

Climate change and cleaner emissions decrease methylmercury export from a headwater peatland catchment

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Introduction

In this supplement, we provide supplemental figures and tables that enhance the main manuscript. This document includes two sections: Supplementary Figures and Supplementary Tables. The Supplementary Figures section contains the hierarchical clustered Pearson correlation matrix, changes in seasonal runoff ratios, and changes in annual proportion event flow. The Supplementary Tables sections contains the mean (\pm one standard deviation) annual flow-weighted concentrations (FWC) for total mercury (THg) and methylmercury (MeHg), mean (\pm one standard deviation) annual temperature, annual and spring runoff ratios, and mean (\pm one standard deviation) wet mercury (Hg) deposition concentration, as well as the independent and joint variability for all hierarchical partitioning regressions. Note, there is a two year gap in the MeHg concentration data due to a now resolved laboratory contamination issue. For more detail of this issue see McCarter et al. (2022) and Sebestyen et al. (2022).

Supplementary Figures



Figure S1. The hierarchical clustered Pearson correlation matrix. Larger circles indicate smaller p-values, while no circle indicate $p > 0.05$. Note, a higher resolution of Figure S1 is provided as an additional supplementary file.

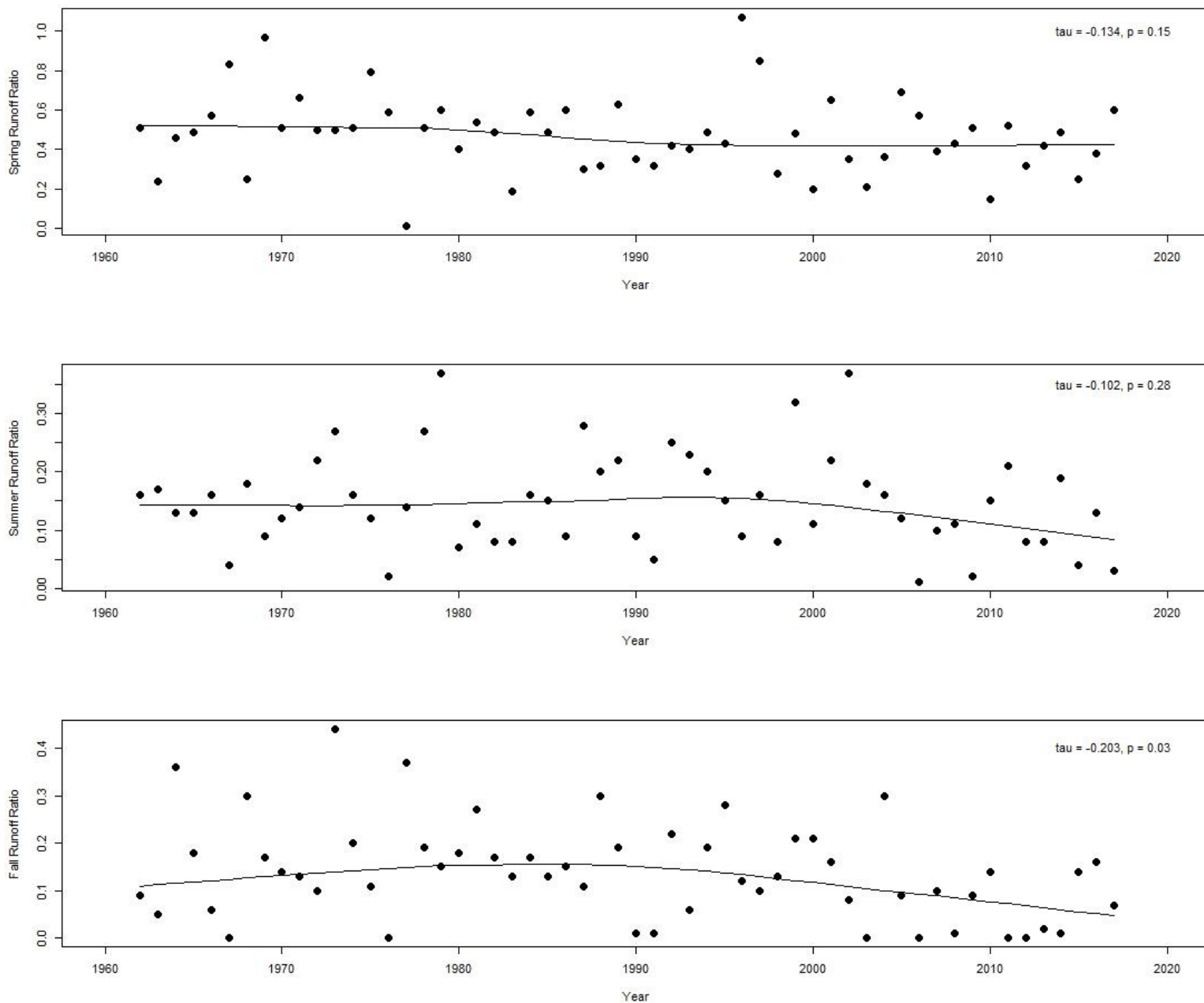


Figure S2. Changes in seasonal runoff ratios 1962 through 2017 at the USDA Forest Service Marcell Experimental Forest. Solid lines are the LOESS smoothed lines resulting from the Mann-Kendall trend tests.

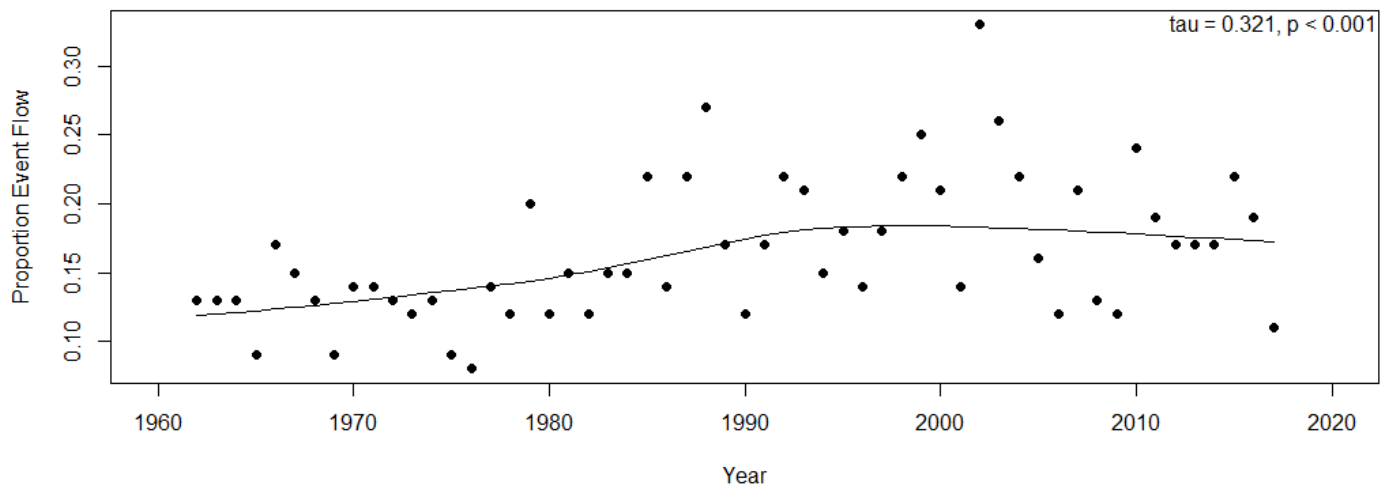


Figure S3. The change in annual event flow from 1962 through 2017 at the USDA Forest Service Marcell Experimental Forest. Solid lines are the LOESS smoothed lines resulting from the Mann-Kendall trend tests.

Supplemental Tables

Table S1. The FWC and yields (\pm one standard deviation) of THg and MeHg, as well as mean (\pm one standard deviation) of mean annual air temperature, annual runoff ratio, mean annual wet Hg deposition concentration, and spring runoff ratio.

