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RESEARCH ARTICLE

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Key Points:

- Lower streamwater methylmercury concentrations and yields due to compounding effects of climate change and cleaner air
- Lower wet atmospheric mercury deposition and runoff ratios offset higher air temperatures, resulting in lower methylmercury concentrations
- Headwater peatland catchments will continue to be net sinks of total mercury; thus, we may not see a net flushing of anthropogenic mercury

Supporting Information:

Supporting Information may be found in the online version of this article.

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Methylmercury Export From a Headwater Peatland Catchment Decreased With Cleaner Emissions Despite Opposing Effect of Climate Warming

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Abstract Peatlands are sources of bioaccumulating neurotoxin methylmercury (MeHg) that is linked to adverse health outcomes. Yet, the compounding impacts of climate change and reductions in atmospheric pollutants on mercury (Hg) export from peatlands are highly uncertain. We investigated the response in annual flow-weighted concentrations (FWC) and yields of total-Hg (THg) and MeHg to cleaner air and climate change using an unprecedented hydroclimatic (55-year; streamflow, air temperature, precipitation, and peatland water tables), depositional chemistry (21-year; Hg and major ions), and streamwater chemistry (~17-year; THg, MeHg, major ions, total organic carbon, and pH) data sets from a reference peatland catchment in Minnesota, USA. Over the hydroclimatic record, annual mean air temperature increased by ~1.8°C, while baseflow and the efficiency that precipitation was converted to runoff (runoff ratio) decreased. Concurrently, precipitation-based deposition of sulfate and Hg declined, where wet Hg deposition declined by ~3–4 μg Hg m⁻². Despite declines in wet Hg deposition over the study period, the catchment accumulated on average 0.04 ± 0.01 g Hg ha⁻¹ yr⁻¹ based on wet Hg deposition minus THg yield alone. Annual MeHg FWC was positively correlated with mean annual air temperatures ($p = 0.03$, $r = 0.51$), runoff ratio ($p < 0.0001$, $r = 0.76$), and wet Hg deposition concentration ($p < 0.0001$, $r = 0.79$). Decreasing wet Hg deposition and annual runoff ratios counterbalanced increased peatland MeHg production due to higher air temperatures, leading to an overall decline in streamwater MeHg FWC. Streamwater MeHg export may continue to decrease only as long as declines in runoff ratio and wet Hg deposition persistently outpace effects of increased air temperature.

Plain Language Summary Climate change and cleaner air are unequivocally altering the movement of toxic mercury and methylmercury. Using long-term and broad environmental measurements, a stream fed by a peatland exhibited decreased methylmercury levels that correlated with lower wet mercury deposition concentration and lower net water yields from the catchment (runoff ratio), which offsets potential increases of methylmercury due to elevated air temperatures. As such, methylmercury export from peatland catchments will likely continue to decrease over time if climate change continues to accelerate the reduction of runoff ratios and atmospheric wet mercury deposition further decreases. Adaptation to future climate and environmental changes would greatly benefit from additional and longer-integrated multidisciplinary data sets. Without such long-term and integrated measures critical insights, such as those gained from this study, would not be possible.

1. Introduction

Climate change is altering stream and river water chemistry (Li et al., 2022; Meyer-Jacob et al., 2019). In boreal and hemi-boreal catchments, wetlands, including peatlands, play a critical role in regulating water chemistry (Gorham et al., 1985; Lam et al., 2022; Schelker et al., 2014), particularly the contaminant mercury (Hg) (Kronberg et al., 2018; Lam et al., 2022; Mitchell et al., 2008b; Tjerngren, Karlsson, et al., 2012). Yet, other concurrent environmental disturbances potentially compound or mask changes in water chemistry from climate change (Li et al., 2021). Even in ecosystems that are assumed to be least impacted and that serve as reference sites, decreased atmospheric deposition of pollutants, such as sulfate (SO₄²⁻) or Hg, with cleaner air legislation (Lee et al., 1998; Pannatier et al., 2011; Sickles & Shadwick, 2015; Zhang et al., 2019) may mask or enhance effects of climate change on aqueous contaminants like Hg. Untangling relative effects of climate and environmental change in reference ecosystems is critical to inform science, management, and policy.

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The bioaccumulating neurotoxin methylmercury (MeHg) is a contaminant of global concern linked to adverse human health conditions (O'Connor et al., 2019). Mercury mobilization, either as MeHg or total-Hg (THg), from uplands and wetlands to downstream water bodies is controlled by interacting hydrological and biogeochemical processes that affect both concentrations and fluxes (Bishop et al., 2020; Branfireun et al., 2020; McCarter, Sebestyen, et al., 2022; O'Connor et al., 2019; Woerndle et al., 2018). Peatlands are often disproportionately large sources of MeHg to surface waters relative to upland mineral soils (Branfireun et al., 1998; Lam et al., 2022; Skjellberg et al., 2003; Woerndle et al., 2018). The net export of MeHg from peatlands to surface waters is a balance between Hg methylation and demethylation (Kronberg et al., 2018; Tjerngren, Meili, et al., 2012), MeHg solubility (Skjellberg, 2008), and the hydrological transport of Hg from a peatland to a stream (McCarter, Sebestyen, et al., 2022). In peatlands, wet atmospheric Hg deposition is usually lower than Hg associated with litterfall (Grigal et al., 2000; Woerndle et al., 2018) but is often more readily available to resident microbial communities (Hsu-Kim et al., 2013), likely resulting in quicker incorporation into the Hg cycle. For instance, Hudelson et al. (2023) observed a strong correlation between atmospheric Hg and climate signals with Hg concentrations in Arctic char. Moreover, elevated atmospheric SO_4^{2-} deposition to peatlands has been linked to increased MeHg concentrations in receiving streamwater (Jeremiason et al., 2006; McCarter, Sebestyen, et al., 2022) but the effect is rapidly reversible once SO_4^{2-} additions decline (McCarter et al., 2017; McCarter, Sebestyen, et al., 2022). While both SO_4^{2-} and Hg deposition have decreased in response to clean air legislation, climate change likely affects the ecohydrological and biogeochemical process that control Hg transformation and movement through catchments (Bishop et al., 2020; Gerson & Driscoll, 2016; Yang et al., 2016).

Model simulations of the effect of climate change on Hg cycling are unclear due to large uncertainties associated with both climate change and Hg biogeochemical processes (Golden et al., 2013), while in-situ peatland experimental warming of boreal peatlands have shown increases in both inorganic Hg and MeHg porewater concentrations (Sun et al., 2023). Yet, few long-term empirical data sets exist to document contemporary responses and trends, particularly at catchment scales. One such long-term study used a 10-year Hg deposition and streamwater Hg record to highlight a decrease in both streamwater THg and MeHg concentrations in the inflow and outflow of a temperate lake in the Adirondack region, USA, driven by declining Hg deposition (Gerson & Driscoll, 2016). However, the response in Hg export from peatland-rich catchments due to the long-term and interacting effects of climate change and cleaner air is uncertain (Sonke et al., 2023), largely due to only a few select places with sustained, routine Hg monitoring in streamwater in conjunction with relevant environmental data collection.

We present an unprecedented 17-year record of annual flow-weighted concentrations (FWC) and yields of THg and MeHg and frame that record within longer 55-year hydroclimatic and ~30-year atmospheric deposition records for Hg and basic chemistry. Combined, we used these data sets to quantify directionality and magnitude of responses and to discern mechanisms that govern Hg concentrations and yields in a headwater hemi-boreal stream. Specifically, we determined: (a) the direction and magnitude of annual THg and MeHg FWC and yields (i.e., Hg export) at an undisturbed reference peatland catchment, (b) the driving climatological, hydrological, and biogeochemical processes (i.e., drivers) of annual Hg export, (c) the impact of intra-annual variability in key drivers on annual Hg export, and (d) explore the trajectory of Hg export by placing the key drivers into the long term hydroclimatic and biogeochemical observations. Our findings are the first catchment-scale observations that show how both THg and MeHg export from peatland-dominated catchments change with interacting effects of cleaner air and climate change.

