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Review

Review of the Influence of Climate Change on the Hydrologic Cycling and Gaseous Fluxes of Mercury in Boreal Peatlands: Implications for Restoration

Randy Kolka [1,*], Caroline Pierce [2,†], Isabella Garrioch [2], Kevin Behrens [2] and Brandy M. Toner [2]

1 USDA Forest Service Northern Research Station, Grand Rapids, MN 55744, USA
2 Department of Soil, Water, and Climate, University of Minnesota-Twin Cities, St. Paul, MN 55108, USA; caroline.pierce5@gmail.com (C.P.); isabella.garrioch@umu.se (I.G.); behre170@umn.edu (K.B.); toner@umn.edu (B.M.T.)
* Correspondence: randall.k.kolka@usda.gov
† Current address: Region 10 Water Division, Environmental Protection Agency, Portland, OR 97205, USA.

Abstract: Mercury (Hg) is a pollutant that bioaccumulates in the food web, leading to health issues in humans and other fauna. Although anthropogenic Hg deposition has decreased over the past 20 years, our watersheds continue to be sources of Hg to downstream communities. Wetlands, especially peatlands in the Boreal Region of the globe, play a vital role in the formation of bioaccumulative methylmercury (MeHg). Few studies have assessed how increases in temperatures such as those that have already occurred and those predicted will influence the hydrologic transport of Hg to downstream communities or the net fluxes of gaseous Hg. The results indicate that peatland pore water concentrations of MeHg are increasing with ecosystem warming, and to some degree with elevated carbon dioxide (eCO_2) in the Spruce and Peatland Responses Under Changing Environments (SPRUCE) experiment at the Marcell Experimental Forest (MEF) in northern Minnesota, USA. Similar to SPRUCE, in the Biological Response to A Changing Environment (BRACE) experiment in Canada, mesocosm pore water MeHg concentrations increased with soil warming. However, long-term peatland watershed streamflow fluxes of MeHg at the MEF indicate that the competing effects of climate warming and decreased atmospheric deposition have led to overall decreases in watershed MeHg transport. Mesocosm studies in the PEATCOSM experiment in Upper Michigan, USA, indicate that simulated fluctuating water tables led to higher concentrations of MeHg in peatland pore water that is available for downstream transport when water tables rise and the next runoff event occurs. Results from a winter peatland soil freeze/thaw simulation from large mesocosm cores from Jennie's Bog at the MEF indicate higher total Hg (THg) upon soil thawing but lower MeHg, likely a result of cold temperatures limiting methylation during thawing. Although there are lower MeHg concentrations after thawing, more THg is available for methylation once soils warm. Results from PEATCOSM and the literature also suggest that plant community changes that result in higher densities of sedges also lead to elevated MeHg in pore water. From a climate warming perspective, it appears that two complementary mechanisms, both related to decomposition, are at play that lead to increased pore water MeHg concentrations with warming. First, warming increases decomposition rates, leading to a higher availability of many ions, including Hg (and sulfur) species. Higher decomposition rates also lead to increases in soluble carbon which complexes with Hg species and assists in downstream hydrologic transport. However, if streamflow is decreasing with climate change as a result of landscape-level changes in evapotranspiration as suggested at MEF, the combination of less direct watershed Hg deposition and lower streamflow results in decreases in the watershed transport of MeHg. Given changes already occurring in extreme events and the rewetting and restoration of hydrology during peatland restoration, it is likely that methylation and pore water MeHg concentrations will increase. However, the landscape-level hydrologic cycle will be key to understanding the connection to downstream aquatic communities. Finally, gaseous Hg fluxes increase with warming and lead to decreases in peatland pools of Hg that may influence future availability for downstream transport.

