Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supplementary Information: Understanding factors influencing the detection of mercury policies in modelled Laurentian Great Lakes wet deposition

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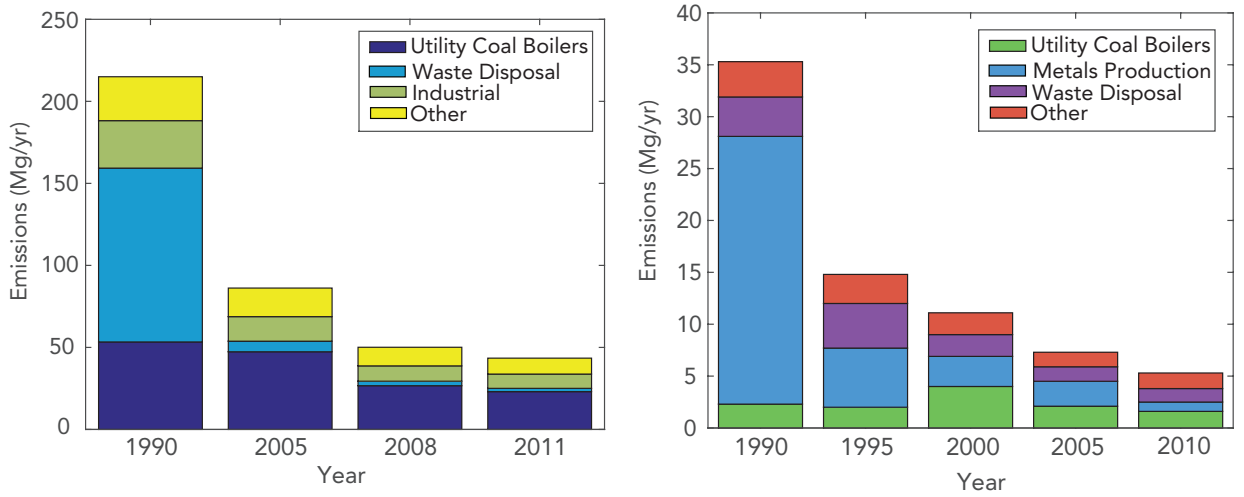


(a) United States

(b) Canada

Figure S1: Emissions trends in the US and Canada. Data for the US is from US EPA [3]. Data from Canada is from Steffen [4].

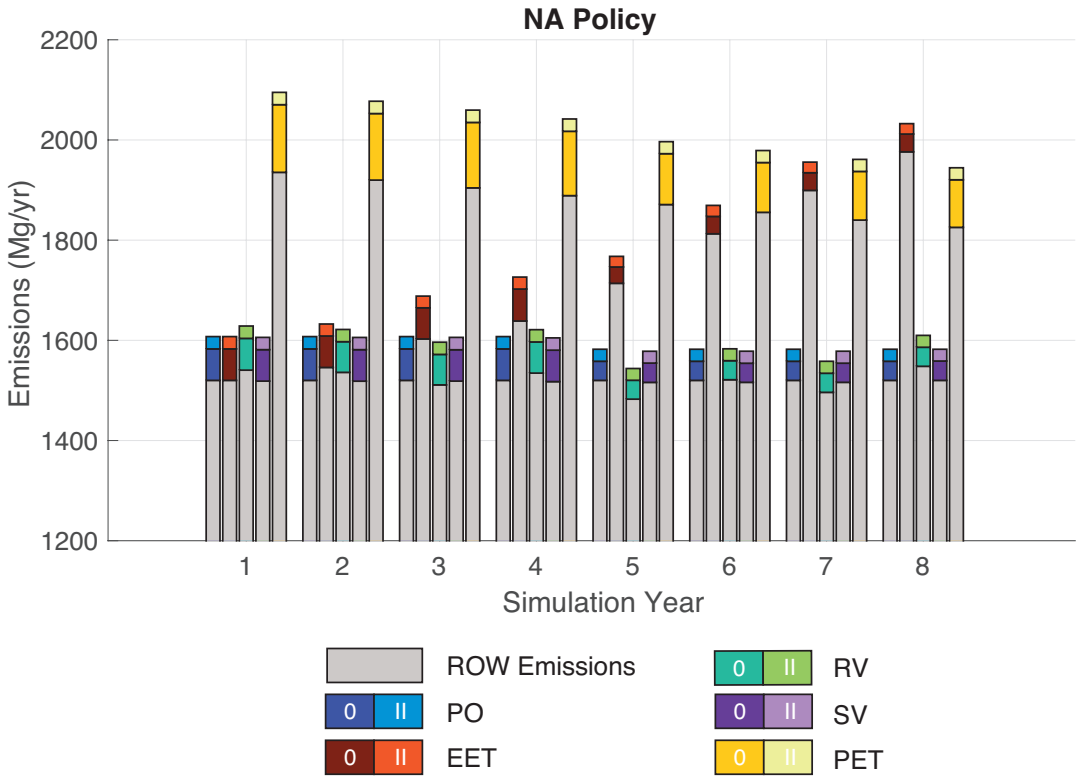


Figure S2: Summary of emissions by year under NA policy and noise scenarios. NA speciated emissions are shown in color (Hg0 = darker shade; Hg2/HgP = lighter shade), and ROW emissions, not speciated, are shown in grey. NA policy is implemented as a step change between 2008 and 2009. The "Meteorological Variability" and "Sectoral Trends + Meteorological Variability" scenarios are not shown as in the first, emissions are identical to "Policy Only," and in the second, emissions follow "Sectoral Trends." Note that the y-axis begins at 1200 Mg/yr.

