

JGR Biogeosciences



COMMENTARY

10.1029/2022JG007065

Key Points:

- A well-executed study sheds new light on the “wicked” problem of how forest harvest influences the mobility and bioaccumulation of mercury
- The study is particularly valuable because it extends across several environmental system boundaries: atmosphere-forest-peatland-biota
- Crossing downstream boundaries can show how more of the stream network embedded in a landscape reacts to land-use and climate change

Correspondence to:

K. Bishop,
kevin.bishop@slu.se

Citation:

Bishop, K., & Eklöf, K. (2022). Boundary-crossing field research marks the way to evidence-based management of mercury in forest landscapes. *Journal of Geophysical Research: Biogeosciences*, 127, e2022JG007065. <https://doi.org/10.1029/2022JG007065>

Received 27 JUN 2022
Accepted 29 JUL 2022

Author Contributions:

Conceptualization: Kevin Bishop, Karin Eklöf
Supervision: Karin Eklöf
Writing – original draft: Kevin Bishop
Writing – review & editing: Kevin Bishop, Karin Eklöf

Boundary-Crossing Field Research Marks the Way to Evidence-Based Management of Mercury in Forest Landscapes

Kevin Bishop¹  and Karin Eklöf¹

¹Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, Uppsala, Sweden

Abstract The atmospheric deposition of long-range atmospheric mercury pollution presents forest managers with a “wicked” problem—forestry operations run the risk of mobilizing this pollution legacy. Management of that risk would benefit from a process-based understanding of how forest management influences the mercury cycle. This commentary highlights the value for building such an understanding of a comprehensive Before-After-Control-Impact study reported by McCarter et al. (2022), <https://doi.org/10.1029/2022JG006826> on the Marcel Experimental Forest in the north-central continental US. That study looked at how different types of forest harvest influenced the movement of mercury through the landscape. The results of this study place it at the minimal end of the range of impacts on Hg mobilization resulting from forest harvest. What makes this paper, together with the companion papers resulting from this study, particularly valuable for improving the understanding of forestry influences on mercury is the number of system boundaries that the study crossed: between land and atmosphere, from a forested hillslope down into a wetland, and finally up into the biota on that wetland.

Plain Language Summary Forest harvest can mobilize toxic mercury from forest soils and move it into living organisms. This mercury originated in air pollution created far away from the forest, but forest managers still need to deal with the risks of this “pollution legacy” to people, fish and wildlife. A recent study in the north-central US took a detailed look at how two different types of forest harvest mobilized mercury in the soil. This study showed a relatively small impact of the forest harvest on mercury relative to some other studies. Since previous studies have found a wide range of mercury responses to forest harvest, this carefully designed and executed study has value in adding to the evidence base about forest management impacts on mercury in the environment. What is particularly valuable about this study is its comprehensiveness, since it crosses a number of environmental system boundaries: between the forest and the atmosphere, from upslope mineral soils into a downslope peatland, and from the wetland environment into the biota.

1. Introduction

McCarter et al. (2022) have provided a carefully planned and executed study of how forest harvest influences the mobility and bioaccumulation of mercury. This is a valuable addition to the knowledge of how one can adapt forestry to reduce the risks of releasing more mercury (Hg) into the environment. Given how ubiquitous the legacy of Hg pollution is in forest soils, and the extensive disturbance that forest management entails, this study is a valuable step on the long path to learning how to better blunt the threat from past atmospheric pollution when conducting forestry operations.

Unlike many of the environmental toxins currently being focused on in research and the public debate, Hg is not an “emerging threat”. Its devastating toxicity is well documented, and it is all too commonly present in food at levels above what are considered safe for consumption by people or wildlife (UN Environment Program, 2019). The fact that Hg is the single largest cause of waters in the European Union (27 countries with 450 million inhabitants) failing to live up to the goals of the EU Water Framework Directive illustrates just how widespread Hg contamination is (Kristensen et al., 2018). The threats posed by Hg pollution unite 137 countries in the UNEP Minamata Convention to reduce human and wildlife exposure to Hg. But simply reducing Hg emissions will not remove the need to manage the problem of the Hg already spread around the globe. This is especially true for the forest landscape where atmospheric deposition of Hg continues to increase concentrations of Hg in forest soils, even where atmospheric concentrations of Hg pollution have declined from peak levels in the latter half of the twentieth century (Shanley & Bishop, 2012). Since runoff concentrations from forest landscapes are large enough

© 2022. The Authors.

