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Complete List of Authors:	Giang, Amanda; University of British Columbia, Institute for Resources, Environment and Sustainability; Massachusetts Institute of Technology, Institute for Data, Systems and Society Song, Shaojie; Massachusetts Institute of Technology, Earth, Atmospheric and Planetary Sciences ; Harvard University, John A. Paulson School of Engineering and Applied Sciences Muntean, Marilena; European Commission Joint Research Centre Ispra Sector, Directorate for Energy, Transport and Climate Janssens-Maenhout, Greet; European Commission Joint Research Centre Ispra Sector, Directorate for Sustainable Resources, Knowledge for Sustainable Development & Food Security Harvey, Abigail ; Massachusetts Institute of Technology, Earth, Atmospheric and Planetary Sciences ; Massachusetts Institute of Technology, Civil and Environmental Engineering Berg, Elizabeth; Massachusetts Institute of Technology, Earth, Atmospheric and Planetary Sciences Selin, Noelle; Massachusetts Institute of Technology, Engineering Systems Division

Mercury is a toxic, global pollutant that can harm human and ecosystem health. Consequently, policies from local to global scales aim to control and reduce anthropogenic emissions of this pollutant. Here, we use chemical transport modelling to evaluate the extent to which these policies may translate into statistically significant changes in wet deposition inputs to the Laurentian Great Lakes region. We find that on a subdecadal scale, sources of noise, such as variability in meteorology or air pollution control performance, may reduce and in some cases obscure policy signals. These results suggest that the magnitude of the policy signal, noise from environmental and human systems, and evaluation timescale should all be considered in both policy design and evaluation.

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

Amanda Giang,[a,e] Shaojie Song,[b,f] Marilena Muntean,[c] Greet Janssens-Maenhout,[d] Abigail Harvey,[b] Elizabeth Berg,[b] Noelle E. Selin[a,b]

[a]Institute for Data, Systems and Society, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
[b]Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
[c]European Commission, Joint Research Centre, Directorate for Energy, Transport and Climate, Air and Climate Unit, Ispra, VA I-21027, Italy
[d]European Commission, Joint Research Centre, Directorate for Sustainable Resources, Knowledge for Sustainable Development & Food Security Unit, Ispra VA I-21027, Italy
[e]Currently at Institute for Resources, Environment and Sustainability and Department of Mechanical Engineering, University of British Columbia, Vancouver, BC V6T 1Z4, Canada
[f]Currently at John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA

Abstract

We use chemical transport modelling to better understand the extent to which policy-related anthropogenic mercury emissions changes (a policy signal) can be statistically detected in wet deposition measurements in the Great Lakes region on the subdecadal scale, given sources of noise. In our modelling experiment, we consider hypothetical regional (North American) and global (rest of the world) policy changes, consistent with existing policy efforts (Δ_{global} = -18%; ($\Delta_{regional}$ = -30%) that divide an eight-year period. The magnitude of statistically significant ($p<0.1$) pre- and post-policy period wet deposition differences, holding all else constant except for the policy change, ranges from -0.3 to -2.0% for the regional policy and -0.8 to -2.7% for the global policy. We then introduce sources of noise—trends and variability in factors that are exogenous to the policy action—and evaluate the extent to which the policy signal can still be detected. For instance, technology-related variability in emissions magnitude and speciation can shift the magnitude of differences between periods, in some cases dampening the policy effect. We find that interannual variability in meteorology has the largest effect of the sources of noise considered, driving deposition differences between periods ±20%, exceeding the magnitude of the policy signal. However, our simulations suggest that gaseous elemental mercury concentration may be more robust to this meteorological variability in this region, and a stronger indicator of local/regional emissions changes. These results highlight the potential challenges of detecting statistically significant policy-related changes in Great Lakes wet deposition within the subdecadal scale.

