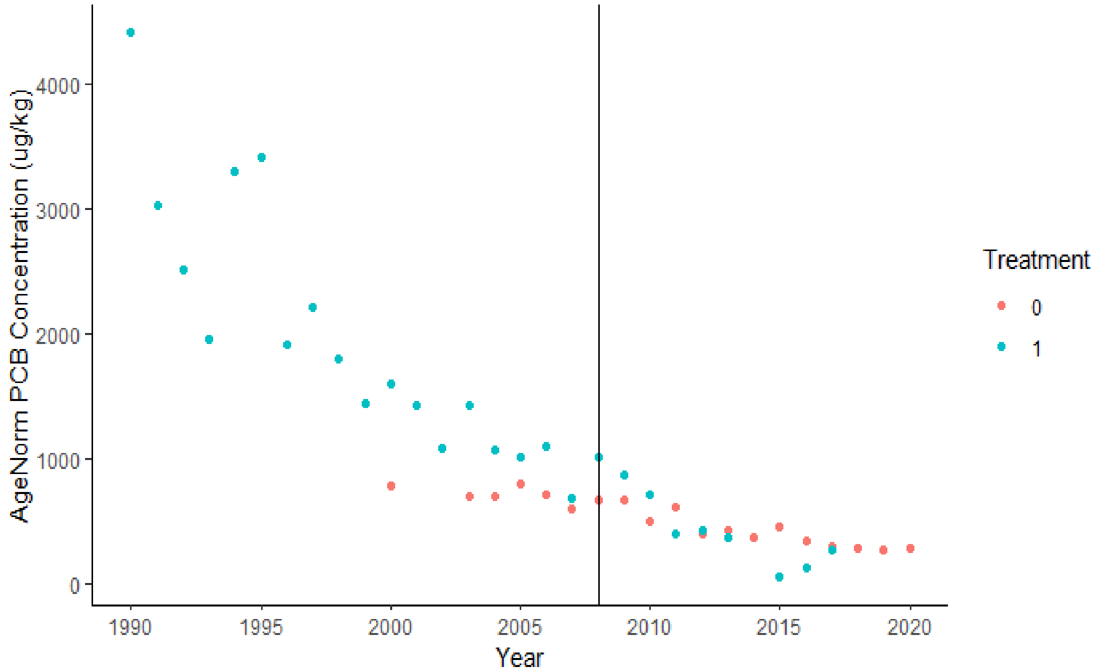

Figure 4-S2. Annual average age-normalized lake trout PCB concentrations for treatment (1; Canadian) and control (0; U.S.) sample locations. The parallel trend assumption is an important assumption for the standard 2x2 difference-in-difference (DID) analysis.

Table 4-S5. Event study (dynamic difference-in-difference (DID)) regression results for age-normalized lake trout PCB concentrations in Lake Ontario. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects. However, since there are significant DID coefficients prior to policy implementation, these data are not suitable for a lake-specific analysis.

	ES_troutHurAge
Dependent Var.:	PCB_AgeNorm1
(Intercept)	64,058.4** (3,266.7)
Lipid_pct	1.181 (2.529)
Treat x TTT = -21	-260.8* (29.59)
Treat x TTT = -17	75.88. (22.40)
Treat x TTT = -16	-53.06 (20.27)
Treat x TTT = -15	307.8** (18.34)
Treat x TTT = -12	75.64. (18.73)
Treat x TTT = -11	-94.66* (21.72)
Treat x TTT = -9	-161.2** (11.92)
Treat x TTT = -8	-194.9** (9.830)
Treat x TTT = -7	-162.8** (11.24)
Treat x TTT = -4	-217.1** (8.063)
Treat x TTT = 2	-32.42 (20.98)
Treat x TTT = 3	-23.22 (13.71)
Treat x TTT = 5	37.30 (17.59)
Treat x TTT = 7	87.52* (13.29)
Treat x TTT = 9	173.1** (16.27)
StationLH-PortAustin	575.7*** (10.43)
StationLH-Rockport	398.7*** (10.74)
Year	-31.82** (1.640)
S.E.: Clustered	by: Station
Observations	714
R2	0.69221
Adj. R2	0.68378

Table 4-S6. Event study (dynamic difference-in-difference (DID)) regression results for age-normalized walleye PCB concentrations in Lake Erie. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects. However, since there are significant DID coefficients prior to policy implementation, these data are not suitable for a lake-specific analysis.

	ES_wallErieAge
Dependent Var.:	PCB_AgeNorm1
(Intercept)	88,698.4* (2,723.9)
Lipid_pct	-67.85. (8.175)
Treat x TTT = -4	897.4** (13.79)
Treat x TTT = -2	730.8*** (0.3199)
Treat x TTT = 0	-336.2* (5.688)
Treat x TTT = 1	92.34. (11.74)
Treat x TTT = 2	47.29. (4.634)
Treat x TTT = 3	335.0** (1.606)
Treat x TTT = 4	137.2** (0.7064)
Treat x TTT = 6	888.4** (10.62)
StationLE-MiddleBassIsland	-255.1** (3.035)
Year	-43.37* (1.316)
S.E.: Clustered	by: Station
Observations	466
R2	0.43390
Adj. R2	0.42018

Table 4-S7. Event study (dynamic difference-in-difference (DID)) regression results for age-normalized lake trout PCB concentrations in Lake Superior. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

	ES_troutSupAge
Dependent Var.:	PCB_AgeNorm1
(Intercept)	37,257.9. (13,721.4)
Lipid_pct	8.190*** (0.2485)
Treat x TTT = -28	147.2 (185.5)
Treat x TTT = -27	-358.9 (187.3)
Treat x TTT = -26	-143.0 (172.3)
Treat x TTT = -25	141.2 (188.2)
Treat x TTT = -24	-175.7 (169.2)
Treat x TTT = -23	82.58 (152.0)
Treat x TTT = -22	-168.5 (144.5)
Treat x TTT = -21	-129.9 (138.8)
Treat x TTT = -20	1,456.6*** (131.6)
Treat x TTT = -19	92.83 (141.7)
Treat x TTT = -18	772.6** (118.1)
Treat x TTT = -17	-92.97 (111.6)
Treat x TTT = -16	290.6* (104.5)
Treat x TTT = -15	449.7 (259.2)
Treat x TTT = -12	145.4 (77.76)
Treat x TTT = -11	-73.43 (72.70)
Treat x TTT = -10	-169.7. (65.14)
Treat x TTT = -9	-118.1 (56.91)
Treat x TTT = -8	-87.95 (56.58)
Treat x TTT = -7	-103.5. (46.04)
Treat x TTT = -6	-61.13 (68.19)
Treat x TTT = -4	148.1** (29.48)
Treat x TTT = -2	-63.32 (63.18)
Treat x TTT = 0	40.42* (13.97)
Treat x TTT = 1	194.9*** (14.30)
Treat x TTT = 2	42.85 (65.04)
Treat x TTT = 3	44.38 (30.34)
Treat x TTT = 4	-130.3 (155.4)
Treat x TTT = 5	69.34 (48.59)
Treat x TTT = 8	-163.4 (199.8)
StationLakeSuperior,WhitefishBay	-471.7*** (40.53)
StationLS-ApostleIsland	-143.4 (68.84)
StationLS-KeweenawPoint	-339.8** (69.74)
StationThunderBay-PieIsland	-369.7** (63.87)
Year	-18.32. (6.836)
S.E.: Clustered	by: Station
Observations	1,978
R2	0.47685
Adj. R2	0.46715