This is an open access article under the terms of the [Creative Commons Attribution License](https://creativecommons.org/licenses/by/4.0/), which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

to start biomagnification to harmful levels in aquatic biota, but too small to drain the forest soil pool, the “legacy” of atmospheric Hg pollution accumulated in the forest soils will persist.

This creates a “wicked problem,” that is one that is resistant to solution due to the combination of complexities in both the science and the societal setting (Lidskog et al., 2018). It is not the forest owners who created the pollution. Nonetheless, harvest and other management activities can increase loadings to aquatic ecosystems. Climate change may exacerbate this problem by either destabilizing some terrestrial Hg pools in ways that increase Hg fluxes from the landscape into surface waters or by changing trophic interactions in ways that can increase the vulnerability of ecosystems to mercury bioaccumulation (Bishop et al., 2020). And while there are recommendations for best management practices in forestry that aim at reducing the risks, the evidence base for these management practices remains weak due in part to the large range in how Hg in runoff responds to forest management (Hsu-Kim et al., 2018).

Ideally there will be validated biogeochemical models that can simulate the effect of different harvest methods on Hg. Catchment scale chemical transport models (CTMs) for Hg exist, for example, INCA-Hg (Futter et al., 2012), RIM-Hg (Eklöf et al., 2015), and those included in the ensemble modeling of Golden et al. (2012). These may someday provide a basis for recommendations about best management practices that are differentiated enough to account for the wide range of soil, climate and vegetation settings where forestry is practiced. But so far applications of catchment CTMs have generally focused on reproducing observed behavior and not been tested as predictors of Hg responses to forest harvest or other perturbations. This is understandable given that the response to forest harvest ranges from manifold increases in fluxes and/or concentrations of different Hg species, to no response (Eklöf et al., 2016). There are hypotheses about why these differences occur after harvest, such as whether reductions in evapotranspiration after harvest lead to inundation of previously well drained soils or simply making wet areas wetter (Kronberg et al., 2016). These hypotheses, however, remain to be tested and quantified in the predictions of CTMs.

2. The Need for Field Studies

Previous reviews of the state of Hg modeling have pointed to the need for a dialog between observation, experimentation and modeling (Sonke et al., 2013; Zhu et al., 2018). However, an increase in modeling studies relative to field studies over recent decades has been noted in the hydrological literature (Blume et al., 2017; Burt & McDonnell, 2015; Kirkby, 2004). Part of this change in publication patterns is driven by the expense and risk of experimental field studies in relation to modeling. This puts a premium on field studies of Hg cycling under different kinds of perturbation where the studies are rigorously executed and designed to look beyond the response at a single point (often a catchment outlet) into what is happening within the soils, and ideally even the biota.

McCarter et al. (2022) is thus a welcome addition to the literature with its thorough analysis of results from an exceptionally comprehensive experiment on responses to forest harvest using before-after-control-impact (BACI) design. Two years of pre-treatment data were collected from three instrumented hillslopes and the wetland which those hillslopes drained into. This was followed by two more years of observations after the forest was harvested on two of the hillslopes. The logging brush was removed on one of these, and left on the other.

The major findings of the paper were that after harvest, total Hg and dissolved organic carbon (DOC) concentrations in water leaving the hillslope decreased, but the total mass of Hg and DOC reaching the wetland increased due to increases in the flux of water moving downslope. In the soils of the down-gradient peatland, the concentrations of methylmercury decreased, while methylation rates and bioaccumulation in invertebrates did not change. That places this site at the minimal impact end of the range of Hg responses to forest harvest reported in the literature reviewed by Eklöf et al. (2016) and commented on earlier.

