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Mercury Sulfur Initiative: Suggested Program of Research for the Upper Great Lakes States

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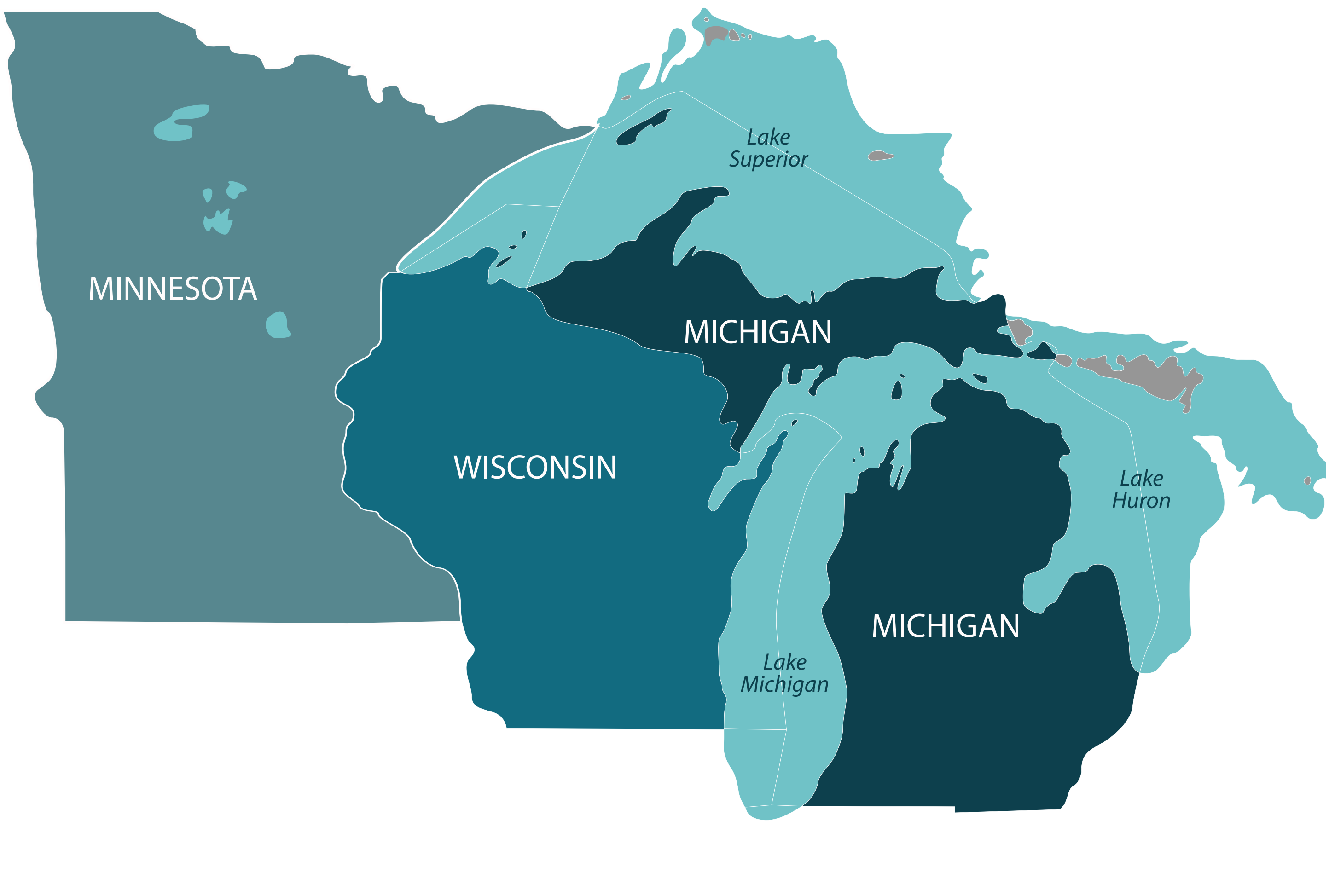

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Abstract

Mercury and sulfur in Upper Great Lakes watersheds of Minnesota, Wisconsin, and Michigan are persistent pollutants that threaten ecosystem services and human health. In 2021, the U.S. Congress directed the U.S. Department of Agriculture, Forest Service's Northern Research Station to study mercury and sulfur pollution in the Great Lakes region over a 5-year period. Specifically, the Northern Research Station was directed to generate science to inform policy regarding mercury and sulfur pollution and to implement demonstration sites for proposed remediation technologies. This report describes the existing scientific resources and cross-disciplinary capabilities that the Forest Service and collaborators contribute to the effort, summarizes existing scientific knowledge, identifies knowledge gaps, and outlines the next steps needed to fill these gaps and develop potential mitigation measures for mercury and sulfur pollution. It details the potential for such pollution to affect valuable resources, including Manoomin (wild rice) and freshwater fish, two ecosystem services that have considerable ecological and socio-economic value and special significance to Indigenous Peoples of the Upper Great Lakes region. How the dynamics of mercury and sulfur may be affected by a changing climate and existing treatments for mercury and sulfur pollution are also reviewed. The report concludes with a series of recommendations for research investments that can provide critical information to land managers and policymakers.

KEYWORDS: mercury, sulfur, freshwater fish, Manoomin, wild rice, climate change, phytotechnology

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Mercury sulfur initiative: suggested program of research for the Upper Great Lakes States

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EXECUTIVE SUMMARY

The Great Lakes region, a landscape dominated by five iconic lakes that border eight states and one Canadian province, is a mosaic of wilderness, wildlife, mining, industry, transportation, and agriculture. In recent years, concentrations of mercury and sulfur, two elements that continue to be persistent pollutants in Upper Great Lakes watersheds of Minnesota, Wisconsin, and Michigan, threaten important ecosystem services such as the production of Manoomin (wild rice; *Zizania palustris*) and fish for consumption. Burning coal, forest fires, taconite processing, and other industries (e.g., cement production, gold mining) contribute mercury and sulfur into the atmosphere. Once in the atmosphere, mercury and sulfur travel both long and short distances before being deposited on land and waters. Mining, manufacturing, utilities, and municipal wastewater production have led to increased levels of sulfur and mercury in lakes and streams. Increased concentrations of sulfide (a sulfur species) in Manoomin waters inhibit Manoomin growth and reproduction. In addition, sulfate (another sulfur species) stimulates the activity of microbes that methylate mercury, thereby producing methylmercury, a neurotoxin that bioaccumulates in fish and poses health risks for humans and the rest of the food chain. The wetlands of the Upper Great Lakes landscape create ideal conditions for the conversion of mercury to methylmercury.