Citation: Kolka, R.; Pierce, C.; Garrioch, I.; Behrens, K.; Toner, B.M. Review of the Influence of Climate Change on the Hydrologic Cycling and Gaseous Fluxes of Mercury in Boreal Peatlands: Implications for Restoration. *Water* **2024**, *16*, 1154. https://doi.org/10.3390/w16081154

Academic Editor: Zbigniew Kabala

Received: 22 January 2024
Revised: 20 March 2024
Accepted: 15 April 2024
Published: 19 April 2024

Keywords: methylmercury; warming; ecosystem manipulation; extreme events; carbon dioxide

1. Introduction

Mercury (Hg) is a pollutant that bioaccumulates in the food web, especially in ecosystems that have many trophic levels such as aquatic environments [1]. As a result, humans and wildlife that consume aquatic organisms are at high risk of negative health effects that include nervous system and neurological disorders [2]. Although research has been conducted over the past 30+ years in developing an understanding of how Hg cycles in the environment, little is known how climate change factors such as warming, elevated carbon dioxide (eCO_2), and changing precipitation regimes influence terrestrial Hg cycles, especially in hotspots of methylation such as Boreal peatlands [3]. Global assessments have indicated that warming is anticipated to enhance soil and sediment methylation, that the increased frequency and severity of extreme events will increase Hg transport to surface waters, and that eCO_2 could enhance terrestrial ecosystem productivity, leading to higher Hg sequestration in terrestrial systems [4–6]. However, few studies assessing the effects of climate change on mercury cycles have been conducted at the ecosystem scale. Notably, wetlands, especially northern peatlands, play a vital role in the global cycling and transport of Hg to downstream areas for bioaccumulation. However, peatlands across the Boreal Region have been drained for a variety of purposes related to agriculture, forestry, and development, many of which have since been abandoned because of poor fertility and difficulty in managing soil water [7].

Restoring peatlands is one of our best opportunities to increase the storage of carbon in our landscapes to mitigate greenhouse gas emissions and rebuild relatively stable soil carbon pools in degraded ecosystems. Wetlands, and more specifically peatlands, have been identified globally as a priority ecosystem for restoration because of the high potential for soil carbon gains compared to other degraded ecosystems [8,9]. The restoration of previously mined peatlands, either for horticulture or fuel, also presents an opportunity to increase ecosystem carbon sequestration [10]. In Minnesota alone, there are an estimated 4000 km of peatland ditches, leading to an estimate of nearly 4 Tg of carbon lost to the atmosphere since disturbance [11], equivalent to about 10% of annual emissions from all sectors in Minnesota in 2020 [12]. However, no research has addressed how peatland restoration influences mercury cycling, including methylation, transport, and ultimately bioaccumulation in the food web, especially when compounded with the interacting effects of climate change.

In this contribution, we synthesize previous and ongoing research addressing climate change effects on the ecosystem-level peatland cycling and transport of Hg as well as the net gaseous fluxes of Hg. There are few studies that have either long-term records (15+ years) or manipulate climate to assess the effect of variables such as warming, changes in precipitation regimes, and eCO_2 on Hg cycles in peatlands. Although there have been efforts to address the implications of climate change on Hg cycling on topics such as permafrost thawing (e.g., [13]) and the increased frequency and intensity of fire (e.g., [14]) that is relevant to peatlands, few studies have used controlled, designed manipulations to directly assess changes in climate variables (i.e., temperature, hydrology, and eCO_2) on Hg cycling in peatlands. The focus of this review is to ascertain what we have learned from the paucity of peatland climate manipulation studies, and one long-term peatland watershed study, on methylation and associated availability for MeHg and to apply that knowledge to understand likely Hg responses to peatland restoration. We do not consider small-scale laboratory incubation studies, because of the difficulty in application to field conditions (e.g., [15]), or more general studies assessing the factors that influence methylation (e.g., [16]) unless there is some aspect of this study related to changes in climate.

As mentioned above, we are not aware of any studies that have investigated how drained peatland restoration influences Hg cycles. Studies have investigated Hg cycles in constructed peatlands such as those implemented to mitigate pollution from the oil sands region of Canada [17]; however, given the massive replacement of soils, we do not consider those studies applicable to ditched peatland restoration. Considering the current interest in restoring drained peatlands to mitigate CO_2 concentrations in our atmosphere (although it exacerbates methane: [18]) and our basic understanding of peatland Hg cycling processes with climate change, expected Hg pore water concentrations and hydrologic fluxes (and gaseous net fluxes of Hg) to restoration and the impacts of climate change on restoration can be predicted.