2. Study Site

The S2 catchment has served as a minimally disturbed reference catchment at the Marcell Experimental Forest (MEF) since the 1960s. The MEF is located in north-central Minnesota with a continental climate and average daily temperature of 3.5°C and annual precipitation of ~800 mm (Sebestyen et al., 2021). The 9.7 ha catchment is comprised of a 6.5 ha upland surrounding a centrally located 3.0 ha forested ombrotrophic bog peatland with a 0.2 ha forested lagg (Hill et al., 2016; Sebestyen et al., 2021). The peatland has an overstory of *Picea Marianna* and an understory of *Sphagnum* mosses, graminoids, and ericaceous shrubs (Verry & Janssens, 2011). The upper ~30 cm of peat is derived from the current forested bog vegetation and transitions from poor fen to rich fen peat deeper through the peat profile (Verry & Janssens, 2011). The peat depth is from 10s of cm to ~7 m (Verry & Janssens, 2011). The uplands are dominated by a mixed deciduous forest of *Populus tremuloides* with other coniferous and deciduous trees (Verry & Janssens, 2011).

Table 1
Data Sources for the Different Measured Parameters, With Measurement Frequency and the Calculations Used for Each Parameter

Data source	Parameters	Measurement frequency	Calculations
Sebestyen et al. (2021)	Streamflow, air temperature, precipitation, adjacent aquifer and peatland water tables	Daily (1962–2017)	All Parameters: Annual and seasonal means, standard deviations, and deviations from long-term mean (1962–1972); event flow and baseflow, proportion of baseflow/event flow; annual and seasonal total precipitation (P); Annual and seasonal runoff ratio, standard deviation of annual runoff ratio (calculated using monthly data); Annual mean potential evapotranspiration (PET), Annual and 10-year mean potential evapotranspiration; P-PET
Sebestyen, Lany, et al. (2022)	THg, MeHg, K ⁺ , Na ⁺ , Ca ²⁺ , Mg ²⁺ , Fe ^{2+/3+} , Cl ⁻ , SO ₄ ²⁻ , PO ₄ ³⁻ , pH, TOC	~bi-weekly (1998–2017, 8–24 samples per year depending on streamflow/no flow)	MeHg/THg: annual and seasonal FWC and yields, standard error of FWC and yields; Cations/anions/pH/TOC: annual FWC and yields
National Atmospheric Deposition Program (MN16) (2021)	Total wet Hg deposition, precipitation wet Hg deposition concentration	Monthly (1996–2017)	Seasonal and annual totals, means, and standard deviation
National Atmospheric Deposition Program (MN16) (2020)	Precipitation concentration of: SO ₄ ²⁻ , Cl ⁻ , NH ₄ ⁺ , NO ₃ ⁻ , Br ⁻ , K ⁺ , Na ⁺ , Ca ²⁺ , Mg ²⁺ , pH	Monthly (1978–2017)	Annual and seasonal means

The peatland and upland both hydrologically feed the lagg surrounding the peatland, which ultimately drains into a small stream (Sebestyen et al., 2011; Verry et al., 2011). The ombrotrophic bog peatland and surrounding uplands are perched above an adjacent aquifer, isolating aquifer water from entering either the uplands or peatland (Verry et al., 2011). Consequently, the uplands provide local groundwater to the lagg through relatively shallow permeable loess sandy loam horizon overlying a low-permeability Koochiching clay loam till (Mitchell et al., 2009; Verry et al., 2011). The water flow at the catchment stream outlet is monitored at a v-notch weir (Verry et al., 2018).

3. Methods

Data were collected within the long-term ecosystem research program at the S2 catchment on the MEF. We are not aware of any other multi-decadal, uninterrupted record for boreal or hemi-boreal streamwater MeHg and THg. The hydroclimatic record consists of the annual mean, minimum, and maximum air temperature, total precipitation, catchment runoff, baseflow/event flow contributions, and the fraction of precipitation that occurs as streamflow (runoff ratio) (Sebestyen et al., 2021). Atmospheric deposition records include the Hg concentration, total mass of wet Hg deposition, and total wet-deposited mass of SO₄²⁻ and hydrogen ions (National Atmospheric Deposition Program (MN16), 2020, 2021). Streamwater chemistry samples were collected 8 to 24 times a year and analyzed for THg, MeHg, total organic carbon (TOC), pH, and major anions and cations (Sebestyen, Lany, et al., 2022). The collated data set also includes calculated runoff ratios (RR), daily baseflow/event flow, proportion event flow, potential evapotranspiration (PET), the 10-year average PET, and the annual potential water deficit/surplus (P-PET) (Table 1). All the data and associated QA/QC come from freely accessible data publications and aggregated annual and/or seasonal values for analysis (Table 1). Additional Hg QA/QC analysis is presented McCarter, Sebestyen, et al. (2022) and the associated supplemental information and references therein.

3.1. Data Calculations

Solute yields were calculated following Sebestyen and Kyllander (2017), whereby concentrations were linearly interpolated between water chemistry samples. Daily yield was determined by multiplying the sample concentration or interpolated concentration by daily streamflow. Daily yields were either seasonally or annually summed following Sebestyen and Kyllander (2017) and then divided by catchment area (9.7 ha) and the total volumetric streamflow to determine annual FWC. Seasons were defined as: Spring: March-May, Summer: June-August, Fall: September-November, Winter: December-February.

The 95% confidence intervals for annual MeHg and THg FWC and yields were determined following Hope et al. (1997). The 95% confidence interval of the monthly FWC, $\text{var}(C_F)$, were determined using,

$$\text{var}(C_F) = \left[\sum (C_i - C_F)^2 \cdot \frac{Q_i}{Q_n} \right] \sum \frac{Q_i^2}{Q_n^2} \quad (1)$$

where, C_F is the monthly FWC for a given solute, C_i is the instantaneous concentration for a given sample, Q_i is the instantaneous discharge, and Q_n is the sum of Q_i . The 95% confidence interval of the yields were then determined by,

$$\text{std.error}(Y) = F \cdot \sqrt{\text{var}(C_F)} \quad (2)$$

where, F is the total annual discharge.

Wet deposition of most solutes and total wet deposition of Hg were measured as part of the NADP. The NADP site is in the MEF about 2 km north of the S2 catchment.

Streamflow was separated into base and event flow following Nathan and McMahon (1990), with a recession coefficient of 0.8. Runoff ratios were calculated by,

$$RR = \frac{Q_s}{P} \quad (3)$$

where, Q_s is the annual total streamflow (mm), and P is the annual total precipitation (mm) for a given time period.

Calculated daily PET (mm) was determined following Hargreaves and Samani (1985),

$$PET = 0.0023 \cdot 0.408 \cdot R_a (T_{avg} + 17.8)(T_{max} - T_{min})^{0.5} \quad (4)$$

where, R_a is the extra-terrestrial solar radiation ($\text{mm H}_2\text{O m}^{-2} \text{ day}^{-1}$), T_{avg} is the average daily temperature ($^{\circ}\text{C}$), T_{max} is the maximum daily temperature ($^{\circ}\text{C}$), and T_{min} is the minimum daily temperature ($^{\circ}\text{C}$).

3.2. Statistical Analysis

Unless otherwise stated, all statistical analysis was performed in R Statistical Software (R Development Core Team, 2021). All parameters listed in Table 1 were then averaged to seasonal or annual scales for comparison to the annual MeHg or THg FWC and yields. The annual Hg FWCs and yields were first compared to all 118 variables using a Pearson correlation matrix with hierarchical clustering (Wei & Simko, 2017) to identify potential controls on either THg or MeHg. Variables with significant moderate correlations ($R^2 > 0.5$, $p > 0.05$) were identified and the initial 118 variables were further reduced by generally choosing annual values over temporally co-variate seasonal values. However, in some instances it was not immediately clear which co-variate should be removed and, in those cases, both were kept for further analysis (i.e., annual and summer average air temperature). This procedure resulted in a total of seven, eight, three, and six variables for annual MeHg FWC, MeHg yield, THg FWC, and THg yield, respectively.

