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Supplementary Information

Decreases in Mercury Wet Deposition over the United States during 2004–2010: Roles of Domestic and Global Background Emission Reductions

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This supplementary material supports the named main text.

Table S1. Comparison of the regression results for Hg wet deposition between direct regression approach and regression after model value subtraction approach.

Region	Site	VWM Hg Concentration Trend						Precipitation Trend[b]	
		Model Subtraction		Direct Regression		Model[a]			
		% yr^{-1}	*p*	% yr^{-1}	*p*	% yr^{-1}	*p*	% yr^{-1}	*p*
Northeast	MA01	**−7.95**[c]	<0.01	**−9.00**	<0.01	*−1.43*	0.26	*2.50*	0.17
	ME02	**−4.02**	0.01	**−5.35**	<0.01	*−1.31*	0.21	*3.54*	0.12
	ME09	**−7.26**	<0.01	**−4.94**	0.01	*2.37*	0.20	**5.24**	0.01
	ME98	**−7.19**	<0.01	**−7.97**	<0.01	*−0.78*	0.34	*−0.44*	0.44
	NS01	**−2.40**	0.06	**−2.90**	0.06	*−0.50*	0.39	*3.41*	0.08
	NY20	**−3.19**	0.04	**−3.16**	0.04	*0.03*	0.49	*−0.62*	0.40
	NY68	**−5.09**	<0.01	**−5.25**	<0.01	*0.12*	0.47	*0.05*	0.49
	ON07	**−5.87**	0.01	**−4.49**	0.01	*1.53*	0.17	*3.08*	0.10
	PA00	*−1.64*	0.23	*−1.26*	0.29	*0.38*	0.40	*−1.73*	0.25
	PA13	**−2.96**	0.06	**−2.50**	0.05	*0.36*	0.41	*−1.23*	0.30
	PA30	**−3.60**	<0.01	*−1.64*	0.13	**1.95**	0.06	*−2.07*	0.17
	PA47	**−4.43**	<0.01	**−4.24**	0.02	*0.19*	0.45	*−0.86*	0.37
	PA60	**−7.38**	<0.01	**−6.83**	0.01	*0.61*	0.34	*2.19*	0.21
	PA72	**−2.35**	0.07	**−2.64**	0.10	*−0.30*	0.41	*0.63*	0.41
	PA90	*−1.07*	0.23	*−0.13*	0.47	*0.94*	0.24	**−4.84**	0.02
	VA28	*−2.52*	0.17	*−2.58*	0.13	*−0.19*	0.46	*−4.81*	0.15
	Regional	**−4.3 ± 2.2**[d]		**−4.1 ± 2.4**		*0.24 ± 1.1*		*0.25 ± 2.9*	