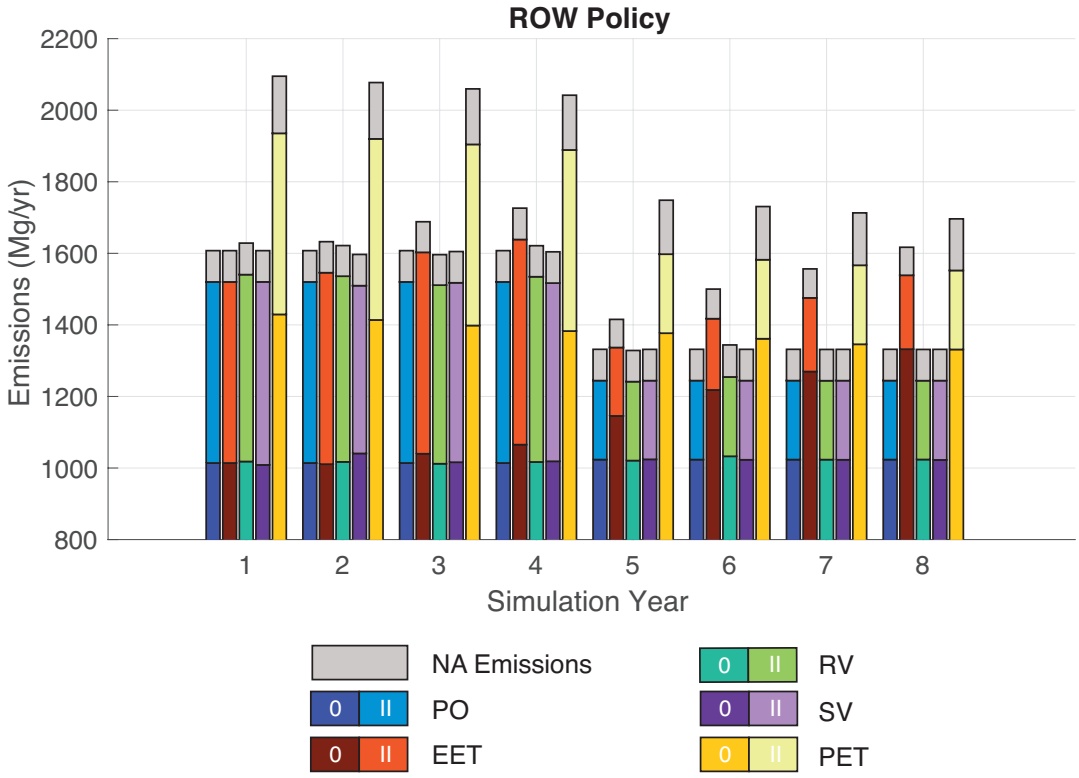


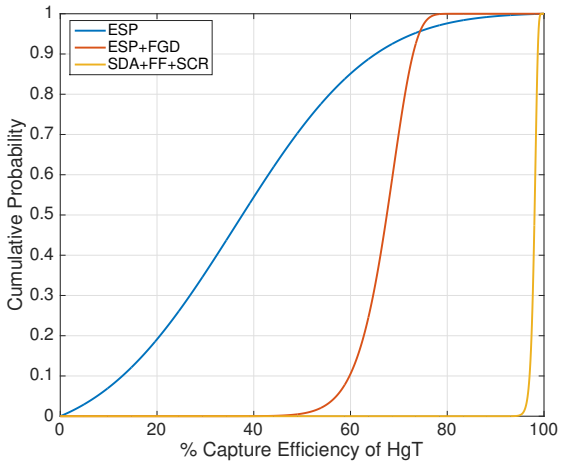
Figure S3: Summary of emissions by year under ROW policy and noise scenarios. ROW speciated emissions are shown in color (Hg0 = darker shade; Hg2/HgP = lighter shade), and NA emissions, not speciated, are shown in grey. ROW policy is implemented as a step change between 2008 and 2009. The "Meteorological Variability" and "Sectoral Trends + Meteorological Variability" scenarios are not shown as in the first, emissions are identical to "Policy Only," and in the second, emissions follow "Sectoral Trends." Note that the y-axis begins at 800 Mg/yr.

Table S1: Summary of emissions by simulation year (1-8) under NA policy. NA policy is implemented as a step change between years 4 and 5. Speciated emissions (0: elemental, II: divalent) are given for both NA and ROW.