Despite reducing the number of potential governing variables, the final variables were likely correlated to each other and necessitated further reduction before performing variable linear regression analysis. Hierarchical partitioning of adjusted R^2 with the *rdacca.hp* function in the *rdacca.hp* package (Lai et al., 2022) was used to assess the relative importance of these independent variables on annual Hg FWC and yield. The independent variables (potential hydroclimatic and chemical drivers) were scaled to the unit variance, and the proportion of variance that could be explained by a given independent variable was determined from 999 randomized data matrix using the *permu.hp* function in *rdacca.hp* (Lai et al., 2022). Hierarchical partitioning determines the proportion of independent and joint variances explained by each variable, allowing for the identification of variables with strong independent correlation with the dependent variable (Chevan & Sutherland, 1991; Lai et al., 2022; Mac Nally, 2000; Olea et al., 2010), in this case either annual Hg FWC or yield. As such, variables with high correlations but with little independent effect due to joint correlations with other variables would not be significant and were excluded from further analysis. Negative variances suggest that the variable is being suppressed by other variables (Olea et al., 2010) and were also excluded from further analysis. Given that there can be significant variance in annual mean data, we chose a relaxed significance threshold of $p < 0.1$ to guide which drivers to utilize for further analysis. As the relationship between Hg and the potential drivers may not follow a linear relationship, we completed the same analysis using the $\log_{10}$ transformed Hg data and included any drivers that met the above criteria. Only one additional driver was identified using the $\log_{10}$ transformed data (MeHg FWC vs. annual mean temperature, $p = 0.06$, %individual variance = 22) and was included in the final analysis. To our knowledge the use of hierarchical partitioning is rare in catchment hydrobiogeochemical studies but is more common in ecological studies with a larger number of correlated variables (Olea et al., 2010).

While hierarchical partitioning regression can account for correlation between independent variables, there can be variability in any given parameter around the annual mean. As such, significant variables ($p < 0.1$ and positive unique variance) were selected for bivariate Deming regression. Deming regression is a form of error in variable orthogonal regression that accounts for the standard deviation of the dependent and independent variables at each data point (Deming, 1943). Note that Deming regression cannot account for negative standard deviations, thus preventing the use of $\log_{10}$ transformed data. As such, all Deming regression was done on the non-transformed data. By using Deming regression rather than other linear or multiple regression techniques, we account for the

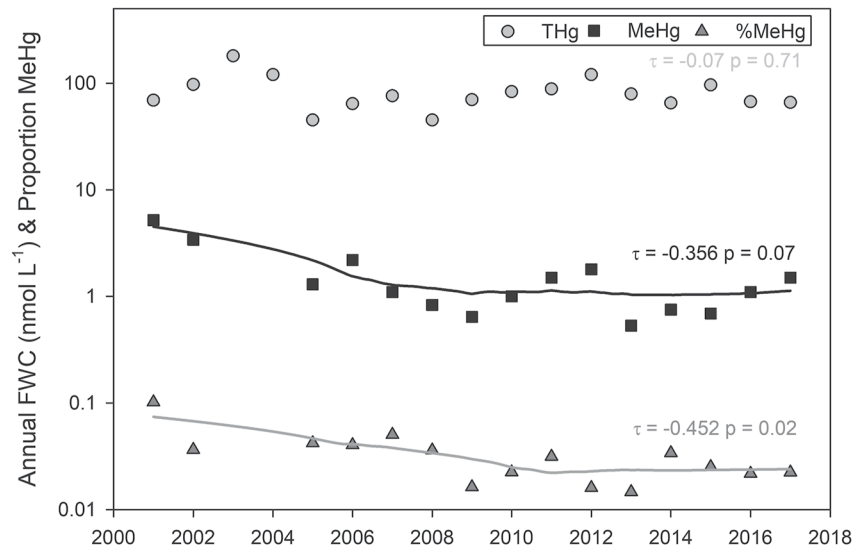


Figure 1. Annual THg and MeHg flow-weighted concentrations and %MeHg from 2001 through 2017 at the S2 catchment in the USDA Forest Service Marcell Experimental Forest. The solid lines are the loess smoothed lines illustrating the results of the Mann-Kendall tests to illustrate the average temporal trends. Note the gap in the MeHg record due to a resolved analytical contamination issue (McCarter, Sebestyen, et al., 2022).

natural variability in annual or seasonal measures, providing a clearer picture of the critical factors controlling Hg export under changing atmospheric conditions.

Deming regression minimizes the sum of squares (SS) residuals in both independent and dependent variable by (Wu & Yu, 2018; York, 1966),

$$SS = \sum_{i=1}^N [\omega(X_i)(x_i - X_i)^2 + \omega(Y_i)(y_i - Y_i)^2] \quad (5)$$

where, X_i is the measured driving variable (e.g., mean annual temperature), Y_i is the measured Hg FWC or yields, x_i is the regressed driving variable, and y_i is the regressed Hg FWC or yields. The individual Hg FWC or yields and driving variable were weighted based on annual variability in X_i and Y_i , respectively, following,

$$\omega(X_i) = \frac{1}{\sigma_{X_i}^2}, \omega(Y_i) = \frac{1}{\sigma_{Y_i}^2} \quad (6)$$

where, σ_{X_i} and σ_{Y_i} are the annual standard deviation of X_i and Y_i , respectively. The error of the independent variables was taken as the standard deviation around the mean of a given driver. All Demming regressions were calculated in SigmaPlot 15.0®.

4. Results and Discussion

There was considerable inter-annual variability in annual THg FWC but no trend through time ($\tau = -0.07$, $p = 0.71$, Figure 1, Table S1 in Supporting Information S1), contrasting the decrease in streamwater THg observed by Gerson and Driscoll (2016) in a temperate lake watershed in northeastern USA over a 10-year period ending in 2016. Here, without considering the intra-annual variability in THg FWC, spring runoff ratio was correlated with annual THg FWC, while annual THg yield correlated with streamflow and THg FWC (Tables S2 and S3, Figure S1 in Supporting Information S1). However, both streamflow and THg FWC are used in the calculation of yield (Sebestyen & Kyllander, 2017), biasing the Pearson correlation analysis. Nonetheless, Mitchell et al. (2008b) found that the snowmelt (12 days) accounted for between 26% and 39% of the annual THg yield from the S2 catchment and the nearby S6 catchment in 2005. When considering the entire spring (here defined as March–May) of 2005 in this study, ~77% of annual THg was exported during the season (Figure 2). Over the study period, the spring often had the largest THg yields ranging from 12% to 97% of the annual yield (Figure 2). This study supports previous work of Mitchell et al. (2008b) that the spring is an important period for Hg mobilization in hemi-boreal peatland-dominated catchments.

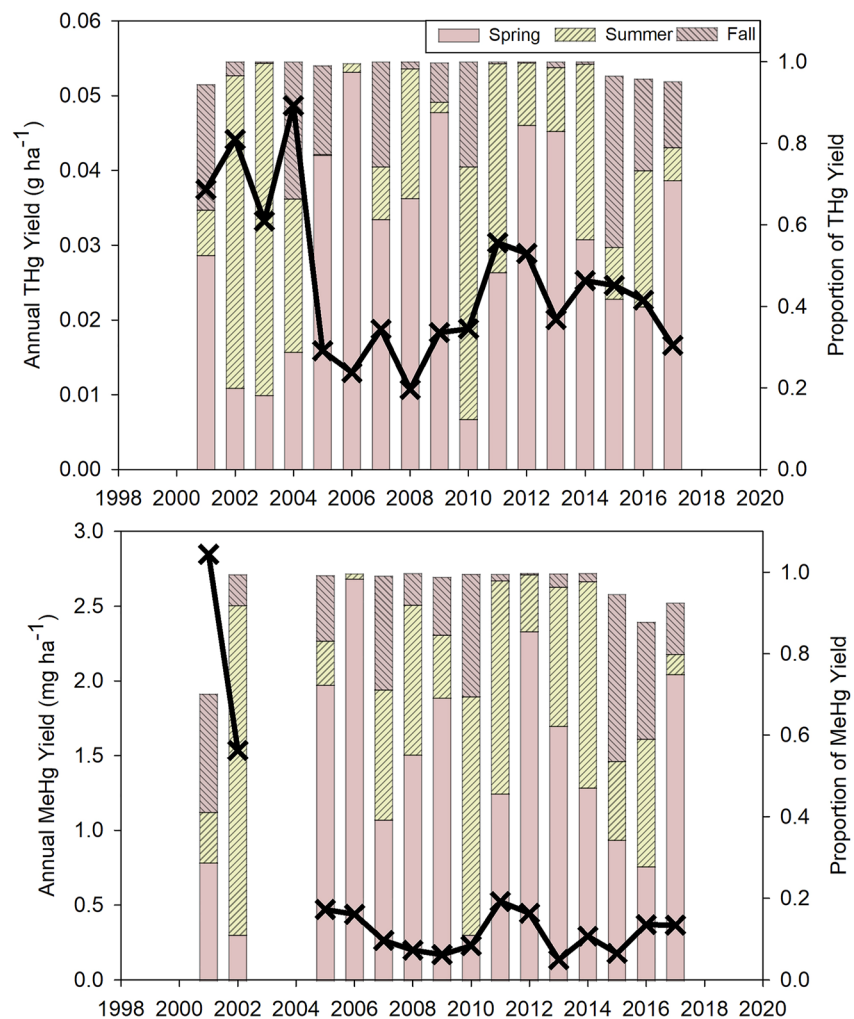


Figure 2. Annual THg (a) and MeHg (b) yields from 2001 through 2017 at the S2 catchment in the USDA Forest Service Marcell Experimental Forest.