Table S1. *Cont.*

Region	Site	VWM Hg Concentration Trend						Precipitation Trend[b]	
		Model Subtraction		Direct Regression		Model[a]			
		% yr^{-1}	*p*	% yr^{-1}	*p*	% yr^{-1}	*p*	% yr^{-1}	*p*
Midwest	IL11	*−0.48*	0.41	*−0.79*	0.34	*−0.30*	0.42	*2.03*	0.27
	IN34	*0.11*	0.47	*0.53*	0.37	*0.31*	0.42	*2.96*	0.20
	KY10	**−2.93**	0.06	*−0.91*	0.30	*2.02*	0.11	*−3.38*	0.22
	MI48	*−2.10*	0.17	*−0.32*	0.44	*1.79*	0.10	*0.70*	0.42
	MN16	*−0.08*	0.49	*−0.07*	0.49	*−0.03*	0.49	*0.88*	0.38
	MN23	*−2.40*	0.13	*−1.03*	0.31	*1.30*	0.15	**4.59**	0.07
	MN27	**−4.14**	0.08	**−3.68**	0.10	*0.52*	0.33	*1.22*	0.37
	MO46	**−4.45**	0.02	**−4.15**	0.05	*0.30*	0.41	*0.89*	0.37
	WI08	**−3.20**	0.07	*−2.25*	0.17	*0.80*	0.24	**6.13**	0.03
	WI22	**−4.25**	0.04	**−4.32**	0.03	*−0.07*	0.48	*4.52*	0.12
	WI31	**−4.78**	0.03	**−4.49**	0.04	*−0.22*	0.44	*3.17*	0.22
	WI36	*−1.96*	0.18	*−1.12*	0.27	*0.72*	0.30	*1.18*	0.34
	WI99	*−2.18*	0.22	*−0.53*	0.40	*1.64*	0.23	*3.48*	0.15
	Regional	**−2.5 ± 1.6**		**−1.8 ± 1.8**		*0.68 ± 0.79*		*2.2 ± 2.4*	
Southeast	AL03	*−0.18*	0.48	**2.64**	0.09	*2.82*	0.18	*1.02*	0.36
	FL05	*−0.58*	0.37	*−1.07*	0.24	*−0.21*	0.43	*−1.02*	0.36
	FL11	*−1.23*	0.24	*−1.28*	0.22	*−0.17*	0.44	*0.29*	0.47
	FL34	*−0.02*	0.49	*−1.24*	0.22	*−0.88*	0.13	**9.57**	<0.01
	GA09	*1.83*	0.15	*0.20*	0.45	**−1.63**	0.07	**−7.96**	0.02
	GA40	*−1.92*	0.22	*0.62*	0.38	**2.61**	0.10	**7.27**	0.01
	MS22	*1.29*	0.26	*−0.29*	0.45	**−1.58**	0.07	*−1.70*	0.33
	NC08	*−1.86*	0.13	*0.95*	0.27	**2.78**	0.02	**−5.08**	0.07
	NC42	**−4.76**	0.02	*0.01*	0.50	**4.77**	<0.01	**−7.11**	0.02
	SC05	*−0.07*	0.49	**3.46**	0.02	**3.52**	0.04	*−2.50*	0.25
	SC19	**−3.76**	0.03	*−1.79*	0.16	**1.97**	0.08	*1.48*	0.33
	TN11	**−1.85**	0.08	*−1.59*	0.11	*0.26*	0.42	**−3.97**	0.06
	TX21	**4.96**	0.03	**6.04**	0.01	*0.60*	0.38	*−3.12*	0.19
	Regional	*−0.63 ± 2.5*		*0.51 ± 2.3*		**1.1 2.1**		*−0.99 ± 5.1*	
West	CA75	**−11.61**	0.06	*−6.61*	0.14	*11.36*	0.26	**12.18**	0.01
	CO97	*2.66*	0.15	**4.46**	0.03	*1.56*	0.18	*1.39*	0.27
	CO99	**6.36**	0.01	**7.69**	<0.01	*0.96*	0.25	*3.94*	0.15
	NV02	**6.93**	0.06	**7.77**	0.04	*0.83*	0.23	*−0.56*	0.45
	WA18	*−2.69*	0.20	*−1.88*	0.27	*0.07*	0.48	*1.73*	0.30
	Regional	*0.33 ± 7.7*		**2.3 ± 6.3**		*2.0 ± 4.7*		**3.7 ± 5.0**	

[a] Trends of VWM Hg concentrations calculated in the BASE model simulation. [b] Trends of precipitation observed at MDN sites. [c] Significant trends ($p < 0.1$) are indicated in bold, insignificant trends are indicated in italics. [d] Regional trends calculated with the random coefficient model. Significant trends ($p < 0.05$) are indicated in bold fonts, trends that are insignificant are indicated in italics.

Figure S1. Map of the 47 Mercury Deposition Network (MDN) sites used in this study. The red rectangles define the boundaries of the four regions considered.