In 2021, the U.S. Congress directed the U.S. Department of Agriculture, Forest Service's Northern Research Station (NRS) to study mercury and sulfur pollution in the Great Lakes region over a 5-year period. Specifically, the Northern Research Station has been directed to generate science and implement demonstration sites for proposed remediation technologies to inform policy regarding mercury and sulfur pollution. The Congressional Directive specifies that the work includes the development of an approach to prevent sulfur species from being transported to areas of mercury accumulation, where it can interact with mercury and form methylmercury.

The objective of this report is to summarize existing scientific knowledge, identify knowledge gaps, and outline next steps needed to fill these gaps to address risks posed by increased mercury and sulfur pollution and investigate potential mitigation measures. It describes the existing scientific resources that the Forest Service and collaborators contribute to the effort and provides a background on mercury and sulfur pollution. Additionally, this report details the potential for such pollution to affect valuable resources, including Manoomin, which is important to Indigenous Peoples of the Upper Great Lakes region, and freshwater fish, both of which have ecological and socio-economic importance in the region. How the dynamics of mercury and sulfur pollution may change under conditions brought about by climate change are discussed, and existing treatments of mercury and sulfur pollution and relevant research are described. The report concludes with the following recommendations for research investments that can provide critical information to land managers and policymakers:

- Understand how climate change and mining activities can influence future mercury storage and transport, sulfur-mercury transformations, and bioaccumulation of mercury in terrestrial and aquatic food chains of the Upper Great Lakes region.
- Build robust models to forecast mercury and sulfur impacts under alternative climate, land use, and policy scenarios and management strategies.

- Conduct laboratory and field experiments to directly test the effects of mercury, sulfur, and climate change on iconic and economically valuable fish species and include the results in model parameterization.
- Develop and improve sulfate treatment and phytoremediation methods to reduce sulfate in industrial discharge and polluted watersheds.
- Create an integrated ecological-economic model and evaluate sulfate abatement strategies that minimize costs to polluters while meeting water quality standards for Manoomin.
- Use data on recreational visits as a case study to estimate the demand for trips to the Boundary Waters Canoe Area Wilderness and welfare loss associated with sulfate and mercury pollution.
- Engage with Tribes to understand the impacts of mercury and sulfur on Manoomin and fish, gain an understanding of what their research needs might be, and incorporate those needs into future research.
- Establish demonstration sites to test the effectiveness of sulfur remediation strategies on surface water before it reaches areas where mercury has accumulated.

CONTENTS

BACKGROUND

Mercury and sulfur pollution have pervasive and widespread effects on soil and water quality in the United States. In 2021, the U.S. Congress directed the U.S. Department of Agriculture (USDA), Forest Service, Northern Research Station (NRS) to conduct research, implement demonstration sites, and generate science to inform policy development regarding mercury and sulfur pollution. One important objective is to prevent sulfur species from being transported to areas of mercury accumulation because the interaction of mercury and sulfate can lead to the formation of the neurotoxin methylmercury. The effects of the relationship between sulfur and mercury in the environment are particularly relevant in the Upper Great Lakes region, where the landscape of abundant wetlands is efficient at converting mercury to methylmercury. In addition to concerns regarding sulfur's interaction with mercury, excess sulfide resulting from mining and other industries is entering wetlands and threatening the sustainability of Manoomin (wild rice) populations. We anticipate that a warming climate will exacerbate this problem because enhanced rates of decomposition in wetlands are tied to increased concentrations of mercury in surface waters. As part of this effort, there is a need to synthesize knowledge concerning: (1) mercury and sulfur cycling, dynamics, and transport; (2) current policy regarding mercury and sulfur levels in the environment; (3) the effects of mercury and sulfur pollution on ecosystems, local communities, and society at large, all in the context of climate change; (4) potential management approaches and treatment options to remove sulfur from the environment, and finally; (5) future research needs.

INTRODUCTION

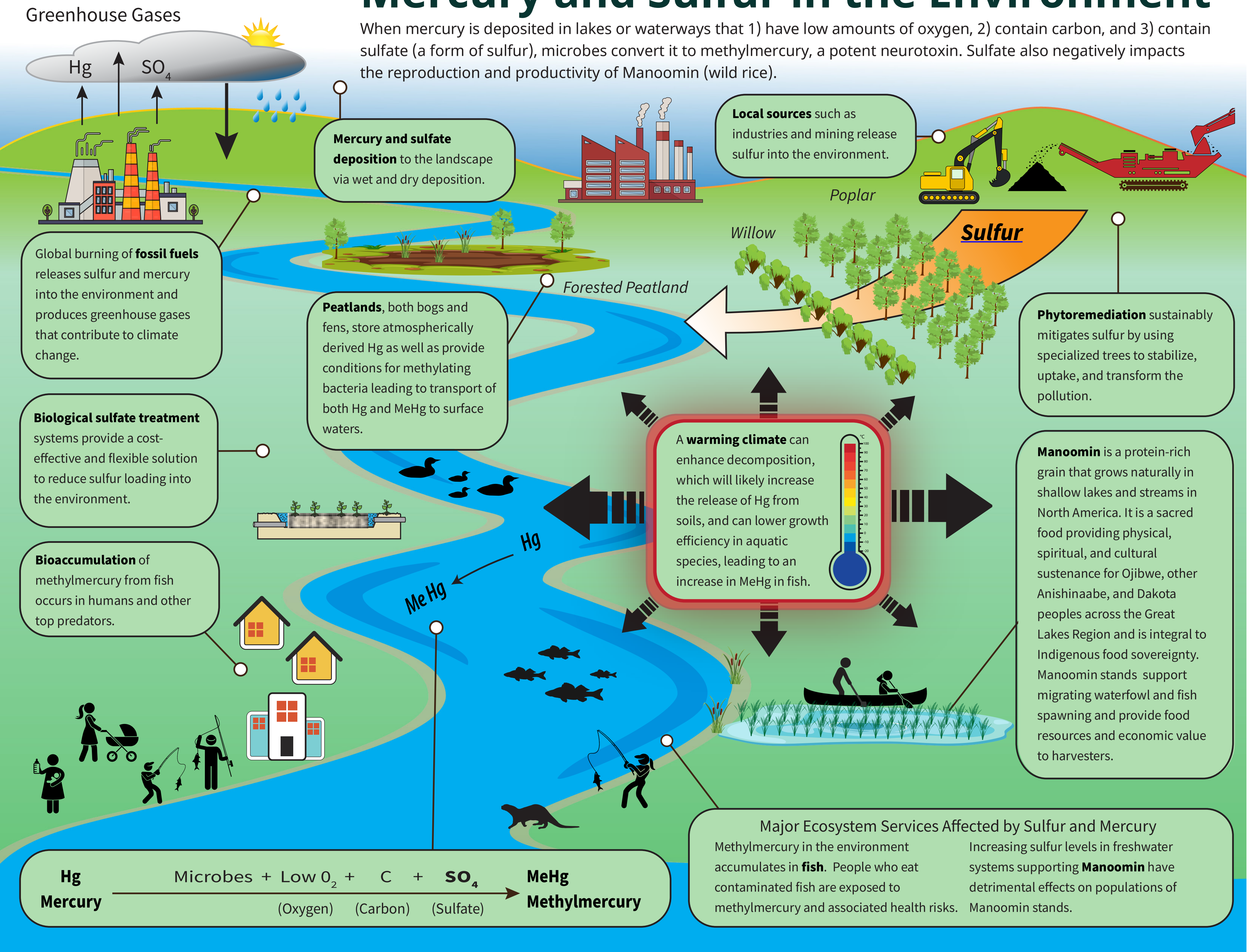
Mercury and sulfur from human industrial activities around the world can negatively influence ecosystem services, making this a global, national, and local environmental concern. Mercury cycles through the environment in several chemical forms. An important step is the conversion of mercury into highly toxic methylmercury through a process called methylation. In this form, methylmercury can enter the food chain where it can bioaccumulate to dangerous levels in top predators including fish, wildlife, and humans (Myrbo et al. 2017). Sulfur can also be converted to various forms important in the environment. Sulfate facilitates the production of methylmercury, and new research (see below) indicates that organic sulfur species might also act in this role. Sulfide in lake sediment is the main culprit in decreasing the production of Manoomin (see page 4).