Once we consider the intra-annual variability in spring runoff ratio and THg FWC using Deming regression, no significant correlation was observed between THg FWC and spring runoff ratio (Figure 3). None of our measured hydroclimatic variables strongly drive THg FWC and streamflow and THg FWC drive THg yields (Figure 3, Table S1 in Supporting Information S1). Despite no apparent effect of climate change on THg at annual time-scales (Figure 3), changes in other unmeasured or time-lagged mechanisms will likely induce shifts in THg (McCarter, Eggert, et al., 2022; McCarter, Sebestyen, et al., 2022), such as experimental evidence that warmer temperatures increase peatland inorganic Hg pore water concentrations from enhanced microbial activity (Sun et al., 2023). As such, there is an urgent need for more careful consideration and study of the interacting temperature-driven processes that influence THg mobilization to aquatic ecosystems.

During the study period, MeHg FWC varied independently of THg FWC, suggesting a disconnect between THg and MeHg transport and cycling processes (Figure 1). Methylmercury FWC decreased ($\tau = -0.356$, $p = 0.07$) over the 17-year streamwater Hg record from the highest annual MeHg FWCs in 2001 and 2002 to the lowest annual FWCs in 2009 and 2013 (Figure 1, Table S1 in Supporting Information S1). The proportion of THg as MeHg (%MeHg) significantly decreased ($\tau = -0.452$, $p = 0.02$) over the 17-year study, paralleling the decrease in MeHg FWC (Figure 1). The observed decrease in both MeHg FWC and yields (Figures 1 and 2) could not be ascribed to a single change in climate, streamwater chemistry, or atmospheric deposition metric (Figure 4). A combination of declines in annual wet Hg deposition concentration (31%) and the runoff ratio (21%) explained 52% of the independent variability in annual MeHg FWC (Table S4 in Supporting Information S1). When using

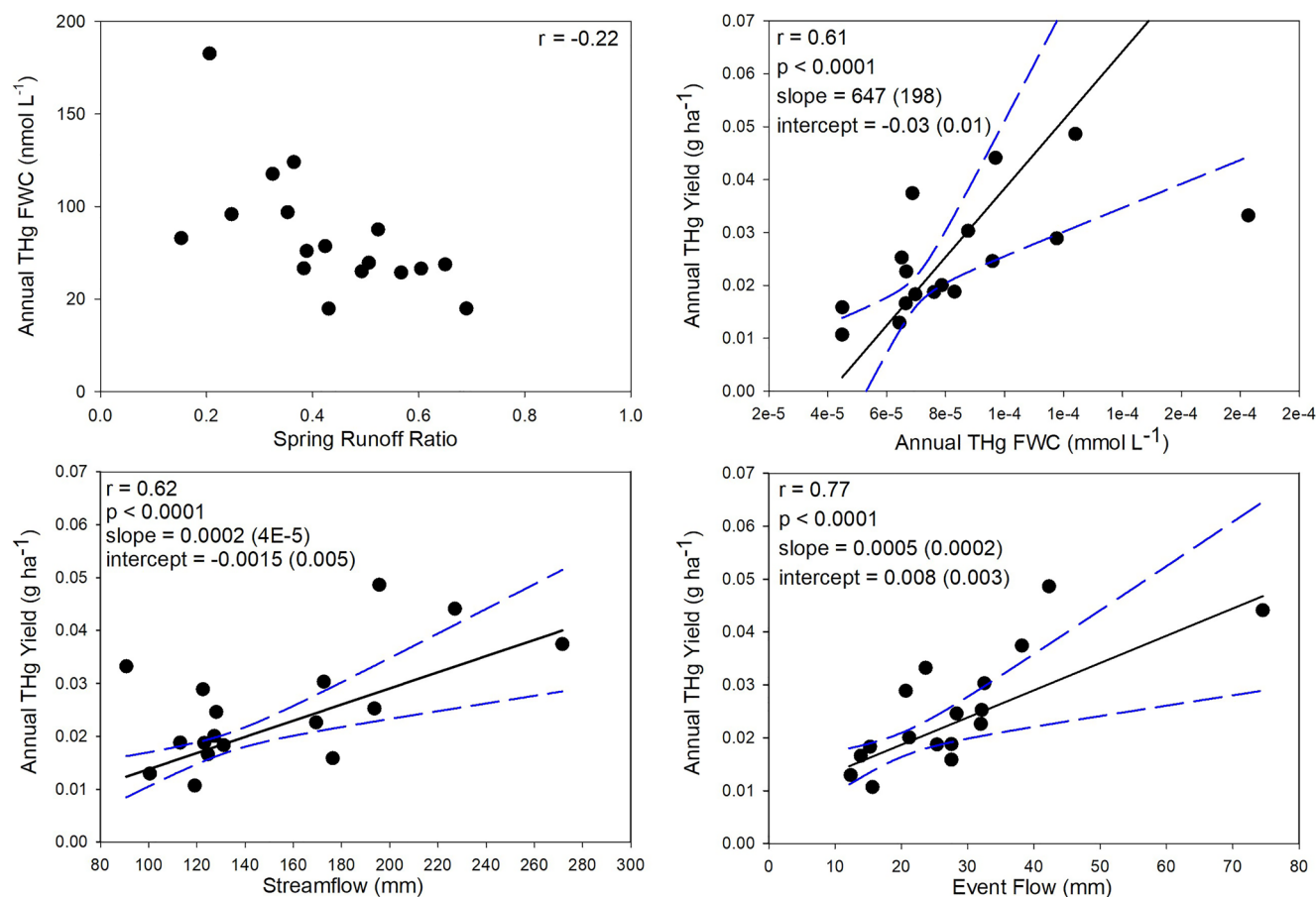


Figure 3. Error-in-variable linear regressions using Deming Regression (solid line) for THg flow-weighted concentrations and yield drivers. Brackets () indicate standard error of regression. Dashed lines represent the 95% confidence interval.

the $\log_{10}$ (MeHg FWC) hierarchical partitioning regression, runoff ratio (20%), wet Hg deposition concentration (24%), and annual mean air temperature (22%) increased the total explained variability to (64%), suggesting that these three drivers are critical for understanding annual MeHg FWC. In contrast, there were only two significant drivers for MeHg yield, MeHg FWC (34%) and annual streamflow (17%), which accounted for 51% of the independent variability but this value increases to 69% when including wet Hg deposition concentration (Table S5 in Supporting Information S1). Thus, changes in MeHg FWC, along with climate-controlled streamflow measures (e.g., declines in baseflow and runoff ratios), govern the total mass of MeHg leaving the catchment to downstream aquatic ecosystems. Interestingly, no streamwater chemistry parameters, including TOC and SO_4^{2-} , were correlated with MeHg FWC or yields. It is important to note that while in both cases, THg and MeHg, the yields were strongly dependent on the FWCs, both FWC and yields are estimated using stream flow. We recommend measuring both Hg concentration and streamflow to properly assess changes in Hg cycling and export. However, we recognize the increased cost associated with such a measurement scheme may preclude geographically extensive Hg measurements. Monitoring a larger number of streams for only Hg concentration with only a select few regionally representative streams for concentration and streamflow may be a more impactful management strategy to assess Hg dynamics at larger spatial scales.