A major take-home from the field study is the importance of hydrology for harvest-related changes in Hg concentration and fluxes. One needs to consult the previously published paper on the hydrological effects of forest harvest at this site (McCarter et al., 2020) to fully appreciate the hydrological influence on both the harvest treatments and between year variations. The general picture is that increased hillslope water yields after harvest (especially when brush was left on the forest floor) diluted the Hg and DOC, while at the same time increasing downslope export of these dissolved constituents from the hillslopes. The analysis goes on to examine intriguing differences between solutes and treatments, as well as the situation in the receiving peatland. A companion paper

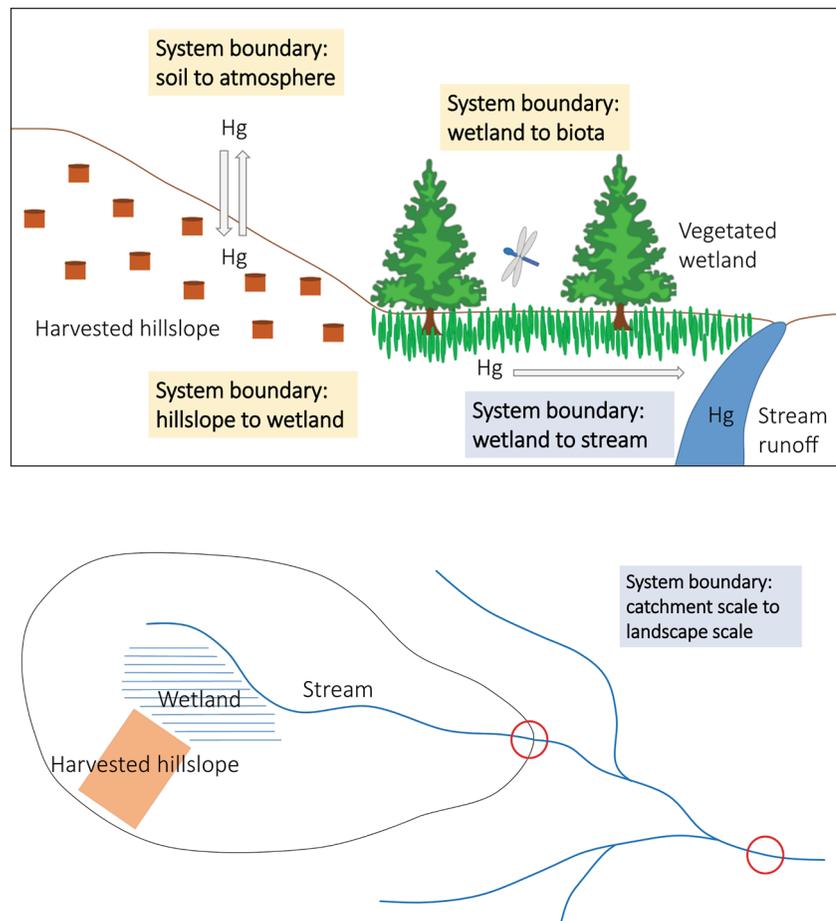


Figure 1. A schematic of environmental system boundaries that are of particular relevance to the fate and transport of mercury pollution deposited from the atmosphere on a forest landscape with both peatlands and a network of watercourses. The high-latitude areas where climate and physiography interact to create such landscapes are characterized by mercury bioaccumulation to levels that are harmful to humans and wildlife. Scientific studies that cross these boundaries improve the understanding of forest harvest effects on the mobilization and subsequent bioaccumulation of mercury in these landscapes. Highlighted in yellow are the environmental system boundaries crossed in a 4 year long forest harvest Before-After-Control-Impact experiment conducted at the Marcell Experimental Forest (North-central USA) that are reported by McCarter et al. (2022) and companion studies. Two more boundaries that warrant consideration are highlighted in blue.

uses Hg isotope tracers to reap further insights from the field experiment (McCarter et al., 2021). These tracers showed the overall dominance of previously accumulated legacy Hg in the Hg moving downslope with the water, but also the larger relative mobility of recently added Hg when comparing the small mass of the added Hg in comparison to the large store of Hg already in the soil.

3. The Value of Crossing Environmental System Boundaries

The comprehensive follow-up of this forest harvest on the Marcell Experimental Forest earns it a prominent place among the studies that comprise the empirical evidence base about forestry's impacts on Hg. But what most distinguishes this study is that it follows the fate of Hg across several system boundaries. Almost all studies that evaluate the effects of land use on Hg have only measured Hg at a single point in the disturbed landscape, in a stream that defines the outlet to a catchment. Few have looked into the landscape, and those that do have tended to stay within the boundaries of a single system: a soil profile, a hillslope, a wetland or an indicator species (Figure 1). This study looks at all of these, linking them together across system boundaries and thereby adding insight into knowledge gaps about forestry effects on Hg.