Sulfate is important because it stimulates the activity of the dominant microbes that methylate mercury (Blythe 2020, Fort et al. 2017). The Upper Great Lakes region contains abundant conifer forests that scavenge the atmosphere of mercury and wetlands that are very efficient at converting mercury into its methylated form, causing concerns about elevated methylmercury levels in the environment (Witt et al. 2009b). Furthermore, industrial activities in the region (e.g., mining, paper production, other manufacturing industries, utilities, and municipal production of wastewater) have led to increased levels of sulfur in lakes and streams (Berndt et al. 2016b, Chapra and Dolan 2012), including seven wild rice lakes in northern Minnesota (US EPA, n.d.b).

Today, mercury and sulfur on the landscape pose ecological and human health risks that may be exacerbated under a changing climate. The following sections detail the scope of the mercury and sulfur problems and how these challenges are relevant for the Upper Great Lakes region, beginning with the major sources of mercury and sulfur in the environment. We use northern Minnesota as our model ecosystem. With its comparable landscapes, similar mercury issues, and importance

Mercury and Sulfur in the Environment

When mercury is deposited in lakes or waterways that 1) have low amounts of oxygen, 2) contain carbon, and 3) contain sulfate (a form of sulfur), microbes convert it to methylmercury, a potent neurotoxin. Sulfate also negatively impacts the reproduction and productivity of Manoomin (wild rice).



of Manoomin, findings from Minnesota can generally be applied across the Upper Great Lakes region. We then discuss the effects of mercury and sulfur on two important ecosystem services, Manoomin production and fish that are safe for consumption. This is followed by outlining the current regulatory framework for both pollutants as it relates to these ecosystem services. Impacts of a changing climate on mercury, sulfur, and their interactions are then addressed, along with management implications. Finally, we recommend research on sustainable treatment options for addressing mercury and sulfur pollution. An approach to addressing key information gaps and developing mitigations strategies is proposed.

MERCURY AND SULFUR: SOURCES AND RECEIVING WATER BODIES

Sources

More than 75 percent of the mercury that enters the environment is a result of human caused emissions into the air from local or global sources. Volcanism and geothermal events are the main natural sources of mercury, and account for less than 25 percent of primary emissions (Driscoll et al. 2013). Activities such as burning coal and processing taconite release mercury into the air. In Minnesota, the amount of mercury released into the environment from the taconite industry ranks second only behind power production (Berndt 2003). Forest fires are another large source of mercury to the atmosphere, with mercury in particulates being deposited downwind from the fire (Witt et al. 2009a). Atmospheric mercury may travel long distances before it falls to the earth with precipitation or dust. Eventually this mercury can end up in lakes, rivers, and oceans. When mercury is deposited in lakes or waterways in which sulfate (a form of sulfur) is present, microbes can convert it to methylmercury, a potent neurotoxin (Jeremiason et al. 2006) that affects fish and birds, animals, and humans who consume the fish. Methylmercury bioaccumulates in aquatic food chains, making it a critical issue in places with an abundance of wetlands, lakes, and rivers.

Sulfur and its various forms including sulfate and sulfide also enter the atmosphere from emissions (e.g., burning high-sulfur coal) and leave the atmosphere in the form of acid rain. While atmospheric deposition tends to be the main source of sulfur in watersheds of the Upper Great Lakes, local point sources exacerbate concentration in water bodies. Similar to other Upper Great Lakes watersheds, sulfur is locally discharged into Minnesota waters from municipal wastewater treatment plants, as well as industrial sources such as taconite mines, glass manufacturers, paper mills, and gravel pits. In Minnesota soils, the pore water concentration of sulfate can be as high as 3350 milligrams per liter, several magnitudes higher than the 10 milligrams per liter limit related to Manoomin production (Berndt et al. 2016a). Both sulfate and sulfide are known to have negative effects on Manoomin, an important cultural resource and commodity in the Great Lakes region (LaFond-Hudson et al. 2020, Myrbo et al. 2017, Pastor et al. 2017). The Minnesota Pollution Control Agency (MPCA) estimates that 19 to 27 percent of waterbodies containing Manoomin are downstream from a permitted discharger of sulfate, and nearly 130 dischargers are located 25 miles or less upstream of a Manoomin water (Governor's Task Force on Wild Rice 2019). The amount of sulfate released ranges widely among dischargers, but new monitoring requirements will help to determine which dischargers need to lower their point-source sulfate concentrations via source reduction or installation of treatment technologies (Baubitz et al. 2021).