4.1. Impacts of Cleaner Air on Hg Export

Sulfur dioxide emissions and subsequent SO_4^{2-} deposition have decreased following clean air legislation first introduced in the 1970s and amended through the 1990s (O'Meara, 1998; Pannatier et al., 2011; Sickles & Shadwick, 2015). The decline in SO_4^{2-} deposition primarily occurred from the 1970–1990s, decreasing from 28 kg ha⁻¹ yr⁻¹ in 1979 to 10 kg ha⁻¹ yr⁻¹ in 1999 at the MEF (National Atmospheric Deposition Program

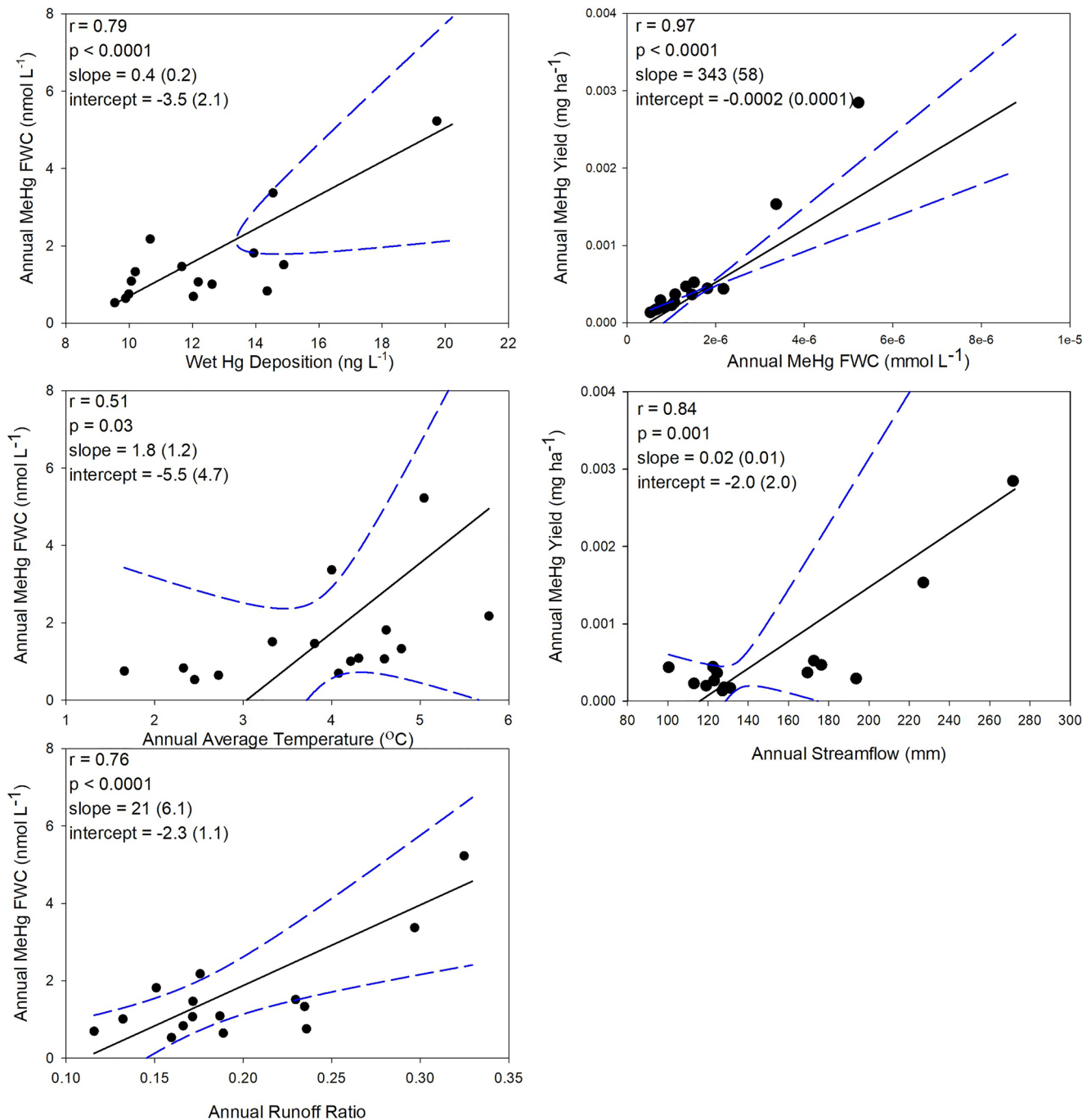


Figure 4. Error-in-variable linear regressions using Deming Regression (solid line) MeHg flow-weighted concentrations (FWC) and yield drivers. Note, annual mean air temperature was significantly related to $\log_{10}$ (MeHg FWC) and was, thus, included here but on the non-transformed MeHg FWC due to the limitations of Deming regression. Brackets () indicate standard error of regression. Dashed lines represent the 95% confidence interval.

(MN16, 2020). During the 2001–2017 streamwater Hg record, annual SO_4^{2-} deposition steadily declined from ~ 10 to $\sim 4 \mu\text{g SO}_4^{2-} \text{m}^{-2}$ (Figure 5), while both annual SO_4^{2-} FWC and yield (Figure S2 in Supporting Information S1) did not systematically respond to the changes in SO_4^{2-} deposition. We expected that atmospheric SO_4^{2-} deposition would be a strong predictor of annual MeHg FWC due to the partial dependence of Hg methylation on microbial SO_4^{2-} reduction in peatlands (Mitchell et al., 2008a; Pierce et al., 2022). However, there was no correlation between SO_4^{2-} deposition and annual MeHg FWC in the outflowing streamwater (Figures S1 and S3 in Supporting Information S1). Given the large store of sulfur in the surficial peat to buffer these lower SO_4^{2-}

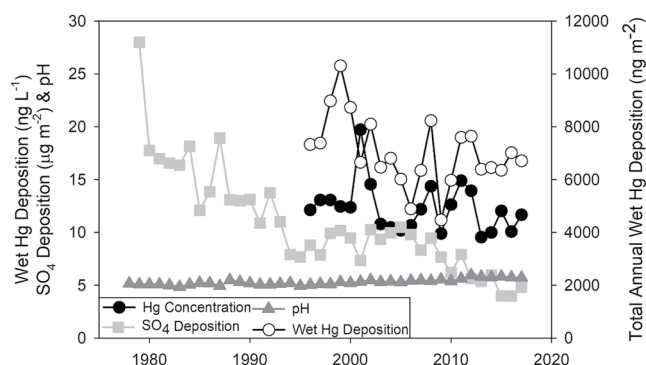


Figure 5. Annual total and concentration of wet Hg deposition, and the annual average precipitation pH and SO_4^{2-} concentration from 1980 through 2017 (Hg: 1995–2017) at the USDA Forest Service Marcell Experimental Forest.