Signif. codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Table 4-S8. Event study (dynamic difference-in-difference (DID)) regression results for age-normalized lake trout PCB concentrations in Lake Ontario. Each treatment-TTT interaction estimates a weight for the DID coefficient. These tables are useful to understand when the policy had the greatest impact. Treatment is a binary variable (0 or 1 for Canadian and U.S. samples, respectively) and TTT is time to treatment, a time variable. Sample station and year are included as fixed effects.

	ES_troutOntAge
Dependent Var.:	PCB_AgeNorm1
(Intercept)	75,650.7*** (315.3)
Lipid_pct	19.07* (5.974)
Treat x TTT = -31	1,210.5* (347.5)
Treat x TTT = -30	7,073.1*** (594.1)
Treat x TTT = -29	2,377.2. (993.8)
Treat x TTT = -28	2,567.3. (1,147.5)
Treat x TTT = -27	936.3 (470.4)
Treat x TTT = -26	4,071.6* (1,473.2)
Treat x TTT = -25	3,873.5. (1,806.7)
Treat x TTT = -24	3,094.4. (1,525.4)
Treat x TTT = -23	1,136.7. (520.6)
Treat x TTT = -22	3,088.1** (667.1)
Treat x TTT = -21	3,543.8. (1,650.2)
Treat x TTT = -20	1,501.9** (276.3)
Treat x TTT = -19	1,085.5 (1,097.4)
Treat x TTT = -18	3,726.3* (1,392.0)
Treat x TTT = -17	2,408.1*** (282.6)
Treat x TTT = -16	1,360.7* (356.5)
Treat x TTT = -15	597.7 (444.8)
Treat x TTT = -14	1,502.1. (700.2)
Treat x TTT = -13	1,556.7*** (189.9)
Treat x TTT = -12	932.8 (474.3)
Treat x TTT = -11	2,213.3** (442.0)
Treat x TTT = -10	578.1* (158.9)
Treat x TTT = -9	285.6 (222.0)
Treat x TTT = -8	838.3* (248.7)
Treat x TTT = -7	738.4* (241.2)
Treat x TTT = -6	94.89 (113.3)
Treat x TTT = -5	-57.38 (238.1)
Treat x TTT = -4	82.79 (107.9)
Treat x TTT = -3	-403.5* (106.1)
Treat x TTT = -2	-53.37 (199.2)
Treat x TTT = 0	211.1 (363.8)
Treat x TTT = 1	-116.6 (196.7)
Treat x TTT = 2	-289.1 (194.9)
Treat x TTT = 3	-599.1** (94.47)
Treat x TTT = 4	-605.0** (149.4)
Treat x TTT = 5	-555.5* (157.1)
Treat x TTT = 9	-312.3 (363.7)
StationLakeOntario,EasternBasin,OswegoNY	342.1 (233.7)
StationLakeOntario,NiagaraontheLake,Ontario	502.5*** (49.11)
StationLakeOntario,PortCredit,Ontario	1,074.4*** (102.3)
StationLO-NorthHamlin	44.02 (304.5)
StationLO-Oswego	11.77 (310.5)
Year	-37.47*** (0.1647)
S.E.: Clustered	by: Station
Observations	4,951
R2	0.53425
Adj. R2	0.53008

Signif. codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

A Spatial Analysis

Using National Lake Assessment (NLA) lakes, inverse distance weighting and geographic weighted regression (GWR) were used to predict dissolved organic carbon (DOC), total nitrogen (NTL), total phosphorus (PTL), and chlorophyll (CHL) in unmonitored National Lake Fish Tissue Survey (NLFTS) lakes. We trained the models using 70% of the NLA lakes (n = ~809). For IDW, 3 lakes were excluded because of missing water quality measurements. For GWR, 13 lakes were excluded because of missing lake area data. (Lake area was used to calculate lake volume, a significant variable in all GWR models.)

Each model was independently validated using 30% of NLA lakes which were excluded from model development. We calculated the RMSE for each model, selecting the one with the lowest RMSE for inclusion in the correlation analysis. For each variable, the IDW interpolation performed the best (i.e., the lowest RMSE).

Table A1. RMSE based on log10() measurements and WQ predictions for the three models considered here. These RMSEs are based on the ~347 validation lakes.

	DOC	NTL	PTL	CHL
IDW	0.01	0.36	0.544	1.27
GWR2A	1.17	0.64	2.06	1.30
GWR2F	1.18	0.66	2.09	1.36

A.1 National Lake Assessment Data

All four water quality parameters - dissolved organic carbon (DOC), total nitrogen (NTL), total phosphorus (PTL), and chlorophyll (CHL) are log-normal and linear relationships exist between the measured water quality variables.

The Average Nearest Neighbor (ANN) analyzes the spatial distribution of the x,y location and considers pairs of points. This is a global statistic and is sensitive to area. The null hypothesis says that samples are randomly distributed. For the NLA data the nearest neighbor ratio is 0.67, indicating clustered sampling ($p < 0.001$).

Getis-Ord High Low Clustering is another global statistic that evaluates whether attributes within the dataset are clustered. The test works best when evaluating high or low clusters, not both. For the 4 water quality parameters (DOC, NTL, PTL, CHL) were randomly distributed. Here we show the results for inverse distance weighted results for DOC using IDW-squared is also random. This is true for the other WQ variables, too. The probability values refer to the likelihood that the observed spatial pattern is due to a random distribution. The z-scores standardize the deviation from the mean. Very high or very low z-scores are associated with small p-values that fall in the tails of the distribution; together they indicate that it is unlikely that the distribution is not random which leads us to reject H0 (that samples are randomly distributed).

Moran's I spatial autocorrelation is another global statistic that evaluates whether the values of an attribute is dispersed, random, or clustered. This approach generalizes the data because it measures deviation from the mean. Here both inverse distance weighting and IDW-squared were considered for all 4 WQ variables. In all instances, the test indicates a random distribution for each variable.