Year	THg FWC	THg Yield	MeHg FWC	MeHg Yield	Mean Annual Air Temperature	Annual Runoff Ratio	Mean Wet Hg Deposition Concentration	Spring Runoff Ratio
	nmol L ⁻¹	g ha ⁻¹	nmol L ⁻¹	mg ha ⁻¹	°C	-	ng L ⁻¹	-
2001	68.8 \pm 16.4	0.04 \pm 0.0017	5.2 \pm 15.6	2.8 \pm 0.078	5 \pm 12.1	0.32 \pm 0.29	19.7 \pm 9.3	0.65 \pm 0.45
2002	96.9 \pm 24	0.04 \pm 0.0023	3.4 \pm 5.4	1.5 \pm 0.001	4 \pm 12.3	0.3 \pm 0.28	14.5 \pm 13.3	0.35 \pm 0.45
2003	182.7 \pm 35.4	0.03 \pm 0.0022	-	-	3.9 \pm 12.3	0.14 \pm 0.01	10.8 \pm 13.3	0.21 \pm 0.13
2004	124 \pm 22.1	0.05 \pm 0.00199	-	-	3.5 \pm 13.4	0.24 \pm 0.29	10.5 \pm 8.8	0.36 \pm 0.78
2005	44.9 \pm 9.5	0.02 \pm 0.0008	1.3 \pm 1.7	0.5 \pm 0.001	4.8 \pm 12.9	0.23 \pm 1	10.2 \pm 6.8	0.69 \pm 2.01
2006	64.3 \pm 5.6	0.01 \pm 0.0004	2.2 \pm 7.7	0.4 \pm 0.002	5.8 \pm 11.5	0.18 \pm 0.59	10.7 \pm 8.5	0.57 \pm 1.09
2007	76 \pm 6.6	0.02 \pm 0.0005	1.1 \pm 9	0.3 \pm 0.002	4.6 \pm 13.4	0.17 \pm 0.25	12.2 \pm 11.7	0.39 \pm 0.41
2008	44.8 \pm 6.1	0.01 \pm 0.0004	0.8 \pm 3.8	0.2 \pm 0.0002	2.3 \pm 13.4	0.17 \pm 0.19	14.4 \pm 24	0.43 \pm 0.3
2009	69.7 \pm 2.1	0.02 \pm 0.0002	0.6 \pm 1.5	0.2 \pm 0.00001	2.7 \pm 12.8	0.19 \pm 0.46	9.9 \pm 15.3	0.51 \pm 0.76
2010	83 \pm 5.3	0.02 \pm 0.0004	1 \pm 5.3	0.2 \pm 0.0001	4.2 \pm 12.5	0.13 \pm 0.08	12.6 \pm 17.4	0.15 \pm 0.07
2011	87.6 \pm 6.7	0.03 \pm 0.0006	1.5 \pm 3.3	0.5 \pm 0.0001	3.3 \pm 12.7	0.23 \pm 0.2	14.9 \pm 14.8	0.52 \pm 0.31
2012	117.7 \pm 62.7	0.03 \pm 0.0045	1.8 \pm 0.2	0.4 \pm 0.00005	4.6 \pm 12	0.15 \pm 0.15	13.9 \pm 14.9	0.32 \pm 0.12
2013	78.7 \pm 4.8	0.02 \pm 0.0003	0.5 \pm 2	0.1 \pm 0.0004	2.5 \pm 13.9	0.16 \pm 0.32	9.5 \pm 8.6	0.42 \pm 0.6
2014	65.1 \pm 10.9	0.03 \pm 0.00097	0.8 \pm 2.7	0.3 \pm 0.0002	1.7 \pm 14.1	0.24 \pm 0.24	10 \pm 8.1	0.49 \pm 0.37
2015	95.9 \pm 15.8	0.02 \pm 0.0012	0.7 \pm 3.1	0.2 \pm 0.0002	4.1 \pm 12.8	0.12 \pm 0.12	12 \pm 12.4	0.25 \pm 0.17
2016	66.6 \pm 4.2	0.02 \pm 0.0004	1.1 \pm 3.3	0.4 \pm 0.0004	4.3 \pm 12.3	0.19 \pm 0.17	10.1 \pm 9.5	0.38 \pm 0.26
2017	66.5 \pm 12.2	0.02 \pm 0.0009	1.5 \pm 1.3	0.4 \pm 0.00005	3.8 \pm 12.6	0.17 \pm 0.28	11.7 \pm 11.3	0.6 \pm 0.34

Table S2. The independent and joint variability explained by each driver of THg FWC from the hierarchal partitioning regression and Z scores. **Bold** and *italic* indicate significant at $p < 0.05$.