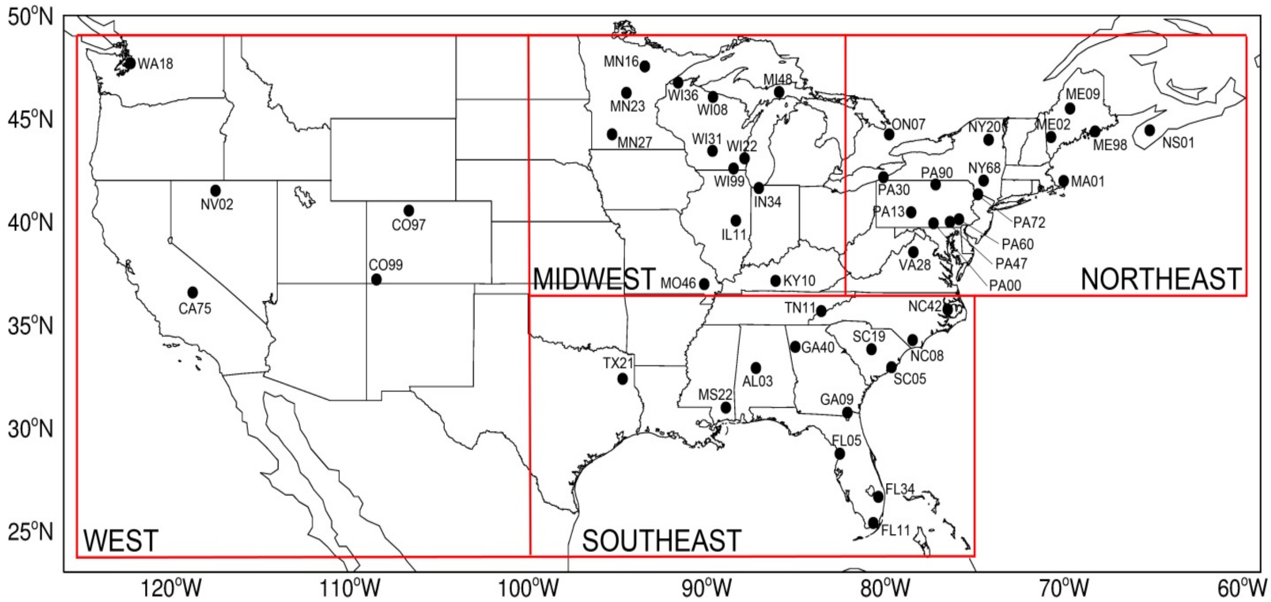


Figure S2. Correlation between observed summer time (JJA) weekly precipitation and Hg wet deposition flux (left column) and between precipitation and Hg concentrations in precipitated rain/snow for 2004−2010. (**Top**): 47 MDN sites. (**Middle**): PA30 site. (**Bottom**): TN11 (bottom panels). The red line is a linear regression for all the points with slope and r^2 values shown in each panel. The shaded area indicates the regime where the Hg concentration in precipitation is less sensitive than Hg wet deposition flux to the change of precipitation depth.