A.2 Water Quality Modeling

Two modeling approaches were considered to predict water quality in unmonitored lakes. Models were trained using 70% of the original data (n ~ 806). Models were validated using the remaining 30% of the NLA lakes (i.e., the validation lakes, n ~ 346). To decide which model was the best, we evaluated the root-mean square error (RMSE) and selected the model with the lowest RMSE.

A.2.1 Inverse Distance Weighting – water quality interpolation

A.2.2 Exploratory Ordinary Least Squares

Dissolved Organic Carbon (DOC)

To interpolate DOC we included 806 lakes (i.e., training lakes). The power was 1.1115. Neighborhood searching method was standard and included up to 15 neighbors with at least 10 neighbors. Full sector type. No angle with a major and minor semi-axis of 1,295,791.913. A map of error appears random.

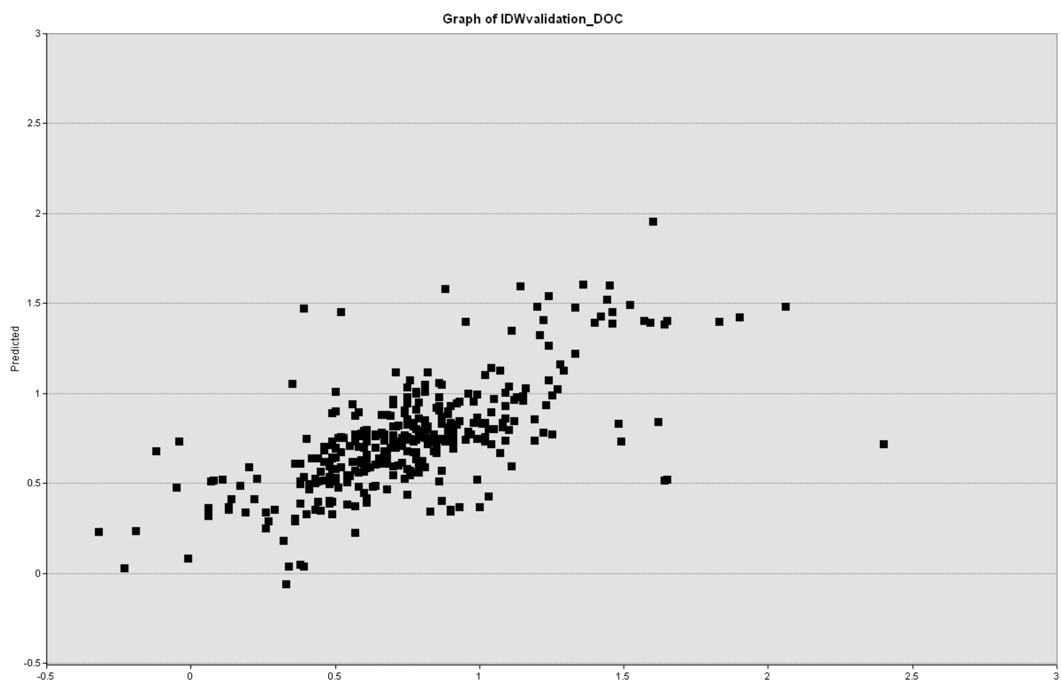

Figure A1. Measured v predicted Log10(DOC) concentrations using the 347 validation lakes.

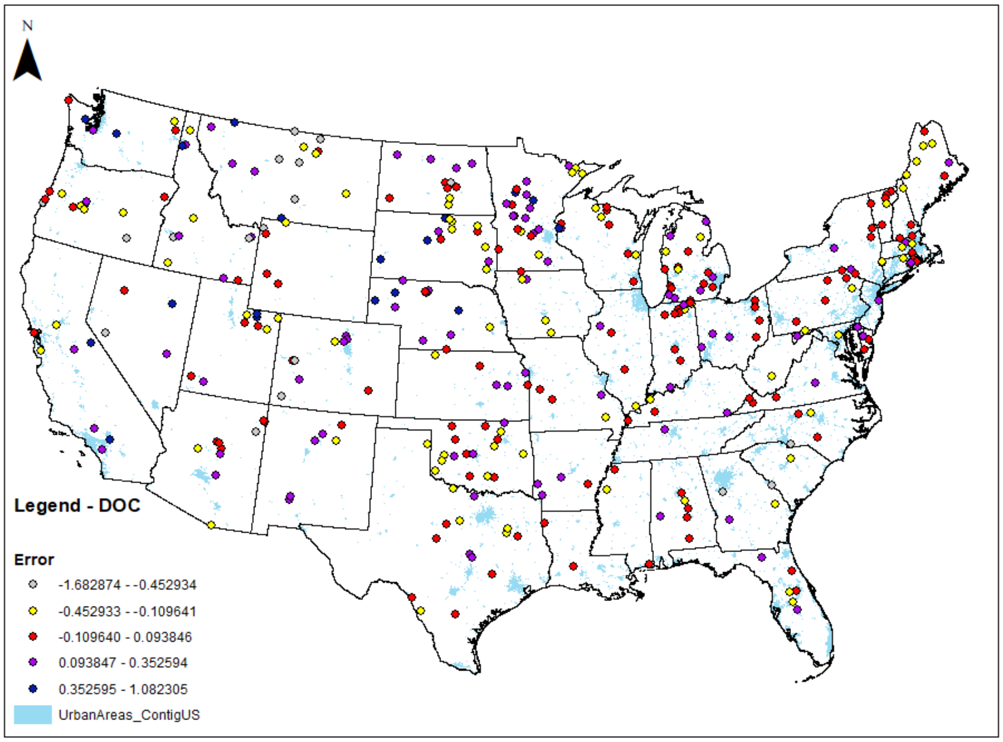

Figure A2. Map of error in DOC_IDW interpolation.

Total Nitrogen (NTL)

To interpolate NTL we included 806 lakes. The power was 1. Neighborhood searching method was standard and included up to 15 neighbors with at least 10 neighbors. Full sector type . No angle with a major and minor semi-axis of 1,295,791.913. A map of error appears random.