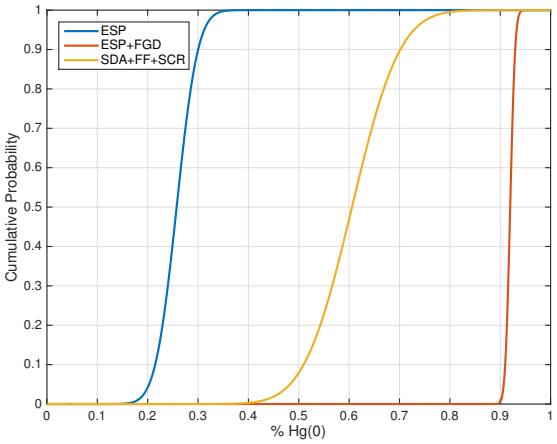
		Pre-NA Policy								Post-NA Policy							
Sim. Year		1		2		3		4		5		6		7		8	
Speciation		0	II	0	II	0	II	0	II	0	II	0	II	0	II	0	II
PO	NA	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	38.1	24.0	38.1	24.0	38.1	24.0	38.1	24.0
	ROW	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1
EET	NA	62.8	24.6	62.8	23.9	62.1	23.4	63.8	23.9	32.6	21.2	34.7	22.0	35.2	21.2	35.5	20.8
	ROW	1014.0	506.1	1010.8	535.1	1039.9	562.9	1065.3	573.3	1132.3	581.5	1204.3	608.4	1254.8	644.5	1317.2	659.1
PET	NA	135.0	24.6	132.8	24.6	130.6	24.6	128.4	24.6	101.6	24.0	99.4	24.0	97.2	24.0	95.0	24.0
	ROW	1429.3	506.1	1413.8	506.1	1398.3	506.1	1382.8	506.1	1364.9	506.1	1349.4	506.1	1333.9	506.1	1319.4	506.1
RV	NA	63.4	24.7	61.2	24.4	60.8	24.4	62.2	24.5	37.7	23.5	38.0	23.7	38.1	23.9	38.0	23.7
	ROW	1018.2	522.3	1017.2	518.9	1012.0	499.1	1016.9	517.7	1004.4	478.2	1012.4	509.0	1007.2	489.0	1017.9	530.3
SV	NA	62.8	24.5	62.9	24.4	62.6	24.7	62.8	24.5	38.5	23.6	38.1	24.0	38.2	23.9	38.6	23.5
	ROW	1008.7	509.9	1040.7	477.8	1016.2	502.4	1018.8	498.8	1003.1	513.0	1015.4	500.8	1010.0	506.2	1033.0	483.2
MV	NA	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	38.1	24.0	38.1	24.0	38.1	24.0	38.1	24.0
	ROW	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1

Table S2: Summary of emissions by simulation year (1-8) under ROW policy. ROW policy is implemented as a step change between years 4 and 5. Speciated emissions (0: elemental, II: divalent) are given for both NA and ROW.

		Pre-ROW Policy								Post-ROW Policy							
Sim. Year		1		2		3		4		5		6		7		8	
Speciation		0	II	0	II	0	II	0	II	0	II	0	II	0	II	0	II
PO	NA	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6
	ROW	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1023.6	220.8	1023.6	220.8	1023.6	220.8	1023.6	220.8
EET	NA	62.8	24.6	62.8	23.9	62.1	23.4	63.8	23.9	56.8	21.8	60.2	22.6	59.3	21.7	57.0	21.3
	ROW	1014.0	506.1	1010.8	535.1	1039.9	562.9	1065.3	573.3	1145.6	245.4	1218.2	255.7	1269.6	264.5	1332.1	265.4
PET	NA	135.0	24.6	132.8	24.6	130.6	24.6	128.4	24.6	126.2	24.6	124.0	24.6	121.8	24.6	119.6	24.6
	ROW	1429.3	506.1	1413.8	506.1	1398.3	506.1	1382.8	506.1	1376.8	220.8	1361.3	220.8	1345.8	220.8	1331.3	220.8
RV	NA	63.4	24.7	61.2	24.4	60.8	24.4	62.2	24.5	62.5	24.6	64.7	24.8	62.9	24.6	59.8	24.3
	ROW	1018.2	522.3	1017.2	518.9	1012.0	499.1	1016.9	517.7	1020.8	220.6	1032.8	221.7	1021.9	220.7	1020.8	220.6
SV	NA	62.8	24.5	62.9	24.4	62.6	24.7	62.8	24.5	62.6	24.7	62.4	24.8	62.9	24.5	62.7	24.6
	ROW	1008.7	509.9	1040.7	477.8	1016.2	502.4	1018.8	498.1	1023.5	220.5	1022.0	220.4	1023.5	220.5	1023.8	220.3
MV	NA	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6	62.8	24.6
	ROW	1014.0	506.1	1014.0	506.1	1014.0	506.1	1014.0	506.1	1023.6	220.8	1023.6	220.8	1023.6	220.8	1023.6	220.8



(a) Removal Fraction



(b) Fraction Hg(0)

Figure S4: Cumulative distribution functions of removal fraction and fraction Hg(0) for different air pollution control configurations at the individual plant level, using data from Bullock and Johnson [1].