2. Review of Boreal Peatland Climate Studies

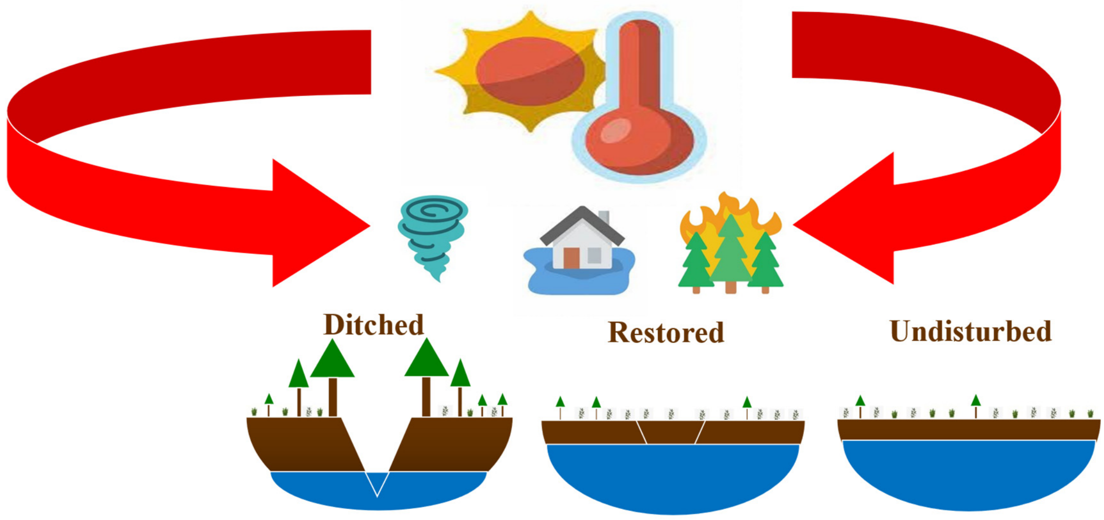
To assess the effect of climate change on Hg cycling, we reviewed studies that either had long-term data that could address ongoing changes in climate, or manipulative studies that have simulated some aspect of climate change. The paucity of research on this topic led to only five studies, some of which are completed, others that are ongoing (Table 1). At the Marcell Experimental Forest (MEF), we have been monitoring Hg in various waters, including streamflow, since 2002. Over that time, we have experienced warming temperatures that have influenced peatland watershed Hg fluxes via streamwater [19]. Also located at the MEF is the SPRUCE (Spruce and Peatland Responses to Changing Environments) experiment, hosted by the USDA Forest Service and the Department of Energy's Oak Ridge National Laboratory, where we are conducting whole peatland ecosystem warming, both above and belowground, as well as exposing half the enclosures to eCO_2 [20]. We have been monitoring pore water as well as passive gaseous Hg fluxes during SPRUCE. In a similar smaller-scale study at the BRACE (Biological Response to A Changing Environment) site in central Ontario, scientists applied both passive and active warming to peatland field plots [21]. The PEATCOSM Study, hosted by USDA Forest Service and Michigan Technological University in Houghton, MI, investigated the effect of varying water tables and changes in plant communities anticipated with climate change [22]. Finally, results from a mesocosm study simulating winter responses on the effects of warming on freeze/thaw cycles on THg and MeHg in peatland pore waters from Jennie's Bog at the MEF were also considered [23]. We do not thoroughly address the implications of climate change on import ions and microorganisms responsible for methylation (e.g., sulfate- and iron-reducing bacteria and methanogens) and Hg transport (e.g., DOC: dissolved organic carbon), though generally, it does not appear that climate change will limit the availability of reducers and associated ions, and there are likely increases in peatland-derived DOC fluxes as a result of greater decomposition with warming [24]. Also, demethylation is not considered, although Arctic lake sediment data suggest demethylation changes little with temperature and net rates of methylation will increase with warming [25].

Table 1. Studies considered in the review of climate change impacts on Hg cycling (* soils for the PEATCOSM mesocosm study from northern Minnesota). Hg water data include both THg and MeHg for all studies.

Name	Location	Manipulation/Source	Hg Data	Time Period
MEF	Minnesota	Long-term records	Streamwater	2002–2017 [19]
SPRUCE	Minnesota	Warming and eCO_2	Pore water, passive gas	2017–2019 [26–28]
BRACE	Canada	Warming	Pore water	2017–2019 [21]
PEATCOSM	Michigan *	Hydrology and plants	Pore water, dynamic gas	2012–2014 [29,30]
Jennie's Bog	Minnesota	Freeze/thaw	Pore water	2015 [23]

3. Climate Change Influences on Peatland Mercury Cycling

Climate change factors that influence peatland hydrologic Hg cycling include those affected by warming, changes in precipitation regimes and extreme events and associated water tables, and eCO_2 (Figure 1). Climate warming continues to be at the forefront of the global climate change discussion, with 2023 being the warmest year on record going back to 1850 [31]. Also presented is a brief review of the implications of climate on gaseous fluxes of Hg.