The first boundary crossed is that between soil and atmosphere, by means of tracer additions that approximated newly added Hg from the atmosphere (McCarter et al., 2021) and flux chamber measurements that looked at Hg returning to the atmosphere from the soil (Mazur et al., 2014, 2015). These identified a loss of Hg from the forest floor after harvest, with Hg mobilized by the harvest moving both down the hillslope as well as up to the atmosphere. Catchment scale studies have generally found undisturbed forest soil to retain over 90% of atmospheric Hg inputs to catchments (Bishop et al., 2020). Crossing this system boundary thereby helps highlight the importance of forest harvest in mobilizing Hg pollution from distant Hg emission sources that is deposited on forests from the atmosphere.

The second system boundary crossed is from the hillslope into a downslope peatland. The instrumented hillslopes are isolated with transverse trenches where the slopes enter into the peatland, and the sectors of the peatland receiving water flow from the upslope harvested areas are sampled separately from peatland sectors below undisturbed hillslopes. It is helpful to know that the rate at which Hg is methylated (which makes Hg more bioavailable) went down in the peatland sectors below the harvested slopes, and that the total amount of Hg in the peatland below the upslope harvest did not measurably change.

By crossing a third system boundary, from the peatland into the biota, an answer was provided to the key question of how bioaccumulation is affected by the forest harvest. This is especially important to look at in the biota itself, since the forest harvest may not only change the amount of MeHg present, but also the food web structure, and thus the vulnerability of that food web to Hg bioaccumulation. McCarter et al. are also to be commended for sampling at the base of the food web, a critical link between water and biota that is often overlooked due to the focus on upper levels of the food web where biomagnification has already raised Hg to levels that are known to be harmful (Wu et al., 2019).

The crossing of a fourth system boundary can even be inferred: from the terrestrial environment to receiving waters. Even though this study itself did not go up to the catchment scale, references to other studies indicate that the mass of Hg moving down the hillslope was several times larger relative to what is found leaving catchments in studies from areas with similar annual water balances. The fact that the hillslope is mobilizing more Hg than is usually seen leaving catchments in runoff suggests that much of the Hg moving down the hillslopes is being sequestered somewhere between the hillslope and the nearest stream. The organic-rich soils of the wetland (or riparian soils in areas without extensive wetlands) are obvious candidates for this sequestering function. The fact that an increase in peatland Hg was not observed in this study does not rule out that possibility for ongoing Hg sequestration in the peatland due to the difficulty of discerning such sequestration given the large size of the Hg store in the peatland relative to the inputs from the hillslope over the course of the study.

To say that studies which cross system boundaries are of special value should not be taken to mean that system boundaries are a hinder. In environmental science, boundaries are of tremendous value since they create a basis for quantification of mass balances (Zhu et al., 2018). The very existence of catchment science, for instance, is predicated on the water divide as a system boundary within which mass balances are established. Useful as it is to have boundaries, many environmental processes manifest themselves most clearly in the transformations occurring at ecotones—system boundaries such as the riparian zone between terrestrial and aquatic systems, cell walls between abiotic and biotic systems, or the base of the food web between organisms and their environment, to say nothing of the boundary between “natural” and human systems.

To understand the cross-boundary transitions entails knowing something of both systems on their respective sides of that boundary. Thus, however, appealing it is in principle to study what happens across system boundaries, the resources required for a study rapidly escalate when attempting to cross a boundary. To compare “before and after” management interventions adds a temporal dimension that studies need to capture as well. This means either long term studies at selected sites, or substituting “space for time” with observations at many sites. It is also not just the required financial resources that escalate when crossing boundaries, there is also a need for more expertise as one moves between disciplinary boundaries in the study of soils, hydrology and ecology.

Unfortunately, in the competition for funds with other environmental threats, Hg research does not have the privilege of being counted among substances of “emerging” concern that have dedicated funding streams. Nonetheless, despite the challenges, McCarter et al. (2022), were able to show that it is possible to cross multiple system boundaries with a study of a management intervention (forest harvest) that spans 4 years (2 years of both pre- and post-treatment). The research infrastructure of the Marcell Experimental Forest was undoubtedly a key

ingredient in making this possible, which is also important to bear in mind as one prioritizes between research infrastructure investment and funding for research projects that require research infrastructures. Given the challenges, it is particularly important to take note of boundary-crossing studies like the subject of this commentary. McCarter et al. (2022) helps build the critical mass of studies needed to provide syntheses and model tests that have a chance of revealing the fundamental ecosystem processes that give the widely varying Hg responses to forest harvest observed at different sites.