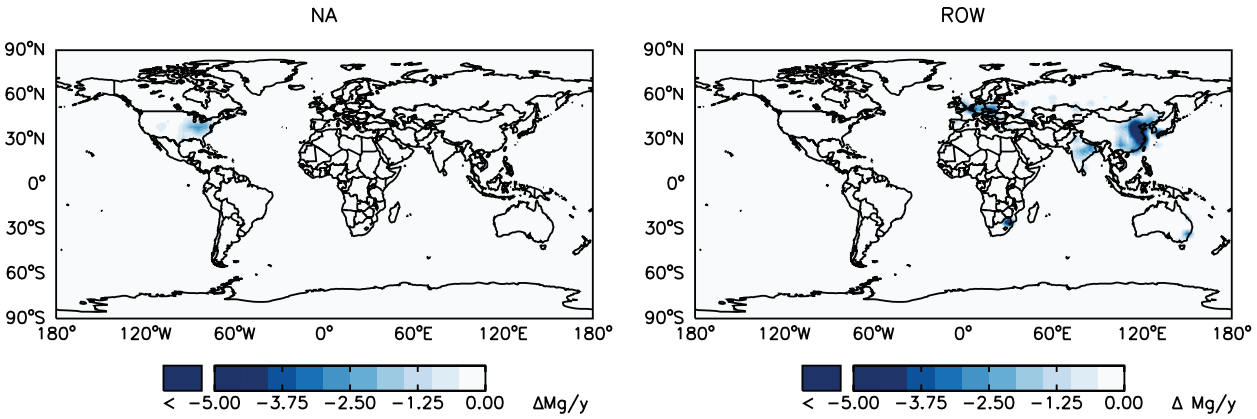


Figure S5: The spatial distribution of emissions changes associated with NA (left) and ROW (right) policy in Mg/y, between the pre- and post-policy period.

Table S3: Parameterization for variability in the population mean of removal fraction and fraction Hg(0) in flue gas, for air pollution control technology configurations. Parameters μ and σ are for normal distributions.

	μ	σ
Removal Fraction		
ESP	0.294	0.039
ESP+FGD	0.778	0.017
SDA+FF+SCR	0.974	0.007
Fraction Hg(0)		
ESP	0.258	0.033
ESP+FGD	0.920	0.008
SDA+FF+SCR	0.606	0.075

S2 Model-Observation Comparison

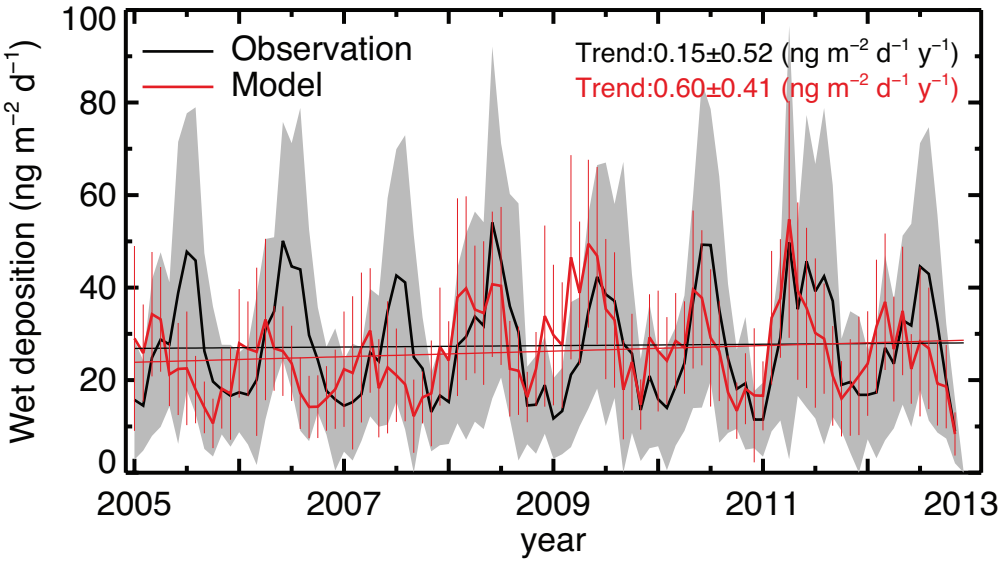


Figure S6: Model-observation comparison of monthly mean wet deposition fluxes from 2005-2012. Mean values from MDN sites are shown in black, with grey shading representing the standard deviation of individual sites. Modelled values are shown in red, with red bars indicating the standard deviation in individual sites. The Pearson correlation coefficient $r = 0.41$ for the entire period, while $r = 0.65$ in the recent period of 2010-2012.