litterfall Hg deposition between 2008 and 2020 was on average $3,700 \pm 500 \text{ ng Hg m}^{-2}$ (Andonie et al., 2021; Mercury Litterfall Network, 2022; Risch et al., 2012), lower than the 1995 litterfall Hg deposition estimate of $12,000 \text{ ng Hg m}^{-2}$ (Grigal et al., 2000). Litterfall Hg concentration has remained relatively constant between the different measurement periods ($\sim 25\text{--}30 \text{ ng Hg g}^{-1}$) but there is a substantial difference in litterfall mass (1995: $\sim 400 \text{ g m}^{-2}$ vs. 2008–2020: $137 \pm 16 \text{ g m}^{-2}$) (Andonie et al., 2021; Grigal et al., 2000; Mercury Litterfall Network, 2022; Risch et al., 2012). Thus, the difference between the two estimates is likely due to methodological differences in the placement of the litterfall collectors or the canopy density. One possibility for the differences in mass deposition maybe the result of the placement of NADP litterfall collectors near the NADP opening, which may lead to lower areal mass litter deposition than a closed canopy forest as measured in Grigal et al. (2000). Despite litterfall often being an important Hg input at the MEF (Grigal et al., 2000; Woerndle et al., 2018), its mobility and availability is often limited relative to wet Hg deposition as additional decomposition or surface erosion processes are often required before impacting streamwater MeHg concentrations or yields (Bishop et al., 2020; Demers et al., 2007). Wet Hg deposition concentrations varied between 9.5 and 14.6 ng L^{-1} over the study period (Figure 5) and was positively correlated ($p < 0.0001$, $r = 0.79$) with the annual MeHg FWC (Figure 4). Given the annual timescales investigated here, recent wet Hg deposition would likely be more bioavailable to the resident methylating microbiota than both legacy Hg that has been bound to organic matter (Chiasson-Gould et al., 2014; Gorski et al., 2008; Hintelmann et al., 2002; Hsu-Kim et al., 2013) and litterfall Hg, where availability to methylating bacteria may lag beyond 1 year as additional decomposition processes are often required to liberate Hg bound in litter (Demers et al., 2007; Pokharel & Obrist, 2011). Thus, potential differences in Hg bioavailability to methylating bacteria due to both inorganic Hg speciation and dissolved organic matter composition (Huang & Mitchell, 2023; Liem-Nguyen et al., 2021) plays a critical role in modulating annual streamwater MeHg FWC in this peatland dominated catchment. Since the majority of streamwater THg is likely from Hg associated with litterfall or gaseous element Hg (Woerndle et al., 2018) and MeHg was strongly associated with MeHg (Figure 4), we suggest that inorganic Hg and MeHg are from different Hg pools on the landscape.

The decline in the wet Hg deposition explained the greatest amount of variability in the annual MeHg FWC (Table S4 in Supporting Information S1), suggesting that further reduction in global atmospheric Hg emissions and subsequent wet deposition would have a substantial and rapid impact in reducing MeHg in streams draining northern peatland catchments. Given the current wet deposition ($6,000\text{--}7,000 \text{ ng Hg m}^{-2}$) is approaching pre-industrial total Hg deposition levels ($\sim 4,000 \text{ ng Hg m}^{-2}$) at the MEF and that $\sim 20\%$ of all Hg deposition is wet Hg deposition, future decreases in wet Hg deposition concentrations may not have as much of a long-term impact and be less important than other drivers in this region. As such, further research on linking atmospheric Hg deposition to precipitation Hg concentration will greatly clarify the likely trajectory of streamwater MeHg over longer timescales.

4.2. Impacts of Climate Change on Hg Export

Climate change is readily apparent at the MEF (Figure 6). From 1962 to 2017, annual mean air temperature ($\tau = 0.27$, $p = 0.003$) increased by $\sim 1.8^\circ\text{C}$, with no trend in precipitation amount ($\tau = 0.01$, $p = 0.94$, Figure 6).

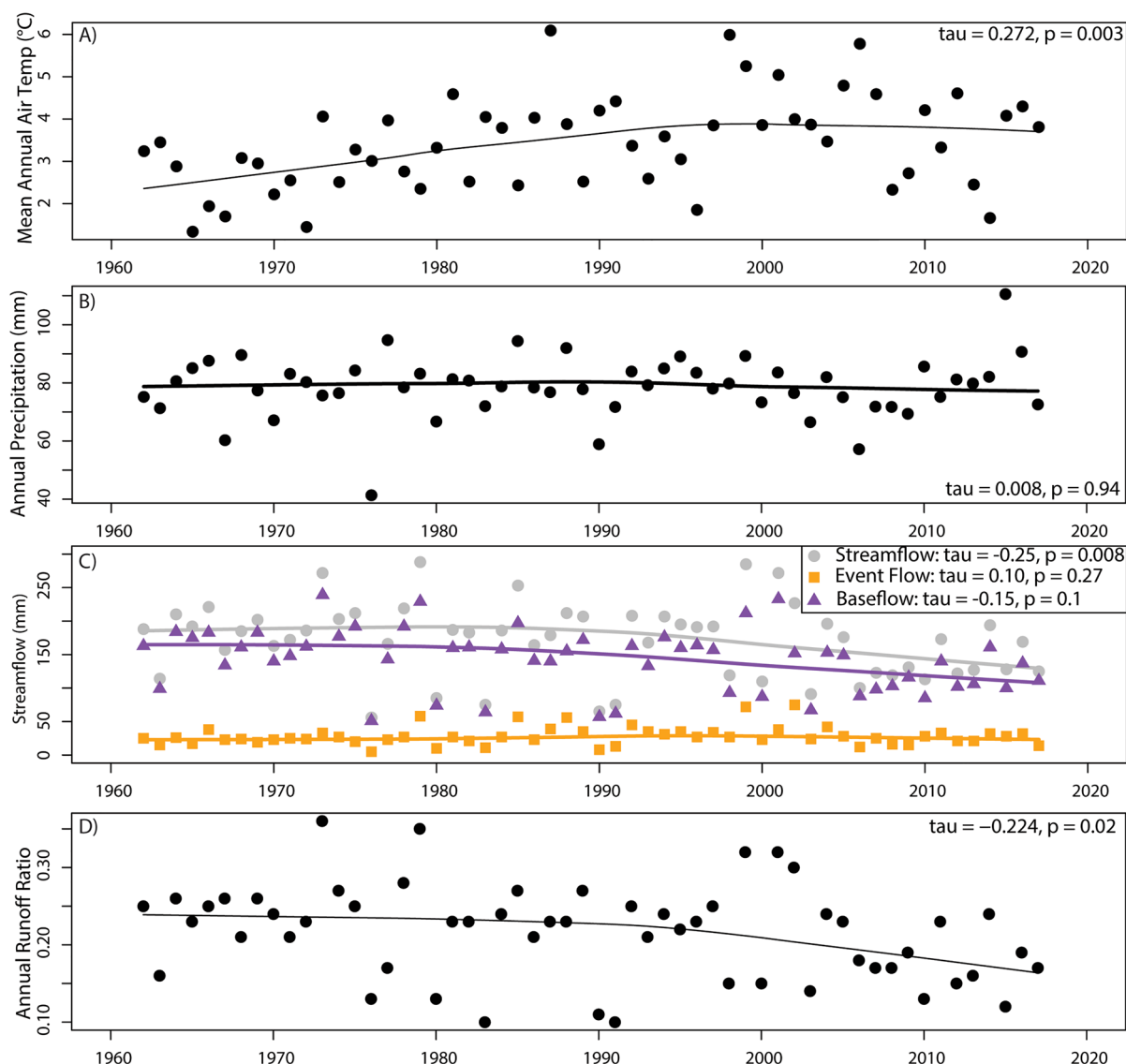


Figure 6. The change in annual average air temperature (a), total precipitation (b), streamflow (c), and runoff ratio (d) from 1962 through 2017 at the USDA Forest Service Marcell Experimental Forest. Solid lines are the loess smoothed lines resulting from the Mann-Kendall trend tests.

Mean annual air temperature was positively correlated with annual MeHg FWC ($1.8 \pm 1.2 \text{ nmol L}^{-1}\text{C}^{-1}$; Figure 4). However, over the Hg sampling period, there was no increase in annual mean air temperature ($\tau = -0.26$, $p = 0.12$), unlike the clear increase from 1962 to 2017 ($\tau = 0.27$, $p = 0.003$, Figure 6). In general, Hg methylation is expected to increase with higher microbial activity at warmer air temperatures (Hsu-Kim et al., 2013; Sun et al., 2023; Yang et al., 2016). Such an increase in microbial activity is thought to be more important in the shoulder seasons (spring and fall) when the average air temperatures are further from optimal microbial activity temperatures (IPCC, 2021). However, neither shoulder season mean, minimum, nor maximum air temperatures were strong predictors of annual MeHg FWC. Here, warmer mean summertime air temperatures (Figure S4 in Supporting Information S1) were positively correlated with annual MeHg FWC (Figure 4, Table S2 in Supporting Information S1) suggesting that warmer summers combined with in situ increases in SO_4^{2-} , and associated changes in microbial Hg methylation, coupled with changes in water table (Sun et al., 2023) will likely produce higher MeHg FWC in receiving streamwater in the near-term. Importantly, these results suggest that annual MeHg FWC responds to mean summer air temperature, which strongly influences mean annual air temperature, and that future climate warming may have an immediate impact on MeHg concentrations in peatland-fed streamwater.