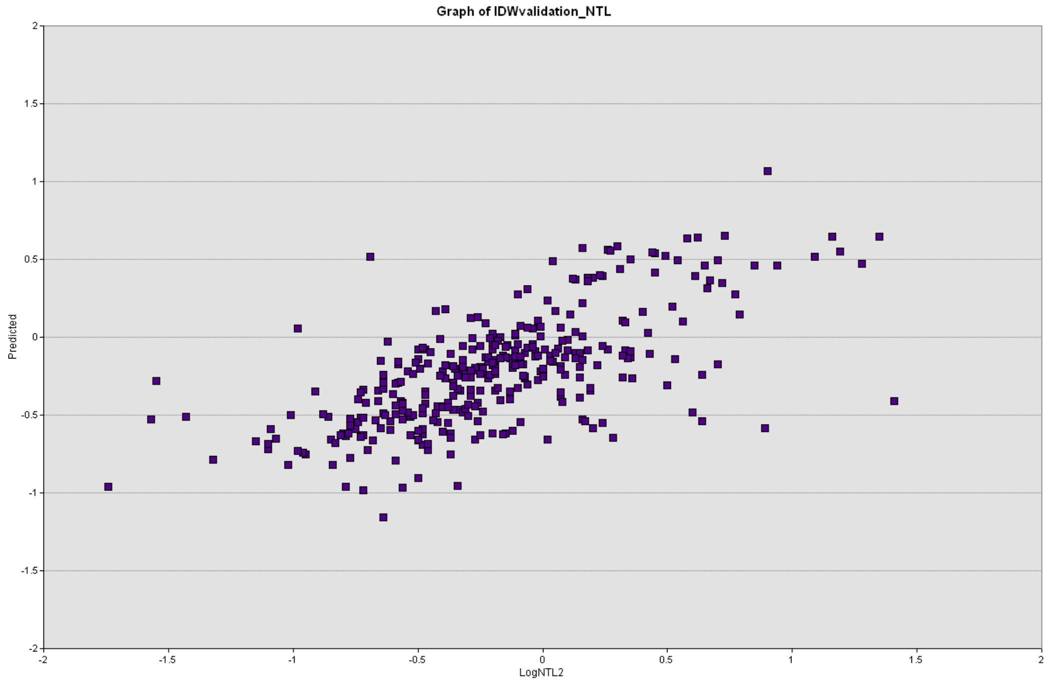

Figure A3. Measured v predicted Log10(NTL) concentrations using the 347 validation lakes.

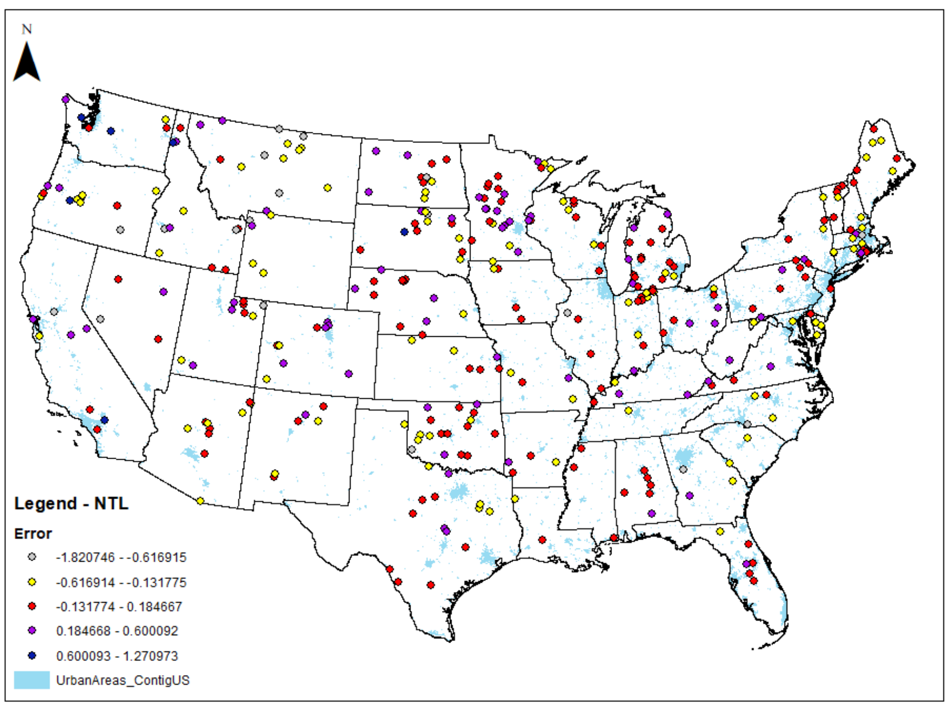

Figure A4. Map of error in NTL_IDW interpolation.

Total Phosphorus (PTL)

To interpolate DOC we included 806 lakes. The power was 1.0620. Neighborhood searching method was standard and included up to 15 neighbors with at least 10 neighbors. Full sector type . No angle with a major and minor semi-axis of 1,295,791.913. A map of error appears random.

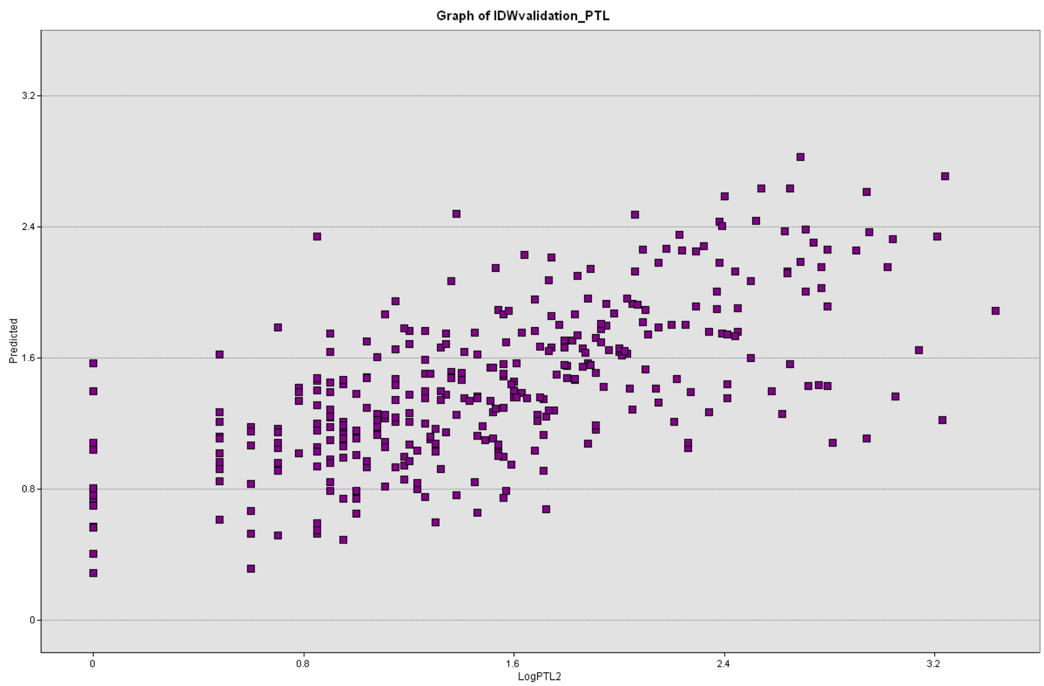

Figure A5. Measured v predicted Log1(PTL) concentrations using the 347 validation lakes.