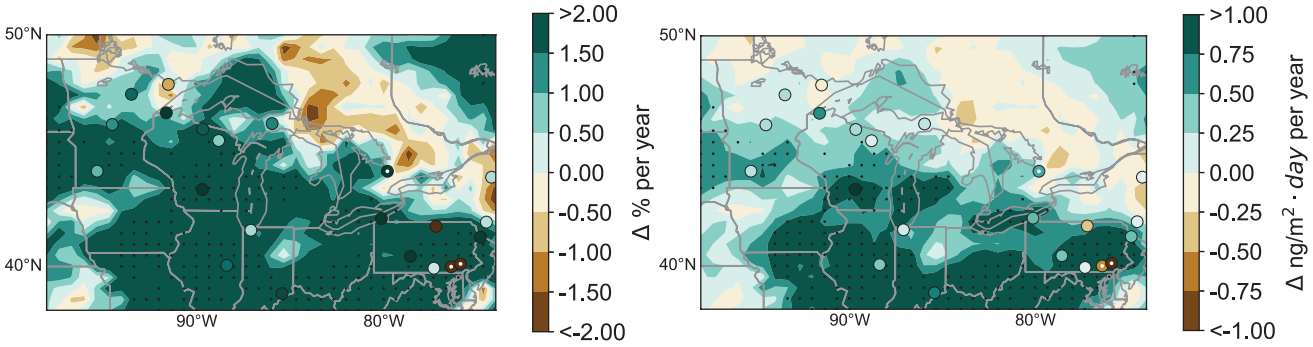


Figure S7: Comparison of modeled (background) and observed (filled circles) 8 year trends (2005-2012) in wet deposition, using SMK trend test and Theil-Sen estimator of slope. Monitoring sites and model grid cells with a significant trend ($p<0.1$) are indicated with a dot. Left compares trends in % change, while Right shows trends in ng/m^2·day per year. (Note: MDN measurements are taken as ng/m^2· week.)

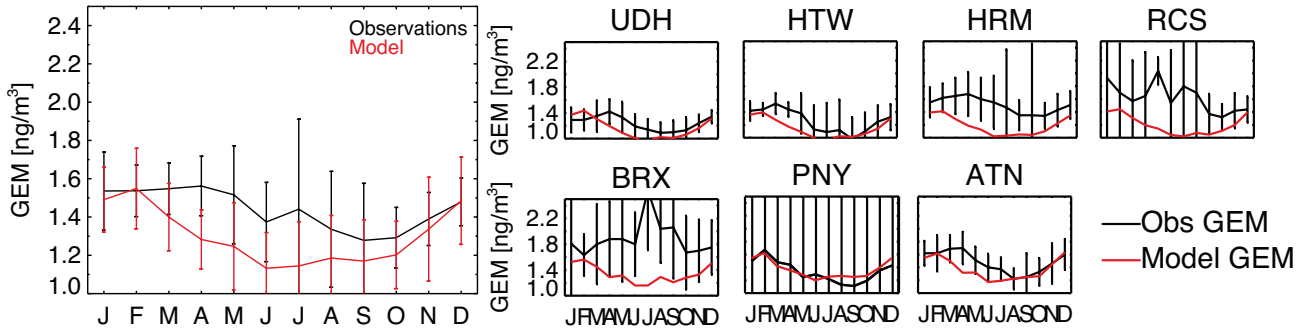
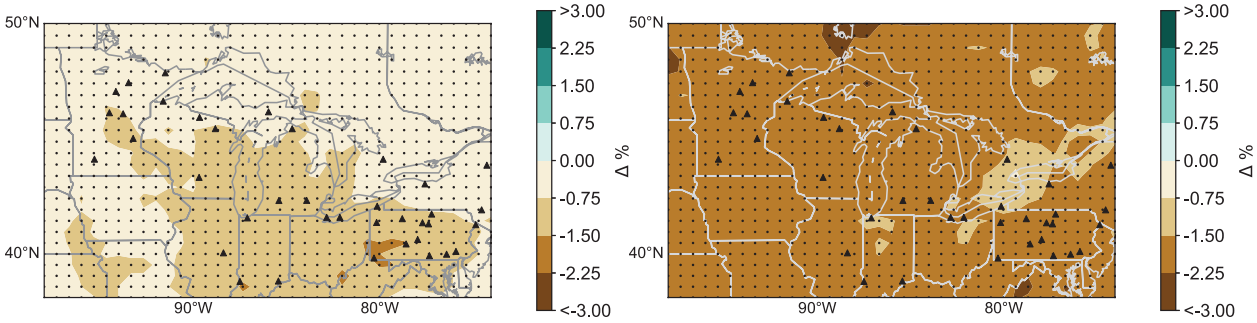


Figure S8: Comparison of modelled (red, 2012) and observed (black, 2013-2015) monthly mean surface gaseous elemental mercury concentrations. Left: Mean site values and standard deviation of individual sites. Right: Seasonal patterns at individual sites and standard deviation across years.