Receiving Water Bodies

Rivers, lakes, and wetlands receive sulfur and mercury from the atmosphere in the form of both wet and dry deposition and from industrial activities that produce sulfate-containing runoff. Preventing sulfur from entering natural systems is a challenge for mining companies, electric utilities, and other industries. Research is needed to understand how to "clean" mine and industrial runoff before it enters the environment as well as how to remove sulfur species that already exist in the environment. There are about 13.4 million acres of peatlands in the Upper Great Lakes region of the United States, with Minnesota leading the way with over 6 million acres of peatlands, second in acreage only to Alaska (Kolka et al. 2016). Wetlands, specifically high-carbon northern peatlands, tend to be sinks of inorganic mercury, and they have been shown to be sources of methylmercury to downstream surface waters (Grigal 2003). The methylation of mercury occurs most readily in environments with low levels of oxygen, such as peatlands or wetlands, and is mediated by microorganisms such as sulfate-reducing bacteria. This conversion process is not entirely understood and may be affected by additional factors including water temperature, water level, dissolved organic carbon concentration, iron concentration, microbial community composition, and algae abundance (Paranjape and Hall 2017; Xu et al. 2019b). Peatlands have high water tables (leading to low oxygen in the soils), large pools of carbon, and adequate sulfate concentrations from decades of atmospheric deposition. As a result, waters that drain peatlands, which are typically headwaters systems, have relatively high methylmercury concentrations that bioaccumulate in downstream food chains.

Forest Inventory and Analysis (FIA) Program: Understanding Mercury and Sulfur Distributions Across the Landscape

The Northern Research Station's Forest Inventory and Analysis (FIA) program has traditionally provided national leadership on a soil sampling campaign conducted as part of FIA's strategic inventory (Amacher and Perry 2010, O'Neill et al. 2005). Regional and national scale analyses utilizing the cutting-edge datasets generated by the inventory allow for the identification of mercury and sulfur hotspots in soils within the Upper Great Lakes region (Perry et al. 2009).

FIA's inventory of forests and soils is intended to support the identification and exploration of regional patterns of soil attributes and their linkages to forest characteristics (Berryman et al. 2020, Horn et al. 2018, Jo et al. 2019, Page-Dumroese et al. 2021). Recent work has highlighted the opportunity to inform strategic estimates of forest soil properties (Domke et al. 2016, 2017). Of particular interest in this context is the description of mercury and sulfur patterns across the landscape and linkages to forest attributes (Perry et al. 2009).

ECOSYSTEM SERVICES AFFECTED BY MERCURY AND SULFATE

Minnesota as a Model for the Upper Great Lakes Region

Glacial processes across the Upper Great Lakes formed similar landscapes (e.g., soils, amounts of wetlands, and forest cover) across the region, making the landscape of Minnesota comparable to that of northern Wisconsin and Michigan. All three states have issues related to mercury deposition and accumulation in the aquatic food chain, fish consumption advisories, and subsequent impacts on recreation. In addition, Manoomin is an important food crop that affects the livelihood of Indigenous Peoples in the region. As a result, the findings and information gathered by this effort are generally applicable across the Upper Great Lakes States.

Ecosystem services, the end products of an ecosystem that are directly used or consumed by people (Boyd and Banzhaf 2007), contribute to the production of benefits and may be affected by environmental policy or management. We use a template for identifying benefits, beneficiaries, and ecosystem services associated with Minnesota waters (Table 1). The template comes from a previous review of the practice of assessing and valuing ecosystem services (Binder et al. 2017). Two important ecosystem services are the production of Manoomin and abundant fish with low mercury concentrations. Beneficiaries of these ecosystem services include Tribal communities and others who tend, harvest, and consume Manoomin; recreational anglers; fisheries (including Tribal fisheries); and fish-eating wildlife that depend on abundant and low mercury concentrations in fish. Sulfur and mercury can negatively affect these ecosystem services. For example, high sulfate concentrations in combination with other factors negatively affect the occurrence of Manoomin (Myrbo et al. 2017). Further, mercury contamination of fish poses health risks for people and wildlife and may limit the use and availability of otherwise healthy, local sources of sustainable food. In Minnesota, the MPCA is responsible for enforcing a limit on sulfate concentration in waters that support Manoomin, and emissions of mercury from coal-power facilities are controlled by the Federal Government through the 1990 Clean Air Act Amendment. Details on the effects of mercury and sulfur pollution on these two ecosystem services, as well as the current regulatory framework for managing these pollutants in Minnesota, are described in this report.

Table 1.—Major ecosystem services affected by mercury and sulfur pollution in Upper Great Lakes region

Land use	Ecosystem service	Benefit	Beneficiary
Shallow and slow-moving water for Manoomin stands	Manoomin harvested	Growing and consuming Manoomin; Fish and wildlife habitat; Shoreline protection	Tribal communities; recreational harvesters; Manoomin consumers
Lakes and rivers used for fishing	Fish with low levels of mercury	Fish as a resource; Biodiversity	Recreational, commercial, and Tribal fisheries and the economies they support; Iconic wildlife species (e.g., loons, otters)

Manoomin and Sulfate

Cultural and Ecological Significance of Manoomin

In their paper on collaborative tribal-university partnerships on Manoomin, Matson et al. (2021) provide a short summary of Manoomin and the institutional and intellectual context for its study. Manoomin means "the good fruit" or "spirit delicacy" in Ojibwe and "wild rice" in English. This protein-rich grain grows naturally in shallow lakes and streams in North America. As a sacred food, Manoomin provides not only physical but also spiritual and cultural sustenance for Ojibwe, other Anishinaabe, and Dakota peoples across the Upper Great Lakes region and is integral to Indigenous food sovereignty. Seasonal cycles of Manoomin are important to Anishinaabe lifeways, but these lifeways are threatened because self-sustaining Manoomin stands are declining due to multiple environmental stressors, including sulfate contamination in supporting waters.

In addition to its cultural significance, Manoomin has ecological functions. It is native to North America, especially the Upper Great Lakes region, including Minnesota, Wisconsin, and Michigan and extending into parts of Canada (Waheed 2021). Manoomin stands support migrating waterfowl and fish spawning and provide food resources and economic value to harvesters. As the beloved state grain in Minnesota, Manoomin is central to debates around environmental policy and management.

Minnesota is home to 11 sovereign Tribal Nations, and much of the Manoomin in Minnesota exists on Tribal reservations and treaty ceded territories (Minnesota Tribal Wild Rice Task Force 2018). State and federal projects and programs have broad impacts on Tribal communities and businesses, and state and federal agencies engage with Tribes both in a formal consultation process and in less formal contexts.

How Sulfate Affects Manoomin and Communities

Sulfate in water is converted to sulfide by microbes in the stream or lake sediment in which Manoomin is rooted. Most researchers agree that as sulfide levels in the sediment increase, the abundance of Manoomin decreases (LaFond-Hudson et al. 2020, Myrbo et al. 2017, Pastor et al. 2017, Pollman et al. 2017). The rate and geochemical conditions at which sulfate is converted to sulfide and how Manoomin is affected is an active area of scientific discussion. The United States Environmental Protection Agency (EPA) recently advocated for Minnesota Manoomin waters with elevated sulfate concentrations be designated as impaired (US EPA, n.d.b).