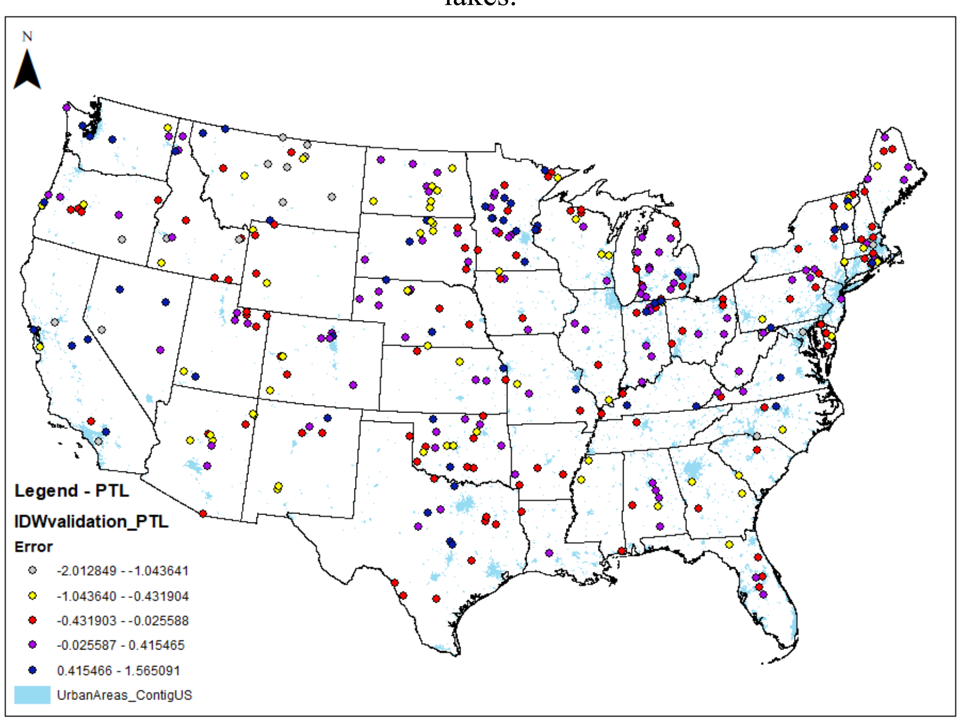

Figure A6. Map of error in PTL_IDW interpolation.

Chlorophyll-a (CHL)

To interpolate DOC we included 806 lakes. The power was 1. Neighborhood searching method was standard and included up to 15 neighbors with at least 10 neighbors. Full sector type . No angle with a major and minor semi-axis of 1,295,791.913. A map of error appears random.

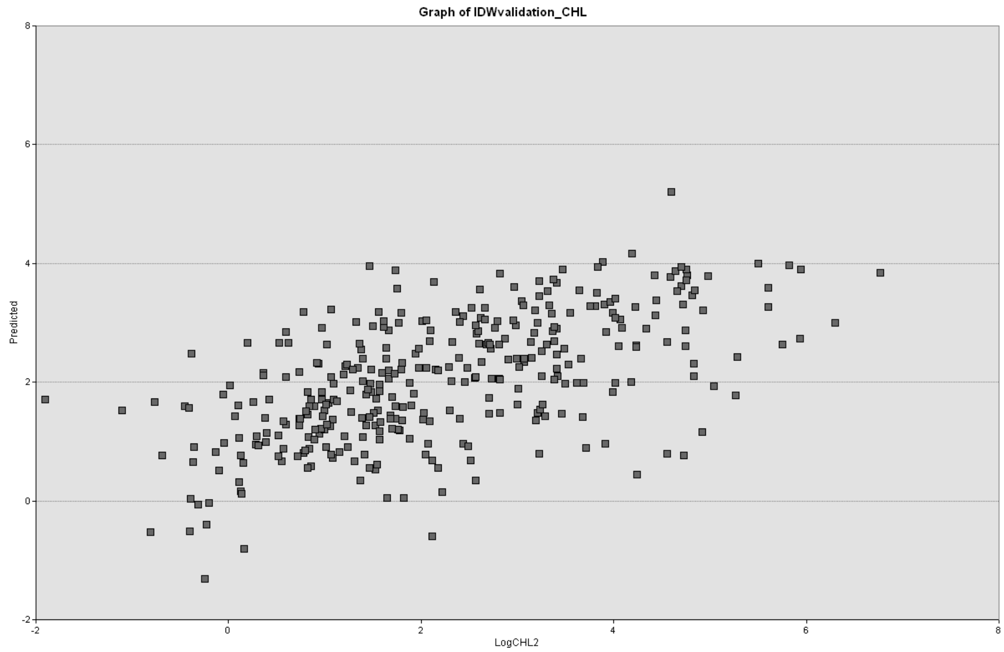

Figure A7. Measured v predicted Log10(CHL) concentrations using the 347 validation lakes.

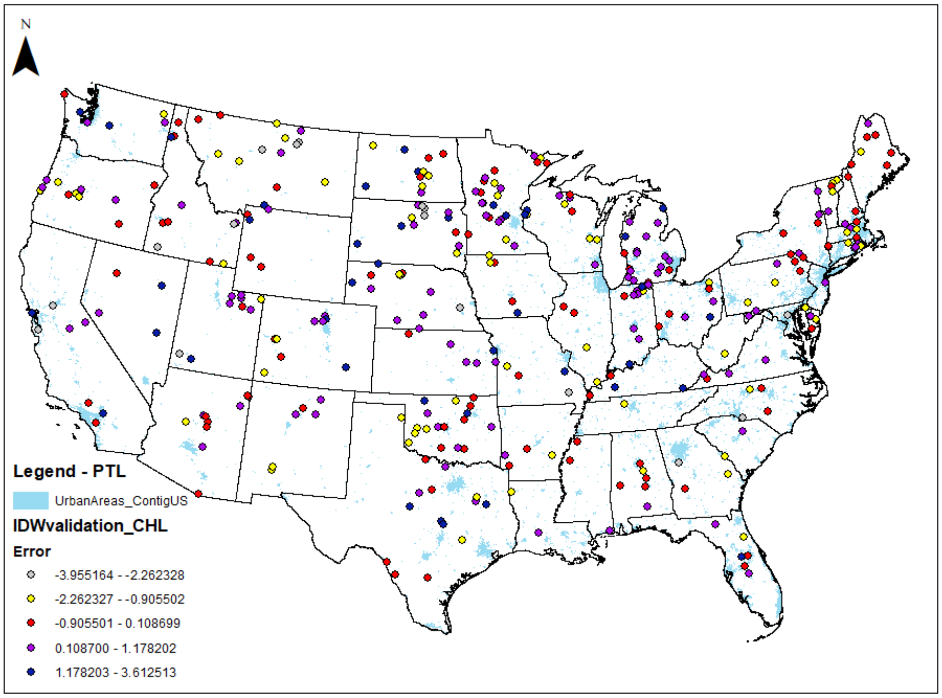

Figure A8. Map of error in CHL_IDW interpolation.