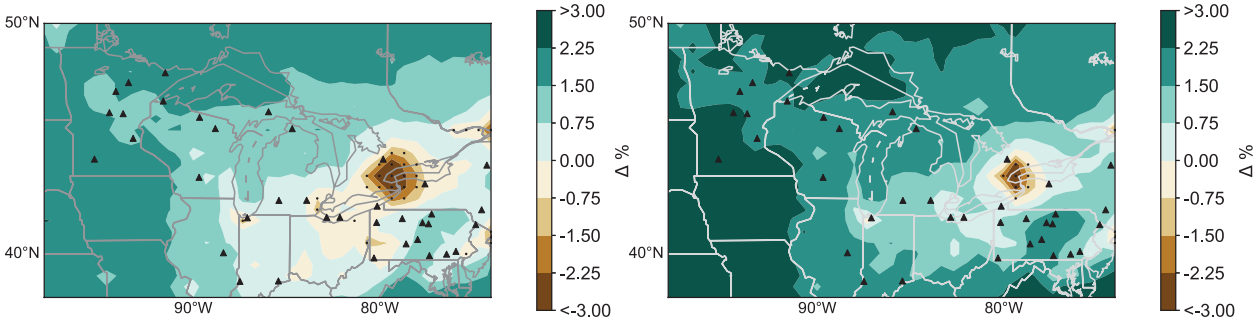
S3 Additional wet deposition results



(a) NA Signal

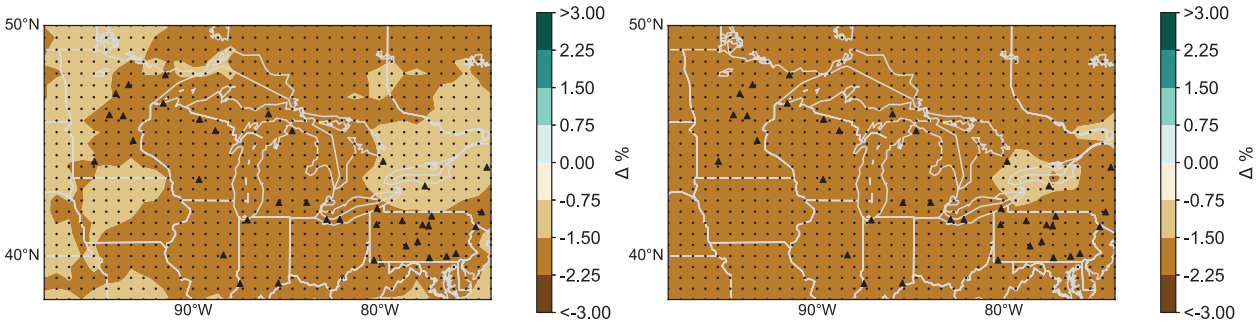
(b) ROW signal

Figure S9: Change in precipitation weighted concentration (%) between pre-policy and post-policy period in Policy Only simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



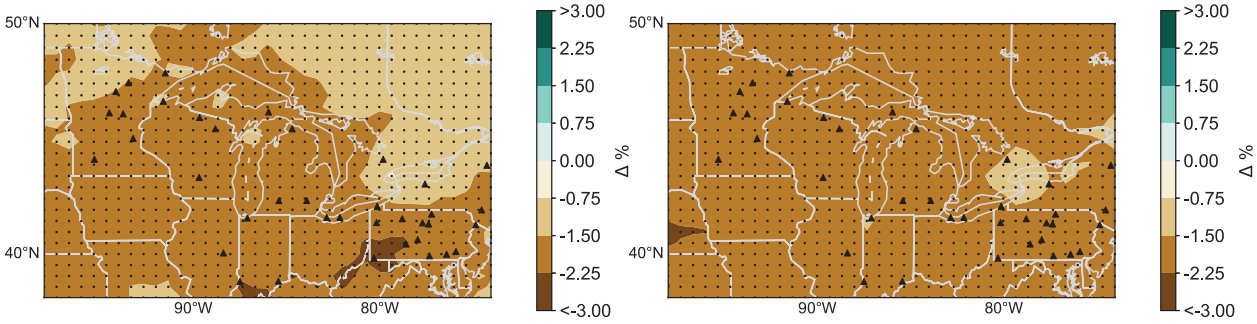
(a) NA Signal (b) ROW signal

Figure S10: Change in precipitation weighted concentration (%) between pre-policy and post-policy period in Energy and Economic Trends simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



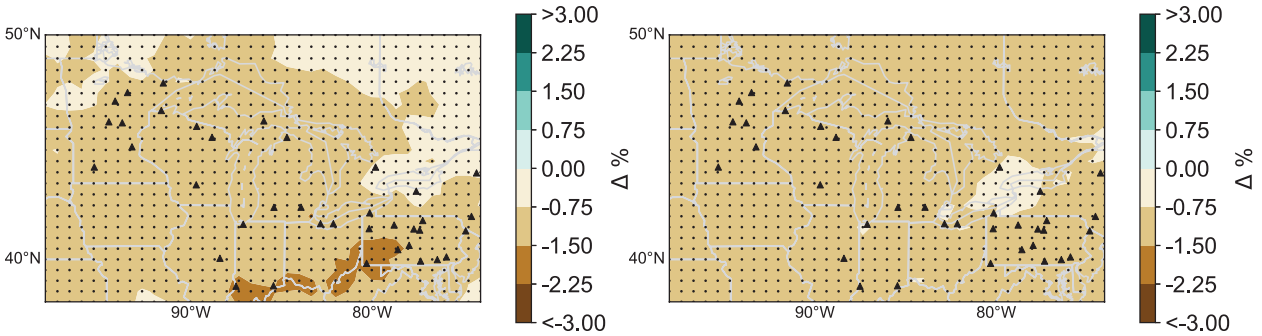
(a) NA Signal (b) ROW signal

Figure S11: Change in wet deposition (%) between pre-policy and post-policy period in Product Emissions Trend simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