Driver	Independent Variability	Joint Variability	Total Variability	% Independent Variability	Z score
Annual Precipitation pH	-0.005	0.003	-0.002	3	-0.580
Spring Runoff Ratio	-0.134	-0.017	-0.151	89	4.370
Summer Precipitation pH	-0.012	-0.004	-0.016	8	-0.350

Table S3. The independent and joint variability explained by each driver of THg yields from the hierarchal partitioning regression and Z scores. **Bold** and *italic* indicate significant at $p < 0.05$.

Driver	Independent Variability	Joint Variability	Total Variability	% Independent Variability	Z score
Annual Streamflow	-0.02	0.015	-0.01	4	-0.05
Annual Event Flow	-0.02	-0.006	-0.02	3	-0.23
Summer Streamflow	-0.01	-0.010	-0.02	2	-0.34
Summer Proportion Event Flow	-0.02	-0.029	-0.04	3	-0.25
Summer Runoff Ratio	-0.02	-0.002	-0.02	3	-0.19
THg FWC	-0.42	-0.044	-0.46	84	14.12

Table S4. The independent and joint variability explained by each driver of MeHg FWC from the hierarchal partitioning regression and Z scores. **Bold** and *italic* indicate significant at $p < 0.05$.

Driver	Independent Variability	Joint Variability	Total Variability	% Independent Variability	Z score
Baseflow	-0.07	-0.11	-0.18	9.6	0.54
Runoff Ratio	-0.15	-0.12	-0.27	20.4	2.25
Annual Avg. Temp	-0.17	-0.07	-0.24	22.8	2.65
Summer Avg. Temp	-0.10	-0.16	-0.26	12.7	1.05
10yr avg. PET	-0.08	-0.19	-0.27	10.7	0.85
Wet Hg Deposition Concentration	-0.18	-0.14	-0.32	23.8	2.58

Table S5. The independent and joint variability explained by each driver of MeHg Yield from the hierarchal partitioning regression and Z scores. Bold and *italic* indicate significant at $p < 0.05$.

Driver	Independent Variability	Joint Variability	Total Variability	% Independent Variability	Z score
Streamflow	-0.13	-0.36	-0.50	12.1	1.07
Runoff Ratio	-0.18	-0.36	-0.53	16.2	1.82
Annual Avg. Temp	-0.10	-0.07	-0.17	9.1	0.73
Event Flow	-0.11	-0.19	-0.30	10.1	0.57
10yr avg. PET	-0.10	-0.30	-0.40	8.8	0.70
Wet Hg Deposition Concentration	-0.12	-0.29	-0.41	11.5	0.87
MeHg FWC	-0.35	-0.57	-0.92	32.2	3.96

References

- McCarter, C. P. R., Sebestyen, S. D., Coleman Wasik, J. K., Engstrom, D. R., Kolka, R. K., Jeremiason, J. D., et al. (2022). Long-Term Experimental Manipulation of Atmospheric Sulfate Deposition to a Peatland: Response of Methylmercury and Related Solute Export in Streamwater. *Environmental Science & Technology*. <https://doi.org/10.1021/acs.est.2c02621>
- Sebestyen, S. D., Lany, N. K., Oleheiser, K., Larson, J., Aspelin, N. A., Nelson, D. J., et al. (2022). *Marcell Experimental Forest chemistry of surface water draining the S6 catchment, 1986 - ongoing*. <https://doi.org/10.6073/pasta/9f8fb5bb7ce3d9549e2cab219fdc65d8>