A.2.3 Exploratory Ordinary Least Squares

Because none of the OLS models passed the spatial autocorrelation test, then these results were used to guide the variable inclusion for the GWR. Since our models have spatial autocorrelation, the OLS models are not properly parameterized. Maps for the residuals have some regional clustering, indicating GWR might be appropriate.

DOC

None of the models passed because of spatial autocorrelation ($p < 0.10$), no adjusted r2 were > 0.5, and the JB p-values were < 0.10. Latitude, wetland, forest, and volume are all strong predictors because they are always significant (100%) and always positive (latitude and wetland) or negative (forest and volume). Lake area is another significant variable (95% of the time) but is sometimes negative (55%) and sometimes positive (46%). Ag land is a strong variable (72% significant) and always positive.Watershed area is somewhat strong (71% significant) and almost always negative (61%). The area ratio is not very strong (significant 30% of the time) but almost always positive (60%).

Table A2. Summary of variable significance for exploratory OLS for DOC. Percent significance refers to how frequently the variable is statistically significant and the percent negative and percent positive explains the frequency that the variable has a negative or positive spatial autocorrelation.

Summary of Variable Significance

Variable	% Significant	% Negative	% Positive
LAT_DD	100.00	0.00	100.00
PCTWETLAND	100.00	0.00	100.00
PCTFOREST	100.00	100.00	0.00
LOGVOLUME_KM3	100.00	100.00	0.00
LOGLKA_KM2	95.18	55.42	44.58
PCTAGRIC	72.34	0.00	100.00
LOGWSA_KM2	71.08	61.45	38.55
LOGARATIO	30.12	39.76	60.24

Some variables have collinearity - lake area with volume (VIF=10.96), watershed area with area ratio and volume (VIF = 17.59), area ratio with watershed area and volume (VIF = 8.96), and volume with lake area, watershed area, and area ratio (VIF = 8.74). Latitude, wetland, forest, agriculture land have no collinearity.

The Jarque-Bera p-values indicate the residuals are not normally distributed ($p < 0.001$). In evaluating the residual spatial autocorrelation, the low p-values ($p < 0.001$) indicate spatial autocorrelation exists.

NTL

None of the models passed because of spatial autocorrelation ($p < 0.10$) indicating that we are missing important explanatory variables. Land cover (forest and agriculture) are strong predictor variables; both are always significant - negatively and positively, respectively. Lake volume is another strong predictor because it is always negatively significant. Lake area is always significant, but it varies (55% of the time it is negative and 45% of the time it is positive). Wetlands are significant 78% of the time and almost always positive. The lake:watershed area ratio and watershed area are often significant (64% and 61% of the time). Area ratio is mostly negative (84%) and watershed area is mostly positive (60%). Latitude explains little.

Table A3. Summary of variable significance for exploratory OLS for NTL. Percent significance refers to how frequently the variable is statistically significant and the percent negative and percent positive explains the frequency that the variable has a negative or positive spatial autocorrelation.

Summary of Variable Significance

Variable	% Significant	% Negative	% Positive
PCTFOREST	100.00	100.00	0.00
PCTAGRIC	100.00	0.00	100.00
LOGLKA_KM2	100.00	55.42	44.58
LOGVOLUME_KM3	100.00	100.00	0.00
PCTWETLAND	77.66	5.32	94.68
LOGARATIO	63.86	84.34	15.66
LOGWSA_KM2	61.45	39.76	60.24
LAT_DD	27.66	8.51	91.49

Some variables are strongly correlated with other explanatory variables. Lake area and lake volume (VIF = 10.96). Watershed area is strongly correlated with area ratio and lake volume (VIF = 17.59). Area ratio is strongly correlated with watershed area and lake volume (VIF = 8.96). Volume is strongly correlated with lake area, area ratio, watershed area (VIF = 8.74). Latitude, and land use have no multicollinearity.

The Jarque-Bera p values indicate the residuals are normally distributed (p = 0.439; not great but still high and not significant). In evaluating the residual spatial autocorrelation you also want a high p-value.

PTL

None of the models passed because of spatial autocorrelation (p < 0.10) indicating that we are missing important explanatory variables. Land cover, area ratio, and volume are strong predictor variables because they are always significant (100% of the time) and often consistently negative (forest - 100%, volume - 100%, and Aratio - 87%) or positive (agriculture - 100%). Lake area is mixed because it's often significant (83% of the time) but sometimes positive and sometimes negative. Watershed area is significant 78% of the time and usually positive (81% of the time). Latitude is somewhat important (52% significant) and almost always negative (86%). Wetland is not very important.

Table A4. Summary of variable significance for exploratory OLS for PTL. Percent significance refers to how frequently the variable is statistically significant and the percent negative and percent positive explains the frequency that the variable has a negative or positive spatial autocorrelation.

Summary of Variable Significance

Variable	% Significant	% Negative	% Positive
PCTFOREST	100.00	100.00	0.00
PCTAGRIC	100.00	0.00	100.00
LOGARATIO	100.00	86.75	13.25
LOGVOLUME_KM3	100.00	100.00	0.00
LOGLKA_KM2	83.13	55.42	44.58
LOGWSA_KM2	78.31	18.07	81.93
LAT_DD	52.13	86.17	13.83
PCTWETLAND	20.21	54.26	45.74

Some variables are strongly correlated with other explanatory variables. Lake area and lake volume (VIF = 10.96). Watershed area is strongly correlated with area ratio and lake volume (VIF = 17.59). Area ratio is strongly correlated with watershed area and lake volume (VIF = 8.96). Volume is strongly correlated with lake area, area ratio, watershed area (VIF = 8.74). Latitude, and land use have no multicollinearity. *Note - this is exactly the same as total nitrogen.

The Jarque-Bera p values indicate the residuals are normally distributed (p = 0.956). The low p-value for spatial autocorrelation (p < 0.001) indicates the residuals are spatially correlated.