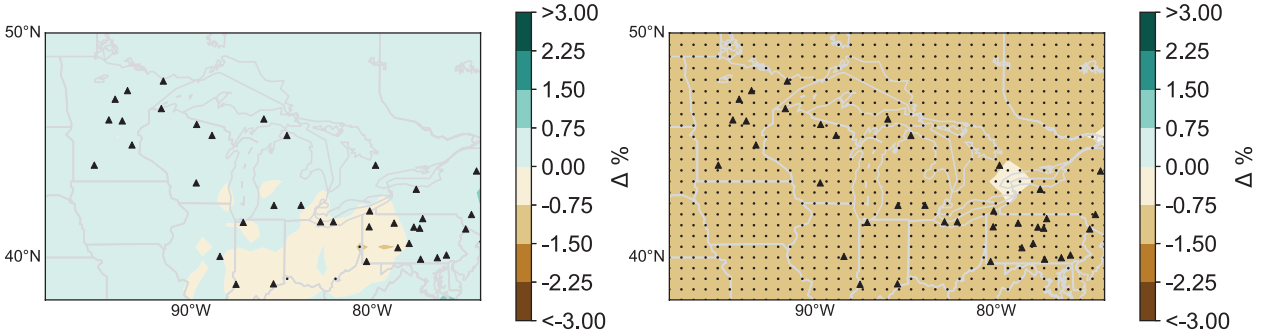
(a) NA Signal (b) ROW signal

Figure S12: Change in precipitation weighted concentration (%) between pre-policy and post-policy period in Product Emissions Trends simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



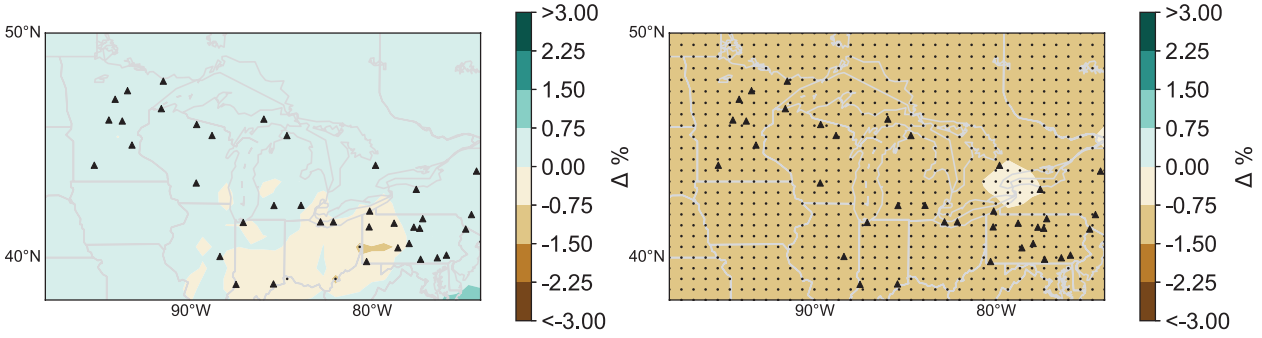
(a) NA Signal (b) ROW signal

Figure S13: Change in precipitation weighted concentration (%) between pre-policy and post-policy period in Removal Variability simulation. Grid cells with a significant (p<0.1) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



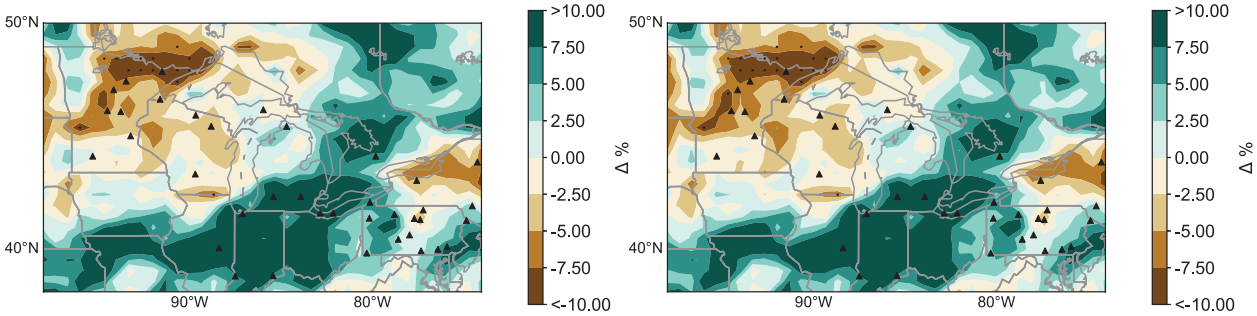
(a) NA Signal (b) ROW signal

Figure S14: Change in wet deposition (%) between pre-policy and post-policy period in Speciation Variability simulation. Grid cells with a significant (p<0.1) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



(a) NA Signal (b) ROW signal

Figure S15: Change in precipitation weighted concentration (%) between pre-policy and post-policy period in Speciation Variability simulation. Grid cells with a significant (p<0.1) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.



(a) NA Signal

(b) ROW signal

Figure S16: Change in precipitation weighted concentration (%) between pre-policy and post-policy period, for interannual meteorological variability simulation. Note the larger color bar range of -10 to 10%, compared to the other plots. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.