In peatlands, other microbially mediated processes, such as heterotrophic respiration or methanogenesis, are sensitive to the timing and magnitude of temperature and water table anomalies but may be temporally limited and masked by using annual means (Feng et al., 2020; Helbig et al., 2022; Le Geay et al., 2023). In the S2 peatland, the seasonal peatland water tables have been disproportionately decreasing due to climate change during the drier summer ($-0.0021 \pm 0.0008 \text{ m yr}^{-1}$, $p < 0.0001$) and fall ($-0.0020 \pm 0.0014 \text{ m yr}^{-1}$, $p = 0.005$) relative to the wetter spring ($-0.0015 \pm 0.0009 \text{ m yr}^{-1}$, $p = 0.002$). Here, annual MeHg FWC was positively correlated ($p < 0.05$) with minimum temperature variability (defined as one standard deviation of the mean minimum temperature; Figure S1 in Supporting Information S1). This increase in temperature variability would likely decrease the amount of time that temperature limitations are impacting Hg methylation rates, while lower water tables increase the oxidative production of SO_4^{2-} (Sun et al., 2023). These two processes are likely to increase future MeHg concentrations in peatlands (Coleman Wasik et al., 2015; Sun et al., 2023). However, higher soil temperatures can increase gaseous sulfur losses from peatlands that can suppress pore water MeHg concentrations and partially offset the aforementioned increases in MeHg concentrations (Åkerblom et al., 2013).

The feedbacks between climate change, shifts in vegetation and microbiological communities, and Hg methylation are often non-linear in peatlands. For instance, the results of the Spruce and Peatland Responses Under Changing Environments (SPRUCE) experiment at MEF suggest that heterotrophic respiration is likely to increase from peatlands under a warming climate (Wilson et al., 2021b) due to enhanced microbial consumption of deeper peat (Wilson et al., 2016, 2021a). Increases in microbial peat consumption would likely increase pore water Hg concentrations similar to the observations of Sun et al. (2023), which is the net result of many interacting processes over a 3-year boreal peatland warming experiment. However, the concurrent increase in vascular plants and fine root production (Malhotra et al., 2020; McPartland et al., 2019) would increase transpiration and modify soil moisture and nutrient conditions through multiple non-linear feedbacks (McCarter et al., 2020; Waddington et al., 2015), while also likely changing Hg deposition inputs via shifts in peatland vegetation (Sun et al., 2023) and associated changes in litterfall rates. Our results suggest that in isolation, further increases in air temperature and variability due to climate change, MeHg concentrations in peatland-fed streams, rivers, and lakes will increase due to this positive feedback but we strongly recommend further interdisciplinary research on the numerous non-linear climate warming feedbacks operating in peatland dominated landscapes and their effect on Hg cycling and mobility.

In general, higher air temperatures increase evapotranspiration losses (Dymond et al., 2014; Helbig et al., 2020), lowering available soil water in the uplands and decreasing peatland water tables, thus converting less precipitation to stream runoff. In the S2 catchment, higher evapotranspiration combined with steady precipitation drove a significant decline in baseflow ($\tau = -0.25$, $p = 0.008$), streamflow ($\tau = -0.15$, $p = 0.1$), the amount of precipitation that became streamflow (runoff ratio, $\tau = -0.224$, $p = 0.02$), and an increase in the proportion of event flow ($\tau = 0.321$, $p < 0.001$), particularly since 2000 (Figure 6 and Figure S5 in Supporting Information S1). Seasonally, the decline in annual runoff ratio was driven by a significant decrease in fall ($\tau = -0.203$, $p = 0.03$), while both the spring ($\tau = -0.134$, $p = 0.15$) and summer ($\tau = -0.102$, $p = 0.28$) did not change during the same period (Figure S4 in Supporting Information S1). Such declines in runoff ratio due to decreasing baseflow and increases in air temperature that drove higher evapotranspiration is consistent with observations at the MEF (Dymond et al., 2014) and projections of climate change (Reshmidevi et al., 2018; Zhang et al., 2023). Despite potential increases in hydrological connectivity between the stream and the catchment during wet periods such as the spring (Jones et al., 2023; Woerndle et al., 2018), the decline in annual runoff ratio was driven by lower water tables during dry periods that offsets any increases in hydrological connectivity during wet periods at annual time scales. Thus, driving an overall decrease in the hydrological connectivity of uplands and the peatland to the stream at annual time scales and precipitation that falls on the catchment over a year is less likely to be transported to the stream during more frequent drier periods (summer and fall).

The flow and pathways that water moves through a catchment is key to regulating downstream aquatic Hg concentrations and cycling (Branfireun et al., 2020) but the mechanisms are often confounded by other landscape features such as ecosystem types or disturbances (Lam et al., 2022). At the MEF, there has been a clear climate change induced decrease in runoff ratio that decreased annual catchment hydrological connectivity but no statistical change in annual event flow. In the S2 catchment, THg is mobilized during high flow events from the upland-peatland interface (lagg), where the lagg Hg was mostly sourced from vegetation litterfall as shown using natural abundance stable Hg isotopes (Woerndle et al., 2018). During low-flow conditions when the upland and peatland largely disconnect from catchment outflow, Hg accumulates as litterfall and other depositional processes

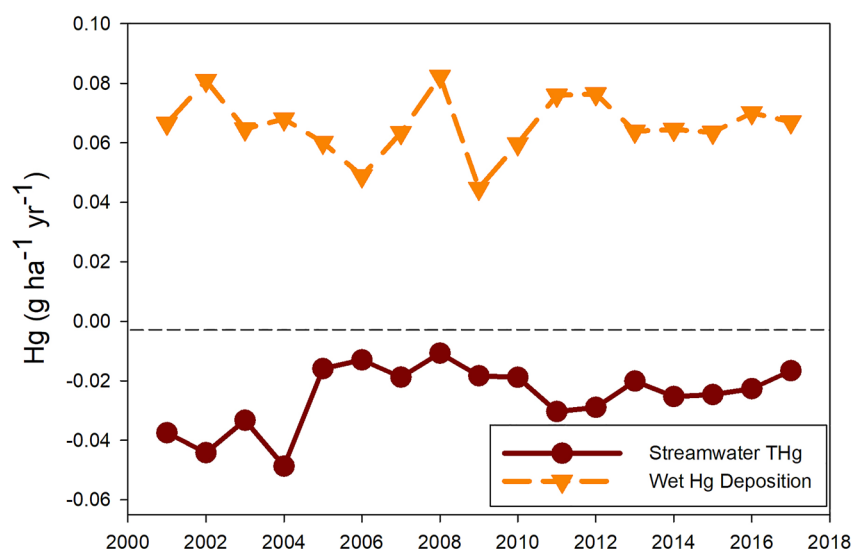


Figure 7. Annual streamwater THg export and wet Hg deposition for the S2 catchment.

and preferential subsurface flow during smaller precipitation events (McCarter et al., 2021; Woerndle et al., 2018). During larger flow events, the accumulated THg is then flushed from the catchment as evidenced by the positive correlation between THg yield, streamflow and event flow. The impact of future climate warming on THg export will likely depend on shifts in surface and near surface flow during high flow events. Further departures from historical runoff patterns will likely decrease our ability to predict THg mobilization from peatland-dominated catchments but a greater research focus on landscape interfaces (in this case upland-lagg-peatland) may provide greater clarity to future THg mobilization.

Climate warming has also reduced the annual runoff ratio (Figure 6), significantly decreasing annual MeHg FWC and yields (Figure 4). With a 10% decline in annual runoff ratio, which is the approximate average decline over the hydroclimatic record, annual MeHg FWC declined by $2.1 \pm 0.6 \text{ nmol L}^{-1}$ and yields by $0.001 \pm 0.0003 \text{ mg ha}^{-1}$. While no seasonal change in hydrology (upland or peatland) correlated with annual MeHg FWC. Peatland water table (i.e., catchment water storage) plays a critical role in facilitating water and solute transport, where higher water tables increase solute transport rates (McCarter & Price, 2017). With the observed decline in peatland water tables due to climate change, it takes longer and more precipitation for the catchment to hydrologically reconnect and transport any MeHg produced in the catchment to the stream and increases likelihood of chemical or microbial demethylation (Barkay & Gu, 2022). At the S2 catchment, photodemethylation would likely be limited due to the stream often being dry for extended durations during summer rather than stagnant surface water, which is also limited in the peatland (Sebestyen et al., 2021). While the increased mean annual air temperature and increased minimum temperature anomalies may increase in situ peatland MeHg production, the produced MeHg would be less likely to reach the catchment outlet due to decreased runoff from the peatland to the stream and provides a critical negative feedback limiting MeHg in our aquatic ecosystems under a warming climate.