Water Flux	High	Low	High/Low
Water Table Variation	High	Low	Low
Spring Pore Water MeHg Conc.	Low	Low	Low
Summer/Fall MeHg Pore Water Conc.	Medium	High	High
Net Gaseous Hg Flux	Low	Low	High
Overall Downstream Impacts	High	Low	High/Low

Figure 1. Mercury cycling interactions with climate and ditched, restored, and undisturbed peatlands. Climate interactions include warming and those impacts of extreme events such as storms (tornados and hurricanes), flooding, and fires resulting from droughts. The associated table indicates the direction of pore water MeHg concentrations, and our relative confidence is denoted by color with green denoting confident and red denoting little confidence because the interaction is untested. The water flux and overall downstream impact from undisturbed peatlands are high when directly connected to surface waters (e.g., streams) and low for seepage wetlands with no outlets. Although the images are small, the ditched and undisturbed peatlands include sedges while the restored peatland does not.

3.1. Warming Effects on Peatland Hydrologic Cycling of Mercury

The SPRUCE experiment is the most extensive ecosystem climate manipulation worldwide. Sponsored by the US Department of Energy, SPRUCE is testing the effects of both aboveground and belowground warming, and eCO_2 on peatland biogeochemical responses, including Hg cycling. Treatments include warming of 0, 2.25, 4.5, 6.75, and 9 °C above ambient conditions, and each temperature treatment has both an ambient (~420 ppm) and elevated (~900 ppm) CO_2 environment. Mercury measurements in SPRUCE began before treatments were installed [32] and have commenced over the first 8 years of a planned 10-year experiment. Since above and belowground warming has been initiated, the results suggest increases in MeHg concentrations in pore water with increasing temperatures [26]. In the BRACE experiment, replicated small plots were installed and supplied with active belowground (1 growing season) and passive aboveground heating (3 growing seasons) in two fen peatlands, one dominated by *Sedge* spp. and the other dominated by *Sphagnum* spp. Collars 1 m in diameter were inserted down to 40 cm in the peat with 8 heated and 8 unheated replicates per treatment, per peatland [21]. Warming led to increases in MeHg concentrations in pore waters but there was little response of THg, indicating that the ratio of MeHg:THg also increased. Concomitant with increases in MeHg in pore waters were increases in DOC and presumably microbial communities responsible for decomposition [21].

3.2. Water Balance Effects on Peatland Hydrologic Mercury Cycling

It is difficult to separate the influence of warming on increasing mercury methylation and the influence climate change has on evapotranspiration and the catchment water balance. In PEATCOSM, relatively undisturbed 1 m^3 peat monoliths were extracted from northern Minnesota and transported via flatbed truck to the PEATCOSM facility in Houghton, MI. Monoliths were inserted into Teflon-coated boxes with the ability to control precipitation inputs using rainout shelters. Treatments included two hydrological scenarios, one that included typical variability and average water table position, and a second that had a higher variability and deeper average water table level. The deeper and more variable water table treatment was designed to simulate changes in extreme weather events and a predicted drier summer environment [22]. PEATCOSM also tested the effect of plant communities with mesocosms that had ericaceous shrubs only (sedges removed), sedges only (shrubs removed), and sedges + ericaceous shrubs (control) (see discussion in Section 3.4). Pore water THg and MeHg and the ratio of MeHg:THg significantly increased in the zone of water table fluctuation in the deeper and more variable water table treatment, indicating that possible increases in decomposition in the aerobic zone drove Hg methylation [29]. Once rewetted by a major precipitation or melt event, much higher MeHg hydrologic flux rates that include the elevated MeHg in deeper pore waters can lead to enhanced downstream transport [33].