S4 Additional atmospheric concentration results

S5 Emissions Inventory Comparison

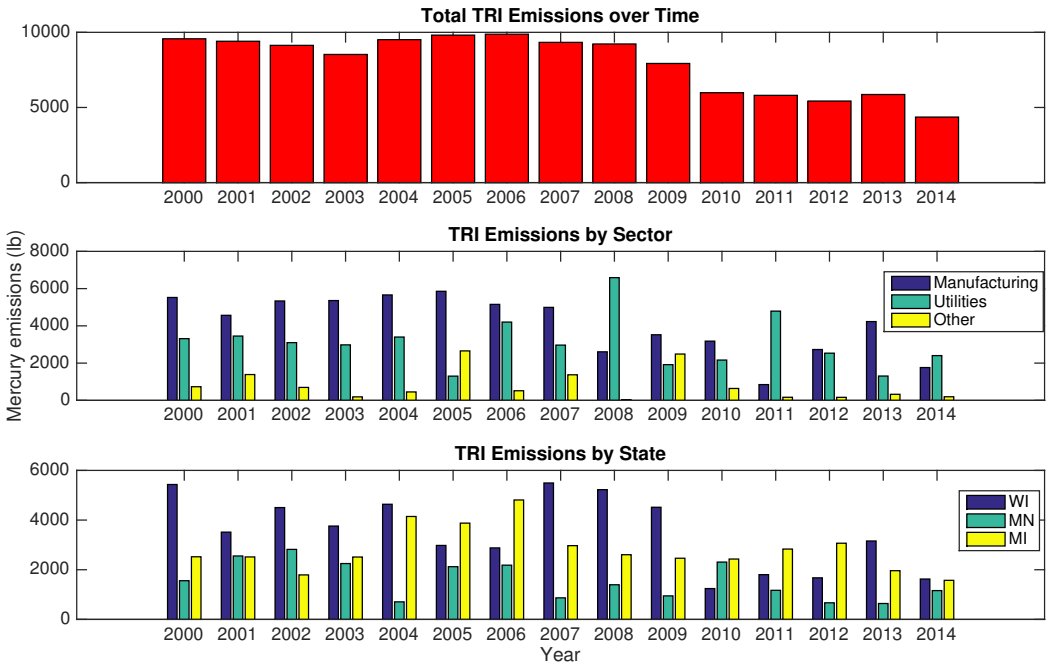


Figure S17: TRI time series for Lake Superior States, broken down by sector and state. For a full description of data sources, see Berg [2].

Table S4: NEI and TRI totals for fossil fuel emissions by state. 1999-2000 numbers are taken from Murray and Holmes [5], which used the 1999 NEI and 2000 TRI inventories. The 1999-2000 numbers only represent coal-fired power plants, while the 2008 and 2011 numbers are for all fossil fuel power generation.

	1999-2000	2008	2011
Michigan			
NEI	3094	2134	1936
TRI	3010	2990	2580
% diff	-2.71	40.09	33.25
Minnesota			
NEI	1265	1207	924
TRI	1497	1254	906
% diff	18.34	3.70	-2.01
Wisconsin			
NEI	2263	2313	1134
TRI	2114	1456	1263
% diff	-6.58	-58.85	11.36

Table S5: Step change in precipitation weighted concentration ($\Delta\%$) between the pre-policy and post-policy period under different simulated scenarios. Significance ($p<0.1$) and size of the step change are calculated using the Mann-Whitney-Wilcoxon Seasonal Rank Sum Test and the Hodges-Lehmann Estimator of Difference. Values in the table represent the average change across all sites and just those with significant changes. The share of sites with a significant change is shown in brackets (% of all sites).

		Policy Signal	Removal Variability	Speciation Variability	Met. Variability	Energy and Economic Trends	Product Trends
NA	all sites	-0.86%	-1.08%	-0.12.%	2.58%	-0.52%	-1.73%
	sig. sites	-0.86% (100%)	-1.08 (100%)	– (0%)	– (0%)	-2.11% (7%)	-1.73% (100%)
ROW	all sites	-1.79	-0.88%	-0.98%	2.47%	1.35%	-1.85%
	sig. sites	-1.79% (100%)	-0.88% (100%)	-0.98% (100%)	-2.47% (0%)	-1.86% (5%)	-1.85% (100%)

References

[1] D. Bullock, S. Johnson, Electric Generating Utility Mercury Speciation Profiles for the Clean Air Mercury Rule, Technical Report, US EPA, Research Triangle Park, NC, 2011.

[2] E. Berg, Mercury emissions inventories in the Lake Superior states, Undergraduate, Massachusetts Institute of Technology, 2016.

[3] US EPA, 2014 National Emissions Inventory , version 1 Technical Support Document, Technical Report, US Environmental Protection Agency, Office of Air Quality Planning and Standards, 2016.

[4] A. Steffen, Canadian Mercury Science Assessment: Summary of Key Results, Technical Report, Environment and Climate Change Canada, Toronto, 2016.

[5] M. Murray, S. A. Holmes, Assessment of mercury emissions inventories for the Great Lakes states, Environmental Research 95 (2004) 282–297.