4. Conclusions

When a critical mass of field studies is achieved, it should be synthesized into validated biogeochemical models that provide a sound basis for guiding management. On the way to that critical mass, there is a need to keep testing hypotheses with more boundary-crossing studies that distinguish the effect of a management intervention from the variation due to weather, season and local site differences. We plead particularly for studies that consider the waterscape as well as the landscape by following the local impact of land-use on aquatic ecosystems further downstream from the first point of stream measurement. The need to consider how a land-use impact on aquatic biota propagates downstream is due to the possibility of land use effects being attenuated downstream. This can be due either to dilution with runoff from unimpacted parts of the landscape (e.g., Schelker et al., 2014), or in-stream processes such as demethylation that might be favored in a well-oxygenated stream environment. These new studies can be purpose built on long-term research infrastructures or exploit operational management interventions.

Since much remains to be done to achieve a better understanding of how to manage Hg in the forest landscape, we think it is particularly important to acknowledge McCarter et al., 2022 for their paper which sets a standard for careful design, execution and analysis of comprehensive, boundary-crossing field studies.

Data Availability Statement

In writing this commentary no data were involved.

Acknowledgments

The authors would like to thank the many colleagues and students who have made it possible for us to conduct field studies. We also appreciate the artistic assistance of Alberto Zanella with the representation of trees in Figure 1.

References

- Bishop, K., Shanley, J. B., Riscassi, A., de Wit, H. A., Eklöf, K., Meng, B., et al. (2020). Recent advances in understanding and measurement of mercury in the environment: Terrestrial Hg cycling. *Science of the Total Environment*, 721, 137647. <https://doi.org/10.1016/j.scitotenv.2020.137647>
- Blume, T., van Meerveld, I., & Weiler, M. (2017). The role of experimental work in hydrological sciences—insights from a community survey. *Hydrological Sciences Journal*, 62(3), 334–337.
- Burt, T. P., & McDonnell, J. J. (2015). Whither field hydrology? The need for discovery science and outrageous hydrological hypotheses. *Water Resources Research*, 51(8), 5919–5928. <https://doi.org/10.1002/2014wr016839>
- Eklöf, K., Kraus, A., Futter, M., Schelker, J., Meili, M., Boyer, E. W., & Bishop, K. (2015). Parsimonious model for simulating total mercury and methylmercury in boreal streams based on riparian flow paths and seasonality. *Environmental Science and Technology*, 49(13), 7851–7859. <https://doi.org/10.1021/acs.est.5b00852>
- Eklöf, K., Lidskog, R., & Bishop, K. (2016). Managing Swedish forestry's impact on mercury in fish: Defining the impact and mitigation measures. *Ambio*, 45(2), 163–174. <https://doi.org/10.1007/s13280-015-0752-7>
- Environment. U. N. (2019). *Global mercury assessment 2018*. UN Environment Programme, Chemicals and Health Branch Geneva (ISBN: 978-92-807-3744-8).
- Futter, M. N., Poste, A. E., Butterfield, D., Dillon, P. J., Whitehead, P. G., Dastoor, A. P., & Lean, D. R. S. (2012). Using the INCA-Hg model of mercury cycling to simulate total and methyl mercury concentrations in forest streams and catchments. *Science of the Total Environment*, 424, 219–231. <https://doi.org/10.1016/j.scitotenv.2012.02.048>
- Golden, H. E., Knights, C. D., Conrads, P. A., Davis, G. M., Feaster, T. D., Journey, C. A., et al. (2012). Characterizing mercury concentrations and fluxes in a Coastal Plain watershed: Insights from dynamic modeling and data. *Journal of Geophysical Research*, 117(G1). <https://doi.org/10.1029/2011jg001806>
- Hsu-Kim, H., Eckley, C. S., Achá, D., Feng, X., Gilmour, C. C., Jonsson, S., & Mitchell, C. P. (2018). Challenges and opportunities for managing aquatic mercury pollution in altered landscapes. *Ambio*, 47(2), 141–169. <https://doi.org/10.1007/s13280-017-1006-7>
- Kirkby, M. J. (2004). Geomorphology: Critical concepts in geography. In *Hillslope geomorphology*, 16.
- Kristensen, P., Whalley, C., Zal, F. N. N., & Christiansen, T. (2018). *European waters assessment of status and pressures 2018*. EEA Report, (7/2018).
- Kronberg, R. M., Jiskra, M., Wiederhold, J. G., Björn, E., & Skjällberg, U. (2016). Methyl mercury formation in hillslope soils of boreal forests: The role of forest harvest and anaerobic microbes. *Environmental Science and Technology*, 50(17), 9177–9186. <https://doi.org/10.1021/acs.est.6b00762>
- Lidskog, R., Bishop, K., Eklöf, K., Ring, E., Åkerblom, S., & Sandström, C. (2018). From wicked problem to governable entity? The effects of forestry on mercury in aquatic ecosystems. *Forest Policy and Economics*, 90, 90–96. <https://doi.org/10.1016/j.forpol.2018.02.001>