1.0 Introduction

Mercury—a bioaccumulative toxin, particularly in its organic forms—poses risks to public health and the environment.[1] Consequently, anthropogenic mercury emissions have been the target of policy action, from local to global scales.[2–5] For instance, emissions to the atmosphere in the United States and Canada have decreased by more than 75% since 1990, from 246 Mg/yr in 1990 to 55 Mg/yr in 2014 in the US,[6] and from 35 Mg/yr in 1990 to 6 Mg/yr in 2010 in Canada [7] (see Supplementary Information Figure S1). Domestic regulations targeting waste incineration (particularly in the US) and metals production (particularly in Canada) contributed to steep declines in the 1990s, and since the mid-2000s, regulations targeting other air pollutants in addition to mercury have contributed to more modest decreases from the electricity generation sector.[8,9] In the future, the United Nations Minamata Convention on Mercury, which entered into force in August 2017, may lead to reductions in emissions globally.[10–13]

Coal combustion is estimated to be the second largest source of anthropogenic mercury emissions globally, after artisanal and small scale gold mining,[14] and given potential growth in energy demand from global economic development,[15] decoupling energy production from mercury emissions is a potentially important part of mitigation efforts.[13] Between the mid 2000s and mid 2010s, regulations targeting pollutant emissions from power plants in the US and Canada—for instance, in the US the Clean Air Interstate Rule and its replacement, the Cross State Air Pollution Rule,[16] and the Clean Air Mercury Rule and its replacement, the Mercury and Air Toxics Standards [17]—led to the increased adoption of end-of-pipe air pollution control devices.[16,18] While many of these controls are not mercury-specific and target particulate matter (PM), SO_2, and NO_x, they also capture mercury as a co-benefit.[19] Globally, the Minamata Convention requires that parties apply best available techniques and best environmental practices for controlling mercury emissions from sources like coal-fired power plants, which includes co-benefit mercury capture from a range of air pollution control devices.[20] In China, adoption of these approaches in the electricity generation sector to address air quality concerns have already led to reductions in mercury emissions per unit coal.[21,22]

To what extent can these, or similar, policy-related emissions decreases be detected in changes in mercury inputs to specific vulnerable ecosystems? In the Laurentian Great Lakes region, where mercury remains a concern for human and wildlife health,[23–25] many community stakeholders in mercury management (including Indigenous communities and recreational anglers) are interested in whether these policies translate into decreases in atmospheric loadings of mercury to aquatic ecosystems, and ultimately, decreases in dietary human exposure from fish.[26,27] As recent source attribution modelling studies have highlighted the importance of both local/regional and global anthropogenic sources for deposition in the Great Lakes basin,[25,28–30] this question is important not only for evaluating the effectiveness of historical and future policy efforts in North America at protecting human health in this region, but also for evaluating the potential impact of prospective policy actions elsewhere in the world in response to the Minamata Convention.

Recent studies report statistically significant declines in observed mercury wet deposition aggregated over North America between the mid-1990s and early 2010s,[8,31,32] and that these long-term, large-scale declines have been driven by anthropogenic emissions changes.[8,33]

However, spatially and temporally disaggregated trends within this larger spatio-temporal region show much heterogeneity.[30,32,34–38] The National Atmospheric Deposition Program (NADP) Mercury Deposition Network (MDN) collects weekly integrated wet deposition samples at monitoring sites in the US and Canada, with a continuous data record beginning in 1996 for the longest running sites.[39] In an analysis of this monitoring data, Weiss-Penzias et al.[32] found significant negative trends in wet deposition concentration in 53% of sites with data from 1997-2013 (ranging from -0.5 to -1.8 % per year), but that this fraction of sites decreased substantially to 6% when considering only the more recent period of 2008-2013, when 30% of sites showed significant positive trends in wet deposition concentration. Regionally, positive trends were concentrated in the central and western areas of the continent, while negative trends were concentrated in the eastern areas.[32]