CHL

None of the models passed because of spatial autocorrelation ($p < 0.10$) indicating that we are missing important explanatory variables. Only 1 model passed based on the minimum r2. Latitude, forest, agriculture, and volume are strong predictor variables because they are always negative (latitude, fores, volume) or positive (agriculture). Wetlands are also a strong predictor because it is significant 80% of the time and almost always significant (94%). Lake area is somewhat useful (significant 57% of the time) but sometimes positive and sometimes negative. Watershed area and area ratio are somewhat good variables (~39% significant) and mostly negative (area ratio - 83% of the time) or positive (watershed area - 70% of the time).

Table A5. Summary of variable significance for exploratory OLS for CHL. Percent significance refers to how frequently the variable is statistically significant and the percent negative and percent positive explains the frequency that the variable has a negative or positive spatial autocorrelation.

Summary of Variable Significance

Variable	% Significant	% Negative	% Positive
LAT_DD	100.00	100.00	0.00
PCTFOREST	100.00	100.00	0.00
PCTAGRIC	100.00	0.00	100.00
LOGVOLUME_KM3	100.00	100.00	0.00
PCTWETLAND	79.79	6.38	93.62
LOGLKA_KM2	56.63	53.01	46.99
LOGWSA_KM2	39.76	30.12	69.88
LOGARATIO	37.35	83.13	16.87

Some variables are strongly correlated with other explanatory variables. Lake area and lake volume (VIF = 10.96). Watershed area is strongly correlated with area ratio and lake volume (VIF = 17.59). Area ratio is strongly correlated with watershed area and lake volume (VIF = 8.96). Volume is strongly correlated with lake area, area ratio, watershed area (VIF = 8.74). Latitude, and land use have no multicollinearity. *Note - this is exactly the same as NTL and PTL.

The Jarque-Bera p values indicate the residuals are normally distributed ($p > 0.90$). The low p-value for spatial autocorrelation ($p < 0.001$) indicates the residuals are spatially correlated.

A.2.4 Geographic Weighted Regression

Explanatory variables were included in GWR based on their performance in the OLS. For each WQ variable, lake and watershed characteristics were used to predict DOC, NTL, PTL, CHL at unmonitored lakes. Included variables were significant 100% of the time.

Table A5. Below are the variables included in the GWR models. For all variables, they are significant in all exploratory OLS models. The directionality of the relationship (+ or

-) is the same 100% of the time except when denoted by a *. For PTL, the lake-watershed area ratio is always significant and is negatively significant 87% of the time.

	DOC	NTL	PTL	CHL
GWR2A	Latitude + Lake volume + Pct wetland -	Lake volume - Pct agriculture +	Lake volume - Lk:Ws area ratio* - Pct agriculture +	Latitude - Lake volume - Pct agriculture +
GWR2F	Latitude + Lake volume + Pct forest -	Lake volume - Pct forest -	Lake volume - Lk:Ws area ratio Pct forest -	Latitude - Lake volume - Pct forest -

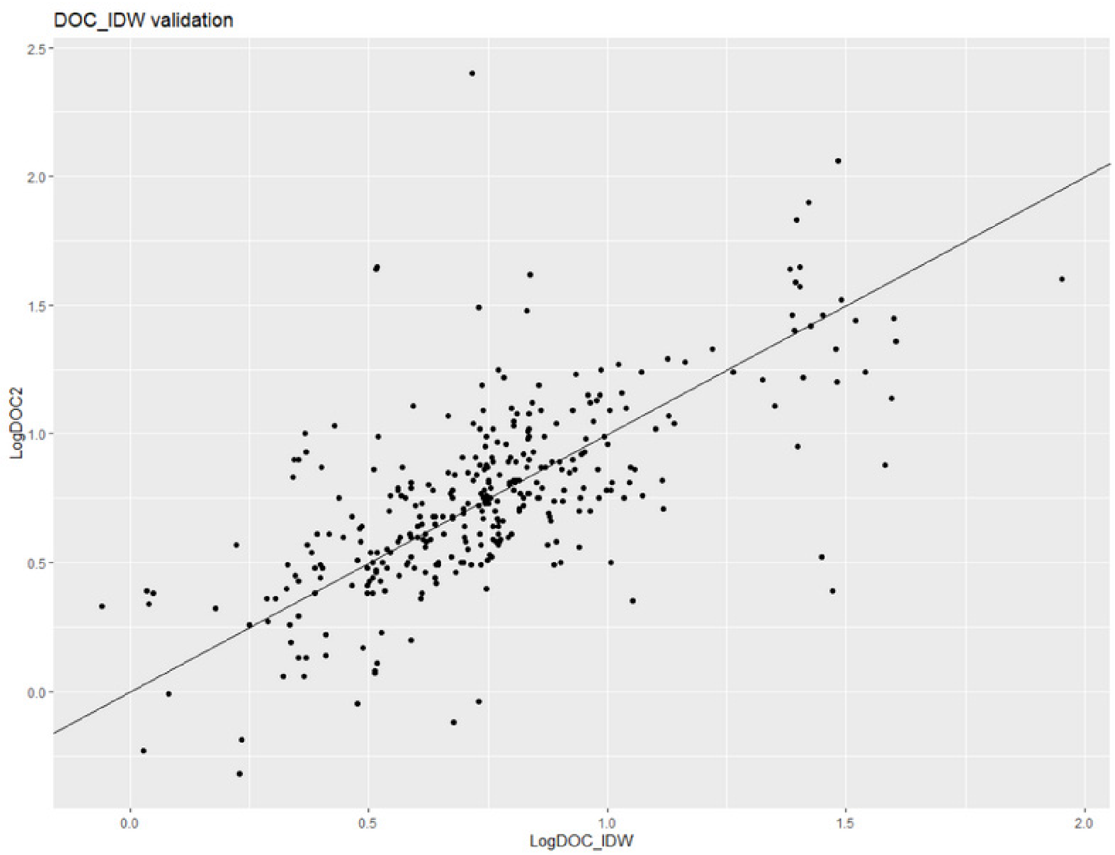

Figure A9. Measured versus inverse distance weighting modeled DOC concentrations in NLA lakes. The line represents a 1:1 slope.