3.3. Elevated CO_2 Effects on Peatland Hydrologic Mercury Cycling

In SPRUCE, data from multiple linear regressions suggest a positive eCO_2 response on peat pore water concentrations of Hg [26], possibly a result of increased overall plant productivity. Plant processes such as photosynthesis and respiration [34] and soil nutrient availability are greater [35] in SPRUCE under eCO_2, which also appears to increase the methylation of Hg. The Th-increased methylation response may be a result of the enhanced decomposition and associated availability of reducing ions and microbes responsible for methylation. No other studies have directly tested the effect of eCO_2 on peatland Hg cycles. Other indirect effects of eCO_2 on plant communities and implications for Hg cycling are considered below.

3.4. Interacting Effects of Climate Change on Peatland Hydrologic Mercury Cycling

Long-term streamflow records (55 years) and Hg concentration measurements (2001–2017) from a peatland watershed at the MEF indicate decreasing streamflow that is a result of increased landscape evapotranspiration and associated decreases in runoff ratios, possibly because of the increasing plant productivity with rising atmospheric CO_2 levels [19]. During the same time period, mean temperatures have increased 1.8 °C, leading to seemingly higher decomposition, nutrient availability, and also increased ecosystem net primary production. In addition, Hg in wet deposition has decreased precipitously since the 1990s as a result of clean air legislation. The combination of factors leading to less streamflow with higher ecosystem productivity, and lower deposition rates, has outweighed increases in methylation rates resulting from climate warming, leading to lower overall watershed MeHg transport downstream [19]. Changes in peatland watershed fluxes of MeHg are expected to continue to decrease if decreases in runoff ratio and wet deposition also continue; however, increases in air temperature that stimulates additional methylation could counteract or mediate this response.

In PEATCOSM, plant community changes resulting from climate change that led to higher densities of sedges and associated soil oxygen with depth also led to elevated MeHg in pore water because of presumably higher decomposition rates [36]. Sedges do appear to outcompete *Sphagnum* spp. and ericaceous shrubs in fen and poor fen peatlands with warming and variable water tables resulting from climate change [37,38]. However, in more nutrient-poor, lower-pH bogs, sedges and other graminoids tend to be outcompeted by ericaceous shrubs with warming [39]. In the SPRUCE bog, warming is leading to much greater densities of ericaceous shrubs at the expense of the *Sphagnum* spp., which have declined precipitously in the warmest treatments [40]. The density of sedges began low and has remained low with warming in SPRUCE. Although eCO_2 tends to increase ecosystem aboveground net primary productivity and can favor sedges in non-peatland environments [41], eCO_2 has not been an important factor in plant community changes in SPRUCE [39].

There is little known about how changes in winters with warming across the Boreal Region influence Hg cycles, especially in the spring, where the bulk of annual Hg transport in watershed runoff occurs annually during snowmelt [42]. It is anticipated that warmer winter temperatures will lead to less snow and more rain in the Boreal Region [43]. Less snow leads to lower insulation and colder soil conditions and more frequent and deeper soil frost, at least until warming overwhelms the formation of soil frost [44]. Mesocosm manipulations of annual temperature cycles that include variations in freeze/thaw rates indicate that peat pore water THg is elevated after the spring thaw, but MeHg concentrations decrease during the freeze/thaw event likely as a result of little microbial activity near freezing temperatures [23]. Elevated pore water THg would presumably be available for methylation once soils warm. Northern latitudes are predicted to experience higher-frequency freeze/thaw events with decreases in snowfall leading to soils being more exposed to variable air temperatures [45]. While the freeze/thaw cycle leads to lower MeHg in peat pore water, it is unknown whether an increased frequency of freeze/thaw cycles or just a shorter soil frost season with climate warming affects early-season MeHg formation and transport.

3.5. Warming Effects on the Gaseous Fluxes of Hg

Warming has also led to elevated net gaseous peatland Hg fluxes in SPRUCE [27,28,30], which decreases the overall future availability of Hg for methylation locally but is available for redeposition elsewhere, potentially in a new methylating environment. Similar to warming, gaseous fluxes of Hg increase with lower water tables [30], likely also a result of higher decomposition rates in the extended aerobic acrotelm zone.