Discussions about sulfate mitigation and the protection of Manoomin raise real fears about the future of Upper Great Lakes communities and environments, especially for Tribes, state-licensed Manoomin harvesters, industry groups, and clean water advocates. Such fears can lead to passionate narratives and a breakdown of understanding and trust. A greater shared understanding of the concerns of Tribes, Manoomin resource users, and other impacted communities is needed to build bridges across political and cultural boundaries.

Fishing and Mercury

Fishing is one of the most popular recreational activities in the Upper Great Lakes region. Between 2000 and 2016, over one million Minnesota residents per year purchased annual fishing licenses (Burkett and Winkler 2018). Given the number of people purchasing fishing licenses, it is not surprising that recreational fishing is a multi-billion-dollar industry. In 2018, the American Sportfishing Association estimated that 1.8 million anglers spent $2.7 billion while fishing in Minnesota, which supported over 28,000 jobs. People enjoy great benefits from fishing, as evidenced by their willingness to pay for their experiences (e.g., Bennear et al. 2005), which has a tremendous effect on the recreation economy of the Upper Great Lakes region.

One of the primary ways people are exposed to mercury pollution is by eating contaminated fish. Methylmercury, the neurotoxic form of mercury, accumulates in algae and invertebrates that are eaten by smaller fish, which are then eaten by larger fish. Fish at the top of the aquatic food chain, such as walleye, can have methylmercury concentrations as high as a million times that of the surrounding water (Morel et al. 1998). Experiments and field studies can be used to understand how factors such as temperature, water chemistry, food sources, and competition influence bioaccumulation in fish (Ward et al. 2010). Results from laboratory studies and clinical human impact studies (Minnesota Department of Health 2011) help inform legislation designed to set emissions and fish concentrations at safe levels. Minnesota's target level for mercury in fish is 0.2 milligrams per kilogram (parts per million, ppm), based on the Environmental Protection Agency's development of a methylmercury criterion for fish tissue to protect human health.

In addition to regulating mercury emissions, the Minnesota Department of Health issues fish consumption guidance statewide and for specific lakes. For example, the 0.2 ppm methylmercury criterion for perch corresponds to a statewide fish consumption advisory threshold for one meal per week for women who are pregnant or intending to become pregnant and children under 15 years of age. Waterbody specific safe-eating guidelines provide advice for lakes and rivers where fish have been tested and contaminant levels are elevated: https://www.health.state.mn.us/communities/environment/fish/docs/eating/specpoplakes.pdf .

CURRENT REGULATORY FRAMEWORK IN MINNESOTA

Sulfate Regulatory Framework

While Michigan and Wisconsin have extensive Manoomin stands, Minnesota's lakes and rivers support the largest remaining naturally-occurring Manoomin beds in the United States. To protect Manoomin, in 1973 Minnesota adopted a Manoomin sulfate standard of 10 milligrams per liter in surface waters, which limited sulfate discharges to 10 milligrams per liter in "water used for the production of wild rice." Shortly thereafter, the EPA approved the standard, and it is still in effect today. The standard, however, does not explicitly define which lakes, rivers, and streams it applies to, and as such, state agencies and Tribal governments have created various lists of Minnesota waters that contain Manoomin. For example, the Minnesota Department of Natural Resources prepared a list of 1,286 lakes and rivers/stream segments with stands of natural wild rice (Hansen 2008, Minnesota DNR 2008). In 2017, the MPCA proposed a list of about 1,300 bodies of water, gleaned from a variety of sources, where the proposed standard would apply (MPCA Wild Rice Waters 2016). The list has been updated on an ongoing basis since then (Governor's Task Force on Wild Rice 2019).

Facilities that discharge sulfate into waterbodies need discharge permits under the National Pollutant Discharge Elimination System. Although Minnesota has issued discharge permits, the numeric sulfate standards have not been enforced by the MPCA because the Minnesota Legislature passed a law in 2015 saying it could not enforce the stands until rule making under the Federal Clean Water Act was complete (Bjorhus 2021) . To meet the State's sulfate standard of 10 milligrams per liter in surface waters, facilities that have permits to discharge treated water to lakes and rivers in Minnesota could be required to reduce sulfate in their discharges if they are upstream from Manoomin waters. Policy supporting reduction of sulfate discharge and sulfate mitigation plans for facilities that contribute substantial amounts of sulfate or sulfide to Manoomin beds could help meet the State's sulfate standard.

Mercury Regulatory Framework

Atmospheric deposition of mercury on lands and waters is a worldwide problem, with much of the emissions coming from coal-fired power plants (Giang and Selin 2016). In response, the EPA has adopted national standards for controlling mercury emissions. The United States is also a cosigner of the United Nations Minamata Convention on Mercury that is a binding global treaty to reduce mercury pollution worldwide.

Although about 90 percent of mercury deposited in Minnesota comes from outside the State, Minnesota's approach to regulating mercury within its boundaries is based on setting a total maximum daily load (TMDL) goal, which was approved by the EPA in 2007 (MPCA 2007). The annual atmospheric deposition goal is 4.4 grams per square kilometer per year for northeastern Minnesota. The plan for achieving this deposition goal includes requiring owners of ferrous mining and processing facilities to reduce emissions by 75 percent from 2008/2010 levels by January 2025 (Maccabee 2010, MPCA 2009). These standards do not take into consideration climate change and the potential effects that increased temperatures and major storm events can have on the amount of mercury and sulfur entering into surface water systems.

CLIMATE CHANGE: EFFECTS ON MERCURY AND SULFATE

Understanding and mitigating the risks of mercury and sulfur pollution to human and ecosystem health must consider the implications of a changing climate (Obrist et al. 2018). In the northeastern and upper midwestern United States, water and soil temperatures are increasing and are projected to rise for the foreseeable future. In addition, changing patterns and amounts of precipitation (rain and snow) have altered the timing, magnitude, and variability of stream and river flows and the drying and wetting of floodplains and wetlands. In Northern Minnesota, annual temperatures have already increased between 2–3 ^{o}C in the past 60 years, with an additional 3–4 ^{o}C projected by 2100 (Liess et al. 2021 Handler et al. 2014).

Climate Change and Bioaccumulation

Climate change influences bioaccumulation in aquatic ecosystems through multiple and complex pathways at multiple scales (Ward et al. 2010). At the base of the food web, warmer temperatures increase the activity of microbes that transform inorganic (and relatively non-toxic) mercury into toxic methylmercury. Increased microbial activity would, therefore, likely increase methylmercury concentrations in soil, sediment, and water. Further, higher water temperatures will increase the metabolic rates of fish and other aquatic species at the top of the food chain. These increased metabolic costs will require individuals to consume more prey (and more toxic methylmercury within those prey), potentially leading to greater bioaccumulation and higher mercury concentrations in the fish, and fish-eating wildlife and people.