These results are consistent with previous analyses of MDN data: Prestbo and Gay[35] found significant decreases in concentration in the range of -1 to -2% per year between 1996-2005 in the Northeast and Mid-Atlantic regions, but no significant trends in the upper Midwest (including Minnesota and Wisconsin) or lower Southeast, and Butler et al.[34] found significant declines in the Northeast and Midwest (defined to include parts of the Ohio River Valley), but no trend in the Southeast from 1998-2005. Focusing on the Great Lakes region, Risch et al.[38] reported small statistically significant decreases in Hg concentration between 2002 and 2008, but no significant trends in wet deposition, as decreases in concentration in precipitation were coupled with increases in precipitation volume. However, considering a 16 year period (2001-2016), Risch and Kenski[18] found statistically significant decreases in Hg wet deposition between 2001-2013 and 2014-2016 (of 16%) in Indiana, suggesting that the implementation of regulations targeting utility boilers, cement kilns, and medical waste incineration in the mid 2010s may have contributed to observed declines in this area rich with local emissions sources. Importantly, changes of this magnitude exceed reported measurement variability in collocated samplers (11% for concentration, 8.5% for deposition), increasing certainty in the observed decrease.[40]

Is the lack of significant negative trends and increasing prevalence of positive ones between the early 2000s and early 2010s (particularly in central and western regions of the US) consistent with the approximately 50% decline in North American emissions (see Figure S1) during this period? Several studies have advanced hypotheses to explain the spatial and temporal pattern of trends observed at North American monitoring sites. These include: uncertainties in both magnitude and speciation of emissions inventories;[8] decreasing influence of local/regional sources given increasing global background concentration of atmospheric mercury, driven by emissions growth in Asia;[32] and meteorological and climatological variability.[37,41,42] Zhang et al.[8] find better agreement between modelled and observed twenty-year (1990-2010) trends in elemental mercury and mercury wet deposition in North America and Europe after revising emissions inventories to take into account decreasing emissions from commercial products and artisanal and small scale gold mining, and changes in flue gas speciation due to adoption of air pollution control devices. Weiss-Penzias et al.[32], based on their interpretation of spatial patterns in observed wet deposition and concentration patterns, suggest that the recent positive trends in the central and western US may be due increases in the transpacific transport of mercury in tropospheric air masses, which have larger influences over these regions. Finally, Gratz et al.[37] propose that interannual variability in local meteorology—particularly precipitation amount and

type—can mask the influence of emissions in wet deposition concentration at a remote northeastern site. Shah et al.[41], using a modelling approach, reach similar conclusions on the contribution of precipitation to variability in wet deposition, while also highlighting the importance of meteorological factors that affect the production and export of divalent mercury to free tropospheric air, like subtropical anticyclones. Mao et al.[42] also point to the contribution of large-scale circulation patterns, such as the North Atlantic Oscillation, on wet deposition trends in the Adirondack region.

The goal of this study is to use atmospheric modelling to better understand which of these hypothesized factors affect the translation of prospective policy-related emissions changes into changes in wet deposition (concentration and flux) in the Great Lakes region on a subdecadal scale, and to quantify their relative influence. In this work, we use modelling experiments to explore the extent to which variability and trends in these intervening factors, exogenous to policy action, can act as "noise" in the detection of a policy "signal" in monitored wet deposition in the Great Lakes. We consider these dynamics for both regional (North American) and global policy signals. We focus on a subdecadal scale because this is a relevant time frame for policy evaluation: for instance, the Minamata Convention calls for effectiveness evaluation six years after entry into force,[5] and certain air quality standards in the US are required to be reviewed on a five-year cycle.[43] We discuss how the results of this analysis can be used to help interpret observed trends, and the potential implications of these signal-to-noise challenges for policy monitoring and design, for instance in the context of the Minamata Convention.