4. Implication of Peatland Restoration and Subsequent Climate Change on Hg Cycles

No studies that we are aware of have assessed how peatland restoration influences Hg cycles, especially in light of possible interactions with climate change. Nonetheless, given previous work on undisturbed peatland systems, we can predict the likely effect of restoration (rewetting and re-establishing plant communities) on Hg cycles and the possible subsequent drivers of climate change on those cycles (Figure 1). The restoration of peatland hydrology will decrease aeration in the upper peatland soils, leading to likely increases in methylation potential; however, based on undisturbed peatlands, it is wetting (anerobic) and drying (aerobic decomposition) cycles that lead to the greatest methylation and downstream transport [29,33]. As the frequency and intensity of extreme drying and wetting events increase with climate change in the Boreal Region [46,47], both restored and undisturbed peatlands will likely become larger sources of MeHg. Also, continued increases in temperatures, especially those in shoulder seasons (spring and fall), and over winter, increase the decomposition "season", also leading to elevated MeHg in peatland pore waters and gaseous fluxes of Hg. The establishment of oxygen pumping plant species such as sedges during restoration can also lead to enhanced MeHg in pore water and the potential for downstream transport [36]. Collectively, the data indicate that to lessen the potential for Hg methylation and downstream transport, and gaseous Hg fluxes, it will be important to manage water levels to keep restored peatland water tables near the surface and to restore plant communities without sedges.

5. Conclusions

The combination of results indicates that a warming climate increases peatland soil water MeHg, leading to the potential for higher MeHg in peatland runoff and downstream transport (Figure 1). However, decreases in Hg deposition and decreases in streamflow resulting from greater landscape-level evapotranspiration can offset warming influences. Importantly, changes in precipitation patterns and associated extreme events will likely lead to elevated methylation rates with more variable water tables. Peatland plant community changes that result in less sedges will likely lower MeHg concentrations in peatland pore water. Although the mechanism is uncertain (possibly increased decomposition), eCO_2 was a significant predictor of increased MeHg concentrations in pore water. Also, changes to winter conditions leading to increased soil freezing can decrease methylation rates but cause higher concentrations of THg to be available for methylation upon thawing. Moreover, net gaseous Hg fluxes have been shown to increase with warming.

Because there is no available literature on the effect of peatland restoration on Hg cycles, we postulate on how restoring ditched peatlands will influence water cycles, soil aeration, and associated decomposition, and the potential for methylation. Overall, we hypothesize that peatland restoration will lead to higher methylation rates than drained peatlands but lower water fluxes and, hence, lower downstream impacts.

6. Future Directions

New studies that directly measure hydrologic and atmospheric fluxes of Hg using before-after-control-impact (BACI) designs or across a gradient of ditched, restored, and undisturbed natural reference systems are necessary to directly assess the effect of peatland restoration on Hg cycles. Also, we need studies that assess how ions and microorganisms responsible for methylation respond to water table/aeration changes resulting from restoration and changes in climate conditions such as extreme events. We need to consider layering climate change variables such as warming, eCO_2, or a water table simulation during restoration studies that integrate both the effect of climate change and restoration to inform future peatland restorations.

Recently funded studies in northern Minnesota have begun to assess the effect of peatland restoration on Hg fluxes. A challenge in these new studies that has been questioned by various organizations is how to monitor for hydrologic fluxes of Hg following the general practice of plugging the ditch during restoration. Although some undisturbed peatlands have natural drainages (i.e., streams) that exit the watershed, other seepage peatlands do not (see Figure 1 for differences in downstream impacts), and when the ditch is plugged during restoration, there tends to be little drainage. The question then becomes how does one assess the effect of restoration on downstream impacts? Because there is a direct relationship between pore water Hg concentrations and watershed fluxes [48], monitoring pore water as an indicator of the potential impacts on downstream waters would be an alternative approach.

Although overlaying a climate change treatment on a Hg/peatland restoration study would be very insightful, the first study priority is to assess the direct effects of restoration alone on Hg cycles. Data from pore water, watershed, and atmospheric fluxes of Hg from field studies could then be modeled under various climate change scenarios [49,50] if field studies that integrate both restoration and climate are not feasible.

Author Contributions: Conceptualization, R.K.; methodology, R.K., C.P., I.G., K.B. and B.M.T.; formal analysis, R.K., C.P., I.G., K.B. and B.M.T.; investigation, R.K., C.P., I.G., K.B. and B.M.T.; writing—review and editing, R.K., C.P., I.G., K.B. and B.M.T.; visualization, R.K. and B.M.T.; supervision, R.K. and B.M.T.; project administration, R.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Conflicts of Interest: The author declares no conflicts of interest.

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