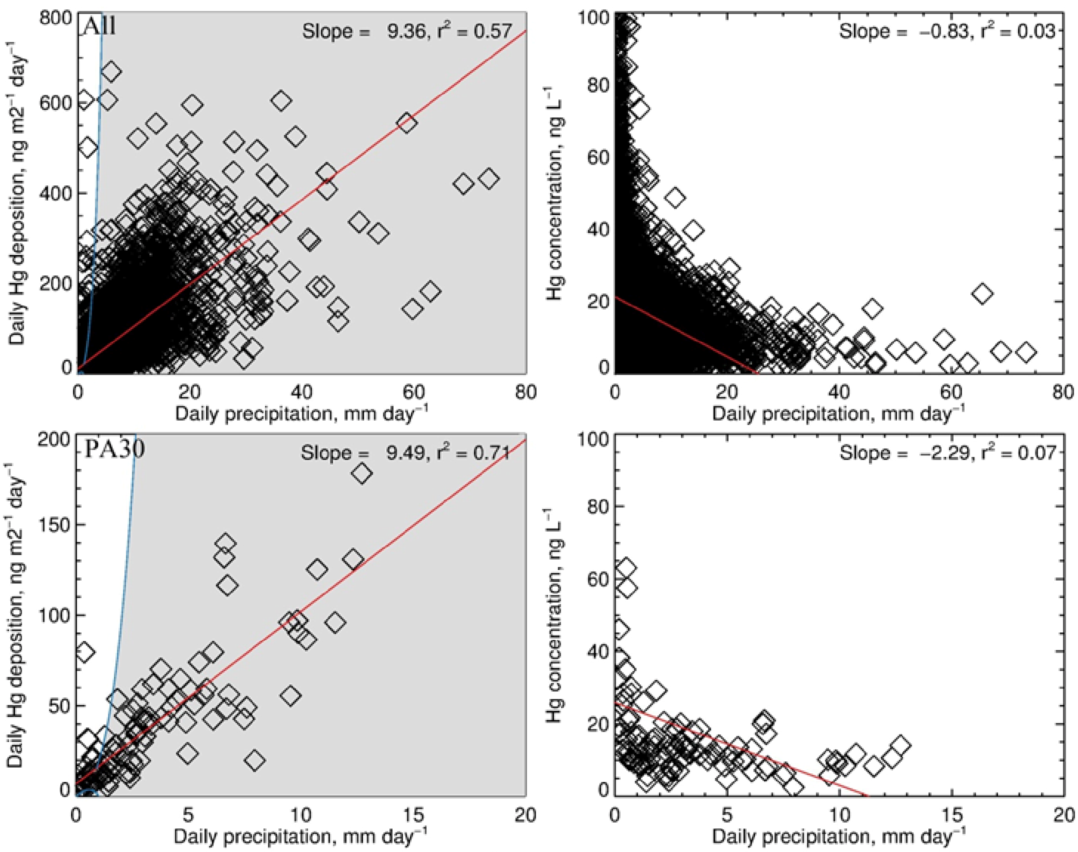
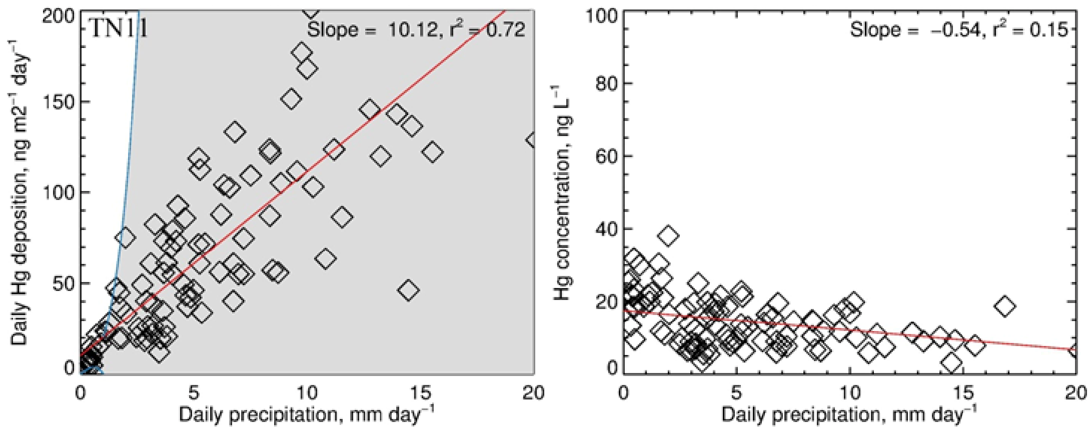


Figure S2. *Cont.*



Sensitivity of Different Analytical Variables to the Change in Precipitation

The sensitivity of Hg wet deposition flux (y) to precipitation depth (x) is defined as:

$$S_{\text{flux}} = \left|\frac{\frac{dy}{y}}{\frac{dx}{x}}\right| = \frac{x}{y}\left|\frac{dy}{dx}\right| = k\frac{x}{y} \quad (1)$$

where k is the slope of the regression line between y and x, as illustrated in the left panels of Figure S2. The sensitivity of Hg concentration in precipitation ($\frac{y}{x}$) to x can be written as:

$$S_{concentration} = \left|\frac{\frac{d\frac{y}{x}}{\frac{y}{x}}}{\frac{dx}{x}}\right| = \frac{1}{x^2}\left(\frac{x}{y}\left|\frac{dy}{dx}\right|\right) - \frac{1}{x^2} = \frac{1}{x^2}\left|S_{flux} - 1\right| \quad (2)$$

Therefore, $S_{\text{concentration}}$ would be less than S_{flux} if:

$$\frac{1}{x^2}\left|S_{flux} - 1\right| < S_{flux} \quad (3)$$

Combining Equations (1) and (3), we obtain:

$$kx^3 > |kx - y| \quad (4)$$

The regime satisfying Equation (4) is shaded in grey on the left column of panels in Figure S2. Among all the data points, 86.4% belong to this regime. At PA30 and TN11, 85.7% and 90.4% of the points fall into this regime, respectively. This indicates less sensitivity for Hg concentration in precipitation to the variability of precipitation depth than Hg wet deposition.