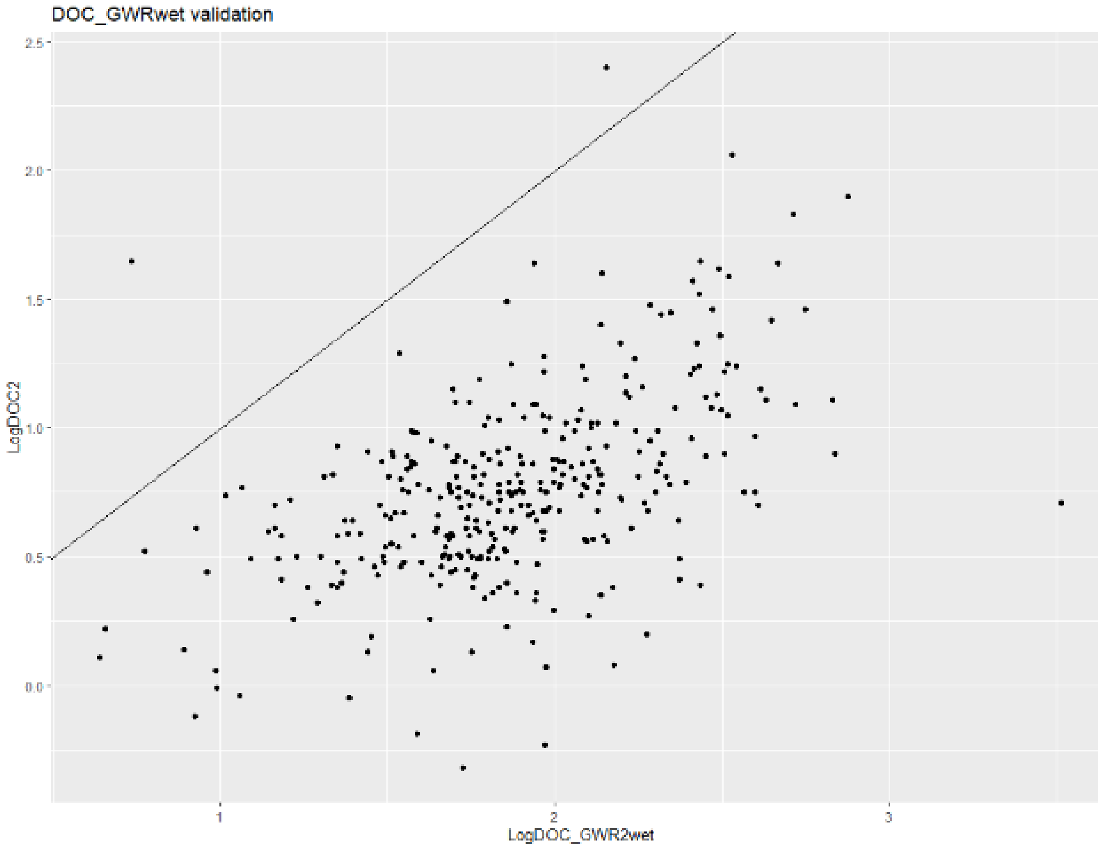

Figure A10. Measured versus geographically weighted regression modeled DOC concentrations in NLA lakes. This modeled used agriculture land use. The line represents a 1:1 slope.

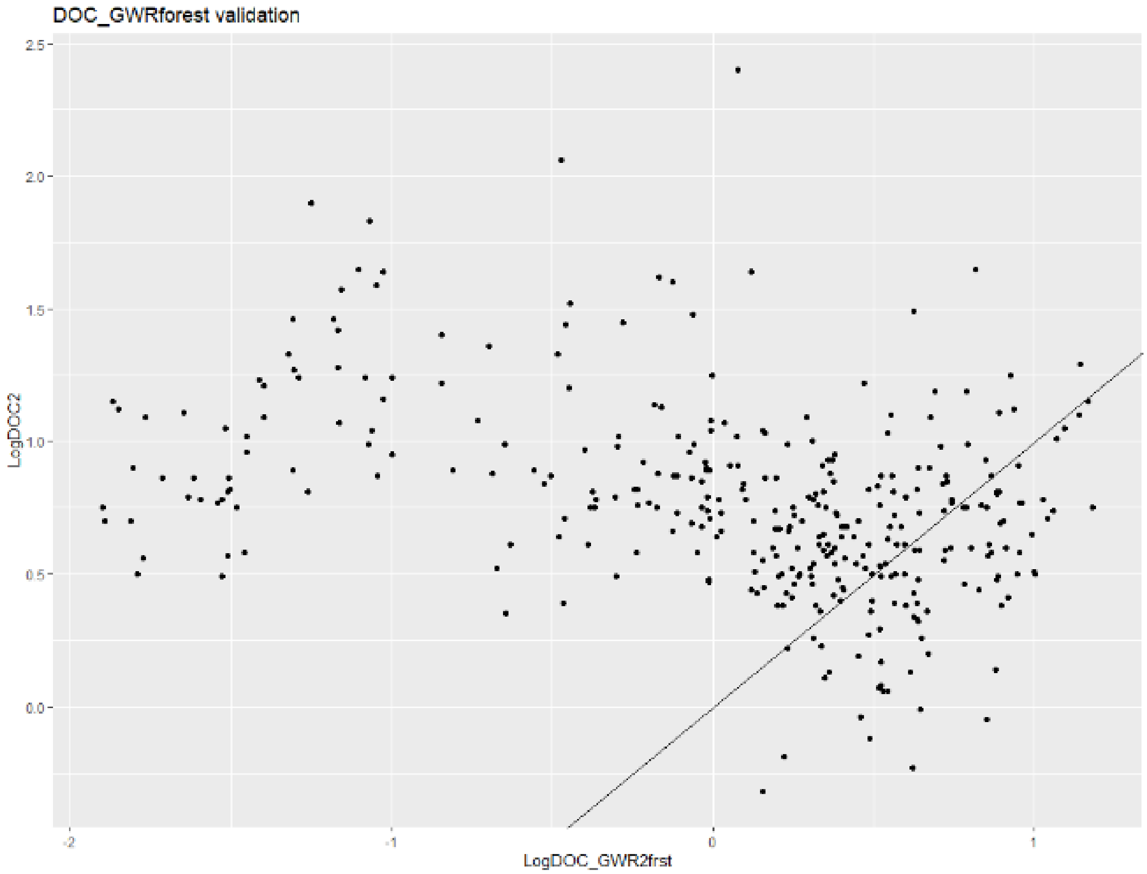

Figure A11. Measured versus geographically weighted regression modeled DOC concentrations in NLA lakes. This modeled used forested land use. The line represents a 1:1 slope.

Table A6. Regression statistics comparing three different water quality modeling techniques – inverse distance weighting, geographic weighted regression with agriculture land use, and geographic weighted regression with forested land use.

		IDW	GWR_2A	GWR_2F
DOC	Slope	0.56	0.58	-0.76
	Intercept	0.33	1.4	0.63
	r2	0.45	0.26	0.13
NTL	Slope	0.49	1.3	1.3
	Intercept	-0.12	-0.22	-0.23
	r2	0.44	0.55	0.52
PTL	Slope	0.44	1.1	1.1
	Intercept	0.76	1.76	1.72
	r2	0.42	0.48	0.5
CHL	Slope	0.40	0.28	0.28
	Intercept	1.2	1.6	1.6
	r2	0.35	0.30	0.24

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