Bioaccumulation and Concurrent Sources of Environmental Change

In addition to a changing climate, the Upper Great Lakes region has experienced and will continue to experience other major trajectories of environmental change. Perhaps most directly relevant to toxic metal accumulation, and in response to emissions reductions associated with the Clean Air Act (1977 and 1990 Amendments), the atmospheric deposition of mercury has generally decreased (Brigham et al. 2021), although not at all Mercury Deposition Network sites in northern Minnesota. In theory, reduced atmospheric deposition of mercury should reduce the amount of methylmercury entering food chains, but in practice, mercury toxicity and transport are much more strongly associated with food web transfer. Sulfate deposition has also greatly decreased over the past 40 years (Greaver et al. 2012), and this reduction in sulfate deposition has had complex effects on mercury pollution. Lower sulfur deposition, by contrast, directly reduces the activity of the bacteria that methylate mercury (Drevnick et al. 2007), thereby reducing the amount of toxic mercury entering the food chain. However, these reductions may be counteracted by other effects of climate change that fuel the activity of methylating microbes and increase toxic methylmercury in aquatic ecosystems.

Climate-change impacts on precipitation type, amount, and intensity will alter hydrological cycles and will also influence mercury cycling. Changes in the duration and frequency of wetting and drying cycles, which are likely to be strongly affected by expected changes in precipitation, influence methylation rates in wetlands and floodplains (hotspots for toxic mercury) (Coleman Wasik et al. 2015, Mitchell et al. 2008). Compared to the influence of increasing temperature, however, the consequences of changes in hydrologic regimes on toxic mercury are more complex and less well-studied, representing a major information gap and science need.

Management Actions Related to Climate Change

A range of practices have been employed to restore aquatic and wetland ecosystems. These strategies are being increasingly evaluated in the context of climate change and climate resilience. However, we have little understanding of how these practices, on their own and interactively with climate change, will influence current and future sulfur and mercury cycling and what the consequent effects will be on ecosystem and human health.

Many, if not all, management approaches that influence mercury dynamics are likely to have complex, and in some cases, non-intuitive effects that may be time- and scale-dependent. One key to understanding these impacts is via the influence of a management action on ecosystem productivity. For example, maintaining and restoring riparian canopy along rivers is a well-established strategy, but one which may, as an unintended consequence, increase the concentration of mercury in river food webs via reductions in prey biomass and growth efficiency of consumers (invertebrates and fish) (Ward et al. 2010). Other planned management activities (e.g., forest harvesting, prescribed fire) or unplanned major disturbances (e.g., wildfire) also tend to release mercury in the environment (Bishop et al. 2009, Kolka et al. 2017, Witt et al. 2009a). Some management strategies may affect mercury via multiple pathways. Floodplains and wetlands are hotspots both for biological production (lowering toxic mercury) and for methylation (increasing toxic mercury) (Rypel et al. 2008). The ultimate outcomes therefore, are dependent on the relative strength of these two processes (production and methylation) in the context of a changing climate.

The complex nature of these interactions underscores the importance of targeted research and application. Empirical surveys and experimental manipulations, in combination with integrative models, can improve the understanding and forecasting of current and future sulfur and mercury pollution risks and inform better response strategies.

Forest Service Research on Climate Change, Mercury, and Sulfur

Research conducted by the Northern Research Station is currently addressing fundamental questions related to climate change and its effects on mercury and sulfur. Initial studies on watershed mercury cycling and interactions with sulfate began in the 1990s at the USDA Forest Service Marcell Experimental Forest (MEF) in northern Minnesota. Since 2012, the NRS has been assessing this interaction within the framework of the Department of Energy (DOE) sponsored SPRUCE (Spruce and Peatland Responses Under Changing Environments) project, the most extensive climate change experiment in the world. https://www.nrs.fs.fed.us/disturbance/climate_change/spruce/.

Preliminary results from the SPRUCE project indicate increases in methylmercury peatland porewaters and runoff with warming. If warming increases methylmercury concentrations in fish, there could be potential implications for human health, outdoor engagement, and the fishing economy. Recent research involves developing techniques with collaborators at the DOE's Oak Ridge National Laboratory to assess the genetics of microbes associated with mercury methylation. The SPRUCE project is a large asset and will allow NRS researchers to further investigate the impact of climate change on mercury and sulfur.

Other important climate-related research occurs at PEATCOSM, NRS's mesocosm facility in Houghton, MI, including the effect of water table levels and vegetation on mercury cycling and methylmercury production. https://www.nrs.fs.fed.us/clean_air_water/local-resources/downloads/peatcosm_information.pdf

TREATMENT OPTIONS

Mitigating the impacts of mercury on the environment requires efforts on multiple fronts. The most obvious is to prevent mercury from entering the environment via the direct release of mercury in wastewater and air. In some cases, established management approaches can mitigate mercury and sulfur pollution and associated impacts (see previous section). A second strategy is to reduce the methylation of previously deposited mercury by minimizing its transport to potential methylation hotspots (e.g., wetlands, other environments with low levels of oxygen).

Similarly, limiting sulfate concentrations and its transport to methylation hotspots would reduce mercury methylation. Sulfur levels can be reduced either by decreasing its direct discharge from industrial sources (Bowell 2004, Johnson and Hallberg 2005) or by implementing techniques to remove sulfur already in the environment (Miao et al. 2012). Northern Research Station and its partners at the Natural Resources Research Institute will assess approaches to both remove sulfur from contaminated surface water and watershed soils. Examples of specific types of sulfur mitigation work are described below.

Sulfate Treatment

Sulfate treatment is possible with a variety of technologies, but cost-effective options are limited (Table 2). Existing treatment options can reduce sulfate concentrations to below the EPA drinking water standard of 250 milligrams of sulfate per liter (US EPA, n.d.a). However, sulfate reduction below the Manoomin standard (10 milligrams of sulfate per liter) relies primarily on membrane-based technologies, which generate large volumes of liquid and solid waste (e.g., brine and sludges) that are challenging and costly to manage.

Table 2.—Summary of costs associated with sulfate treatments (Bolton and Menk, Inc. and Barr Engineering Company 2018)

Cost category	Bioreactor	Reverse osmosis	Electrodialysis	Membrane filtration
Capital Cost[a]	\$0.32 M	\$0.82 M	\$0.69 M	\$0.50 M
Operating Costs	\$0.30 /m^3	\$0.87 /m^3	\$0.48 /m^3	\$0.27 /m^3
Sludge production	Low-Moderate	Low	Low	Moderate-high
Maintenance	Moderate	High	High	High

[a]Per 1000 m^3/d; Abbreviations: m^3 = cubic meters; d = day; M = millions

Sulfate concentrations in waste streams are categorized into three regimes based on the sulfate source: 50–250 ppm from municipal water treatment facilities; 250–2000 ppm from agriculture, industry, and mining pit lakes; and >2000 ppm from power utilities and the metallurgy mining industry (Bavin and Berndt 2008; Bolton & Menk, Inc. and Barr Engineering Company 2018; Johnson et al. 2019). To achieve Minnesota's stringent Manoomin sulfate standard, each regime may require two or more remediation technologies to be applied in tandem. Further, cost estimates of both water treatment for sulfate and the reduction of sulfate generated at the source are needed.