4.3. Future Hg Export Dynamics

Despite wet atmospheric Hg deposition declining over the study period, the streamwater Hg export from the catchment was consistently lower than wet Hg deposition (Figure 7). The net difference between wet Hg deposition and streamwater Hg export was consistent throughout the study period, where the catchment retained on average $0.04 \pm 0.01 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$. This Hg retention resulted in between 13% and 71% (avg. $38 \pm 13\%$) of the wet Hg deposition to be exported via the stream in any given year and encompasses the Hg retention estimates of Kolka et al. (2001) from a more detailed THg budget study in 1995. However, wet Hg deposition is not the only Hg input into catchments and often is a minor flux relative to other depositional pathways, such as dry deposition or litterfall (Grigal et al., 2000; Risch et al., 2012; Woerndle et al., 2018). In 1995, Grigal et al. (2000) observed throughfall ($0.13 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$) and litter inputs including ($0.12 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$) to be nearly double that of wet Hg deposition ($0.07 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$) when accounting for the differences between upland and peatland

vegetation in the S2 catchment. While by 2007–2020, litterfall Hg deposition was ~60% of wet Hg deposition at MEF (Andonie et al., 2021; Mercury Litterfall Network, 2022; Risch et al., 2012), although lower litterfall mass deposition related to the near proximity of the NADP site opening could account for this decrease (see earlier discussion). Mazur et al. (2014) observed direct gaseous Hg deposition on the upland forest floor $0.49 \pm 0.18 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$ of the nearby S7 catchment assuming a snow-free period of March through November. A nearby peatland net total gaseous Hg flux was on average $-0.09 \pm 0.02 \text{ g Hg ha}^{-1} \text{ yr}^{-1}$ from the mesocosms at the SPRUCE chambers located in the MEF S1 catchment assuming the same snow-free period (Haynes et al., 2017). Woerndle et al. (2018) highlighted the importance of these other fluxes to streamwater THg, estimating between 73% and 95% of streamwater THg was from dry deposition of gaseous elemental Hg based on stable Hg isotopes. Further, in a Swedish boreal peatland, Li et al. (2023) observed that ~30% of all Hg deposited is emitted back to the atmosphere based on a Hg isotope mass balance. While these other Hg input pathways were not measured as part of this study, it is likely that total terrestrial Hg stored in the S2 catchment is increasing since the aqueous export of THg is relatively small compared to the other various Hg fluxes and the associated uncertainties with those estimates. As such, even with further reductions in atmospheric Hg deposition, these catchments will likely remain Hg sinks on the landscape. Importantly, even under pre-industrial Hg atmospheric concentrations and deposition, these catchments would likely be weak net sinks for atmospheric Hg and, as such, we may never see a net flushing of anthropogenic Hg from hemi-boreal or boreal peatland-dominated catchments.

Droughts and subsequent hydrological recovery are commonly linked to increased concentrations of metals, such as Hg or lead, from peatlands (Jeremiasen et al., 2018; Szkokan-Emilson et al., 2013) and MeHg concentrations can increase within peatlands (Coleman Wasik et al., 2015). There were several droughts (1967/1968, 1976/1977, 1990/1991, and 2006/2007) with annual precipitation <603 mm (<77% of the 1962–2017 mean) during the long-term record. Annual runoff ratios during drought years (0.17 ± 0.05) were significantly lower ($p = 0.04$, $t = 2.23$, $df = 10$) than the long-term mean (1962–2017, 0.22 ± 0.02), indicating that on average the entire catchment (both uplands and peatlands) was less hydrologically connected to the stream during droughts. During the 2006/07 drought, Coleman Wasik et al. (2015) observed SO_4^{2-} regeneration and subsequent elevated pore water MeHg concentrations of a nearby peatland and an increase in THg pore water concentrations after the drought. Despite increases in pore water MeHg concentrations, there was no detection of elevated annual MeHg FWC in the stream following the 2006/07 drought. While elevated THg FWC and yields may occur after droughts, potentially covarying with runoff ratios (Figure 3), elevated yields were also not observed during the Hg record and the response was relatively limited at the annual scale. Regardless of internal Hg cycling during and after droughts, the export of Hg from peatland catchments was more dependent on the efficiency that precipitation is converted to streamflow, which declines during drought years, limiting both MeHg and THg yield under more frequent climate change induced droughts (IPCC, 2021).

Under an increasingly warming climate, MeHg production within peatlands will likely increase the pore water MeHg concentrations (Sun et al., 2023; Yang et al., 2016). However, with higher temperatures and water losses to the atmosphere, annual streamflow decreases. A resulting reduction in annual hydrological connectivity between MeHg sources and biological sinks will likely be critical in modulating aquatic MeHg concentrations under future climates. Additionally, as the seemingly microbially available atmospheric wet Hg deposition continues to decline and the Hg in a catchment becomes increasingly sourced from less bioavailable legacy Hg sources, such as those bound to thiol-containing organic matter (Liem-Nguyen et al., 2021), we may see a further reduction in streamwater MeHg due to lower Hg bioavailability. Our results using this unprecedented data set suggest that MeHg export from headwater peatland catchments will continue to decrease over time if climate change continues to accelerate the reduction of runoff ratios and atmospheric wet Hg deposition further decreases.

5. Conclusions

Climate change and cleaner air are unequivocally altering the ecohydrological and biogeochemical processes that underpin Hg cycling and export in peatland-rich catchments. We present a clear linkage between decreasing wet Hg deposition concentration due to reductions in atmospheric Hg over North America (cleaner air) that when coupled with the climate change induced reductions in runoff ratios more than offsets the increase in MeHg production due to warmer air temperatures. Yet, the response of THg to cleaner air and climate change was much less clear. As such, we highlight the clear need for further research into unraveling how future changes in timing and strength of hydrological connectivity within and from catchments will impact Hg export, determine

the ubiquity and strength of these relationships across a greater number of catchments, and elucidate how these changes will feedback on Hg cycling in wetland and aquatic ecosystems. These critical insights would not be possible without the long-term consistent monitoring of the comprehensive array of the environmental measurements at the MEF. To better adapt to future climate and environmental change, there is a need for more and longer integrated multidisciplinary data sets.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The streamwater chemistry data used in the study are freely available at the Environmental Data Initiative via <https://doi.org/10.6073/pasta/a47f5019f2ce2aff6cbca0e555939950> with CC BY 4.0 (Sebestyen, Lany, et al., 2022). The daily precipitation data used in the study are freely available at the Environmental Data Initiative via <https://doi.org/10.6073/pasta/bfcb6628794b8ba1fb1159b9b75e3f58> with CC BY 4.0 (Sebestyen et al., 2021, 2022a). The daily air temperature data used in the study are freely available at the Environmental Data Initiative via <https://doi.org/10.6073/pasta/c93d5e0bba0b40f68f10244fcb68d25b> with CC BY 4.0 (Sebestyen et al., 2021, 2022c). The daily groundwater and peatland water table data used in the study are freely available at the Environmental Data Initiative via <https://doi.org/10.6073/pasta/6e9348a1c691c10271c9373bd31da67f> with CC BY 4.0 (Sebestyen et al., 2021). The daily streamflow data used in the study are freely available at the Research Data Archive via <https://doi.org/10.2737/RDS-2018-0009> (Sebestyen et al., 2021; Verry et al., 2018). The biweekly atmospheric deposition chemistry data are freely available at the National Trends Network (site MN16) via <https://nadp.slh.wisc.edu/sites/ntn-MN16/> (NADP (MN16), 2020). The monthly atmospheric mercury deposition chemistry data are freely available at the Mercury Deposition Network (site MN16) via <https://nadp.slh.wisc.edu/sites/mdn-MN16/> (NADP (MN16), 2021).

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