- Mazur, M., Mitchell, C. P. J., Eckley, C. S., Eggert, S. L., Kolka, R. K., Sebestyen, S. D., & Swain, E. B. (2014). Gaseous mercury fluxes from forest soils in response to forest harvesting intensity: A field manipulation experiment. *Science of the Total Environment*, *496*, 678–687. <https://doi.org/10.1016/j.scitotenv.2014.06.058>
- Mazur, M. E., Eckley, C. S., & Mitchell, C. P. (2015). Susceptibility of soil bound mercury to gaseous emission as a function of source depth: An enriched isotope tracer investigation. *Environmental Science and Technology*, *49*(15), 9143–9149. <https://doi.org/10.1021/acs.est.5b01747>
- McCarter, C. P., Eggert, S. L., Sebestyen, S. D., Kolka, R. K., & Mitchell, C. P. (2022). Effects of clearcutting and residual biomass harvesting on hillslope mercury mobilization and downgradient mercury accumulation. *Journal of Geophysical Research: Biogeosciences*, *127*(4), e2022JG006826. <https://doi.org/10.1029/2022jg006826>
- McCarter, C. P., Sebestyen, S. D., Eggert, S. L., Kolka, R. K., & Mitchell, C. P. (2020). Changes in hillslope hydrology in a perched, shallow soil system due to clearcutting and residual biomass removal. *Hydrological Processes*, *34*(26), 5354–5369. <https://doi.org/10.1002/hyp.13948>
- McCarter, C. P., Sebestyen, S. D., Eggert, S. L., Kolka, R. K., & Mitchell, C. P. (2021). Differential subsurface mobilization of ambient mercury and isotopically enriched mercury tracers in a harvested and residue harvested hardwood forest in northern Minnesota. *Biogeochemistry*, *154*(1), 119–138. <https://doi.org/10.1007/s10533-021-00801-y>
- Schelker, J., Öhman, K., Löfgren, S., & Laudon, H. (2014). Scaling of increased dissolved organic carbon inputs by forest clear-cutting—What arrives downstream? *Journal of Hydrology*, *508*, 299–306. <https://doi.org/10.1016/j.jhydrol.2013.09.056>
- Shanley, J. B., & Bishop, K. (2012). Mercury cycling in terrestrial watersheds. In *Mercury in the environment: Pattern and process* (pp. 119–141). Sonke, J. E., Heimbürger, L. E., & Dommergue, A. (2013). Mercury biogeochemistry: Paradigm shifts, outstanding issues and research needs. *Comptes Rendus Geoscience*, *345*(5–6), 213–224. <https://doi.org/10.1016/j.crte.2013.05.002>
- Wu, P., Kainz, M. J., Bravo, A. G., Åkerblom, S., Sonesten, L., & Bishop, K. (2019). The importance of bioconcentration into the pelagic food web base for methylmercury biomagnification: A meta-analysis. *Science of the Total Environment*, *646*, 357–367. <https://doi.org/10.1016/j.scitotenv.2018.07.328>
- Zhu, S., Zhang, Z., & Žagar, D. (2018). Mercury transport and fate models in aquatic systems: A review and synthesis. *Science of the Total Environment*, *639*, 538–549. <https://doi.org/10.1016/j.scitotenv.2018.04.397>