In comparison to material- and energy-intensive membrane technologies, biological sulfate reduction is a promising and economical alternative for sulfate removal. Most biological treatment methods use naturally occurring microbes to convert sulfate in wastewater to sulfide, which can be sequestered to form an unreactive and manageable solid phase. This process could be applied in both active (e.g., bioreactors) and passive (e.g., constructed wetlands) treatment systems (Bowell 2004, Johnson and Hallberg 2005, Miao et al. 2012, Muyzer and Stams 2008). However, past studies on biological sulfate treatment have focused solely on the fundamental scientific mechanisms rather than engineering applications. At present, much of the research on biological sulfate treatment has been conducted via small-scale laboratory studies (Deen 2021, Takaki 2018). Based on promising laboratory results, it is now important to demonstrate the feasibility, reliability, and affordability of biological sulfate treatment systems for treating wastewater at an intermediate (pilot) scale. Such efforts will increase confidence in the efficacy of biological sulfate treatment by establishing a scientific and engineering foundation for the technology.

Levels of certainty and understanding will increase with scaled-up sulfate treatment research. Scaled-up research requires partnerships that provide access to high-sulfate wastewater for a meaningful demonstration of the technology and an understanding of site-specific sulfate issues. Cost-effective treatment options will benefit both the industries that need to meet water quality standards and the public who relies on clean water and healthy environments for recreation and sustenance. This could lead to lower infrastructure maintenance costs, reduced wastewater volumes, and improved water quality. Our research will be a tool to accelerate implementation of emerging technologies associated not only with Minnesota water quality efforts, but also for other communities within the Upper Great Lakes region, as well as across the United States.

Targeted Sulfur Treatment Option: Phytoremediation

Phytoremediation is a proven approach that uses natural physiological processes of plants and their associated soil microorganisms to clean contaminants from polluted soils, sediment, and waters (McCutcheon and Schnoor 2003, Rai 2008). Phytotechnologies have been successfully implemented to remediate a broad range of organic and inorganic contaminants, including explosives, petroleum hydrocarbons, landfill leachate, and dredging sludge (McCutcheon and Schnoor 2003, Pilipović et al. 2019, Zalesny et al. 2007). It is this versatility that makes phytoremediation a possible candidate to address sulfur problems in the Upper Great Lakes region, particularly when part of a landscape level co-strategy to reduce sulfur in impacted watersheds.

All plants require sulfur for physiological processes related to growth and development (Gallardo et al. 2003, Marschner and Marschner 2012). However, plants differ in their uptake and metabolism of sulfur, and thus there is broad variability in the potential for remediation of sulfur (Ernst 1998, Marschner and Marschner 2012, Rennenberg et al 1990). The performance of systems designed for phytoremediation of sulfur are also largely influenced by site conditions such as climatic and soil variables. Therefore, selecting the proper plants that are matched to the contaminants and specific site conditions is of utmost importance for reclamation (Mosseler and Major 2017;,Pilipović et al. 2021, Xu et al. 2019a). Use of plant materials following maturity will likely be in wood products or converted to biochar for longer-term storage.

To address the need for such selection, NRS researchers and partners have developed, tested, and refined phyto-recurrent selection. This green tool utilizes multiple testing cycles in the greenhouse and field to choose superior-performing tree varieties based on growth, physiological, and biomass parameters (Zalesny and Bauer 2007, Zalesny et al. 2007). Short rotation woody crops (SRWC) such as hybrids of poplars (*Populus*) and willows (*Salix*) are the most effective trees used worldwide for phytoremediation. These species exhibit key traits that make them preferable candidates for remediation activities, including extensive root systems and rapid growth (Isebrands et al. 2014, Rockwood et al. 2004, Zalesny et al. 2016). Previous studies have revealed promising results regarding poplar and willow for phytoremediation of mine wastes (Brunner et al. 2009, Guittonny-Larchevêque and Lortie 2017, Mosseler and Major 2017, Suo et al. 2021), though results specifically related to remediation of sulfur species at the field scale are limited and not well-tested for effectiveness.

Building upon this research, phyto-recurrent selection can be utilized in the context of sulfur mitigation to select and outplant SRWCs on contaminate sites. Specifically, this effort will (1) select tree varieties to maximize sulfur remediation; (2) establish demonstration plantings and conduct monitoring to evaluate the efficacy of SRWC-based phytoremediation of sulfur; (3) develop specialized laboratory methods to quantify sulfur species throughout the system; and (4) conduct education, outreach, and technology transfer activities that involve engaging directly with community members, Tribal citizens, and other stakeholders.

Bioeconomic Analysis

To meet Minnesota's sulfate standard of 10 milligrams per liter in surface waters for the production of Manoomin, facilities such as municipal wastewater treatment plants that have permits to discharge treated water to lakes and rivers in Minnesota could be required to reduce sulfate in their discharges if they are upstream from Manoomin waters. Strategies for reducing sulfate discharge and sulfate mitigation plans are needed for those facilities to meet the State's sulfate standard while minimizing costs to the polluters. Further, mechanisms for financing the costs of pollution control are needed to prevent undue burden on rural communities and industries. We envision building a bioeconomic optimization model for the development of cost-effective sulfate abatement strategies. The decision maker is the state regulator who weighs different strategies for reducing sulfate discharge and cleanup. The decisions include the selection of facilities for required discharge reduction and the location and type of sulfate mitigation measures. The economic parameters include the costs of these treatment activities, and the benefits include the avoided costs of sulfate pollution, especially the avoided reduction in Manoomin production. Estimating these benefits will require modeling the hydrology of watersheds that support Manoomin in northern Minnesota as well as information on the location of Manoomin waters and upstream sulfate discharge sites. The expected outcomes of this policy analysis include the following:

- Cost-effective deployment strategies for sulfate abatement technology to meet water quality standards for Manoomin surface waters.
- Tradeoffs between sulfate abatement costs and upper bounds on sulfate loads.
- Prioritization of sites for sulfate abatement.
- Mechanisms for financing pollution control strategies that minimize economic impacts on rural communities.

SUMMARY OF CURRENT AND POTENTIAL FOREST SERVICE RESEARCH

Addressing Mercury and Sulfur in the Upper Great Lakes Region

The Forest Service is focusing on four subject areas to address mercury and sulfur interactions inthe Upper Great Lakes region. The Northern Research Station has developed research goals and identified specific questions for each of these areas:

- Ecosystem Services
- Climate Change
- Sulfate Engineering Solutions
- Phytoremediation

Ecosystem Services

Ecosystem services, such as the production of Manoomin and abundant fish with low mercury concentrations, contribute to the wellbeing of important communities who benefit from pristine northern waters, including Tribal communities and recreational anglers. The production of these ecosystem services and their benefits to communities may be affected by environmental policy and management decisions related to pollution control. Understanding the benefits and costs of these decisions is critical for developing sound ecosystem management strategies.

Research Goals

1. Create an integrated ecological-economic model and evaluate sulfate abatement strategies that minimize costs to polluters while meeting water quality standards for Manoomin.
2. Use data on recreational visits to estimate the demand for trips to the Boundary Waters Canoe Area Wilderness and welfare loss associated with sulfur and mercury pollution.

Specific Questions

- How can we protect Manoomin from sulfate pollution while minimizing costs for communities and industries responsible for the pollution?
- What is the potential loss in recreation value of the Boundary Waters Canoe Area Wilderness if mining discharge contaminates surface waters in the Wilderness Area?
- What are the costs to anglers to mitigate mercury levels in fish?

Climate Change

The Forest Service is acutely aware that climate change is driving many ecosystem changes in the environment. Understanding the additive effects of climate change and how systems are reacting to the changes is critical for planning future land management actions and understanding how communities will be further impacted.

Research Goals

1. Harness the unique infrastructure of the SPRUCE project to study the effects of climate change on mercury and sulfur in wetland environments.
2. In partnership with land managers (e.g., National Forest System), use ongoing surveys, databases, experiments, and ecological models to understand complex impacts and make robust forecasts under alternative climate change scenarios.

Specific Questions

- How will warmer, wetter, and more extreme conditions impact mercury and sulfur cycles, ecosystems, fish concentrations, and habitats, and the valuable services they provide?
- How do we assess the combined effect of management actions and climate change to inform future mitigation adaptation strategies?
- How do we incorporate other sources of environmental change or disturbance in these models and forecasts (e.g., land use change, forest harvesting, fire)?

Engineering Solutions

Northern Research Station scientists have been partnering with academia to investigate the potential for **engineering solutions** to remove excess sulfate from polluted waters. The application of current research methods is ready for testing at demonstration sites. Results will help to determine the upscale value of treatments.

Research Goals

1. Utilize promising laboratory results with fundamental scientific basis to implement sulfur treatment systems at an intermediate scale in the field.
2. Establish a scientific/engineering foundation to increase confidence in the biological sulfur treatment process.

Specific Questions

- What are the performance efficiencies and final products of biological sulfate treatment systems at the demonstration scale?
- Which water chemistry and site-specific characteristics will be important for increasing the reliability and development of large-scale treatment?

Phytoremediation

Phytoremediation technologies are highly desired applications that allow for treatment of pollutants as well as the greening of landscapes. Northern Research Station researchers have current demonstration locations that are actively treating contaminants.

Research Goal

1. Apply information from research and demonstration sites to areas in watersheds where mercury and sulfur have a high potential for interaction and downstream assets are at risk.
2. Apply phytoremediation technologies to utilize decades of NRS experience with short rotation woody crops and phyto-recurrent selection to address knowledge gaps and provide on-the-ground remediation strategies for sulfur.

Specific Questions

- What are the thresholds of tolerance and toxicity for the phytoremediation of sulfur species?
- Which varieties of short rotation woody crops will maximize phytoremediation effectiveness of sulfur species?
- How effective are field-scale, short rotation woody crop-based phytoremediation systems at mitigating sulfur species?

CONCLUSIONS AND RECOMMENDATIONS

The objective of this report is to summarize existing scientific knowledge, identify knowledge gaps, and outline next steps needed to fill these gaps to address risks posed by increased mercury and sulfur pollution and investigate potential mitigation measures. The Forest Service has a long-standing interest and need to understand and develop good insight into mercury and sulfur interactions and where those interactions are occurring on the landscape. Local levels of mercury are influenced by local, national, and global industrial activities as well as landscape influences such as the area of wetlands in watersheds. Great Lakes States have stringent water monitoring programs and fish consumption guidelines to reduce health risks of mercury exposure. However, current policy is still in flux about how sulfur should be regulated and what levels can be achieved to protect resources such as Manoomin. While there is a general understanding that mercury negatively affects human health and safety, the impacts on society of sulfur and its interaction with mercury are less clear. More research is needed to investigate these complicated, intertwined effects, especially given changing climate. Further, treatments for sulfur in waters are not well established, nor economical. This hurdle not only affects the industries that need to treat effluent and waste for sulfur, but also the remediation of contaminated landscapes and watersheds where current sulfur levels are high.

The Forest Service and its partners have multiple resources, including the unique SPRUCE experiment, Forest Inventory and Analysis data sets (see https://www.fia.fs.fed.us/tools-data/index.php), and collaborative skills, to develop solutions that can be utilized as part of the management of mercury and sulfur in the environment and to address the complex interaction between ecological and socioeconomic drivers in the Upper Great Lakes region. We provide research and technology development recommendations (See Box) that can address the knowledge gaps and remediation needs to reduce risks from bioaccumulation of mercury and transport of sulfur species in aquatic systems.

Northern Research Station Recommended Research Investments

- Understand how climate change and mining activities can influence future mercury storage and transport, sulfur-mercury transformations, and bioaccumulation of mercury in terrestrial and aquatic food chains of the Upper Great Lakes region.
- Build robust models to forecast mercury and sulfur impacts under alternative climate, land use, and policy scenarios and management strategies.
- Conduct laboratory and field experiments to directly test the effects of mercury, sulfur, and climate change on iconic and economically valuable fish species and include the results in model parameterization.
- Develop and improve sulfate treatment and phytoremediation methods to reduce sulfate in industrial discharge and polluted watersheds.
- Create an integrated ecological-economic model and evaluate sulfate abatement strategies that minimize costs to polluters while meeting water quality standards for Manoomin.
- Use data on recreational visits as a case study to estimate the demand for trips to the Boundary Waters Canoe Area Wilderness and welfare loss associated with sulfate and mercury pollution.
- Engage with Tribes to understand the impacts of mercury and sulfur on Manoomin and fish, gain an understanding of what their research needs might be, and incorporate those needs into future research.
- Establish demonstration sites to test the effectiveness of sulfur remediation strategies on surface water before it reaches areas where mercury has accumulated.

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