2.0 Methods

2.1 Overall approach

We begin with an analysis of historical observations from 2005-2012, to replicate trends reported in the literature and evaluate the ability of the chemical transport model to capture spatial patterns and magnitudes of wet deposition over the Great Lakes region. Then, in our modelling experiment, we consider a hypothetical step policy change, consistent with existing policy efforts, requiring the application of increased air pollution control devices in the electricity generation sector that divides this eight-year period, resulting in a four-year pre-policy period and a four-year post-policy period (Table 1). We consider a regional policy, targeting North America (NA), and a policy that targets the rest of the world (ROW) separately, to evaluate the influence of regional and global "policy signals" on the Great Lakes region. We define "policy signal" as the percent difference between pre- and post-policy period for any given metric (precipitation weighted concentration, deposition, and precipitation). We first evaluate the strength of this signal over the region holding all else constant except emissions in the sector targeted by policy. We then introduce sources of noise—trends and variability in factors that are exogenous to the policy action—and evaluate the extent to which the policy signal can still be detected. These scenarios are summarized in Table 2 and the emissions resulting from these scenarios are summarized numerically in Tables S1 and S2 and visually in Figures S2 and S3. The scenarios are also described in detail in Section 2.4.2. To explore whether all atmospheric indicators are equally sensitive to these sources of noise, for selected scenarios we repeat the analysis using modelled gaseous elemental mercury (GEM) concentration as the indicator of interest.

As has been noted in the literature, observed patterns and trends of wet deposition—particularly on the decadal scale—are highly dependent on which start and end dates are chosen;[32] the goal of this analysis is to provide insight into the factors that lead to this variability. In this light, our focus on the eight-year period of 2005-2012 is illustrative in that it is meant to illuminate the relative influence of sources of noise that operate on a decadal scale. Given this goal, while selecting a different eight-year window would have been possible, our focus on 2005-2012 is due to the richer availability of data (in monitoring, detailed sectoral emissions, air pollution control technology) during this period, which supports the development of "noise" scenarios that reflect real-world variability. Similarly, our focus on wet deposition (flux and concentration) as the metric of interest is due to the larger spatial and temporal coverage of wet deposition monitoring stations (compared to atmospheric mercury concentration) and its importance as a vector for mercury inputs into the Great Lakes[44]—both factors that contribute to the continuing relevance of wet deposition observations as a means of evaluating past and future policy efforts.

Table 1 Technology standard policy change targeting emissions from electricity generation sector. Removal fractions and speciation profiles are based on data from Bullock and Johnson,[45] collected for the US EPA.

	Region	**Technology**	**Emissions**
Pre-Policy Period (4 years)	NA	ESP+FGD *Removal: 77.8%* *Speciation: 92% Hg(0), 8% Hg(II)*	87 Mg/yr
	ROW	ESP *Removal: 29.4%* *Speciation: 26% Hg(0), 74% Hg(II)*	1520 Mg/yr
		Policy Change	
Post-Policy Period (4 years)	NA	SDA+FF+SCR *Removal: 97.8%* *Speciation: 49% Hg(0), 51% Hg(II)*	62 Mg/yr
	ROW	ESP+FGD *Removal: 77.8%* *Speciation: 92% Hg(0), 8% Hg(II)*	1244 Mg/yr

Table 2 Descriptions of modelling experiments. For all experiments, the step policy change (described for the Policy Only experiment) is applied after year 4, leading to 4 year pre- and post-policy periods.

Modelling Experiment		**Description**	**Emissions Years**	**Meteorological Years**
No Noise	Policy Only	See Table 1 for a full description of the policy scenario; all else held constant except for policy change	2005 × 8	2005 × 8
Trend	Energy and Economic Trends	Trends in energy and economic activity for all sectors, and control technology adoption in sectors not targeted by regulation	2005-2012	2005 × 8
	Product Emission Trends	Decreasing trend in additional source of	2005 × 8	2005 × 8

		Hg(0) emissions from commercial products		
Variability	Removal Variability	Interannual variability in the removal fraction of air pollution control devices in the power generation sector	2005 × 8	2005 × 8
	Speciation Variability	Interannual variability in fraction Hg(0) of end-of-pipe emissions in the power generation sector	2005 × 8	2005 × 8
	Meteorological Variability	Interannual variability in meteorology (e.g. precipitation magnitude and type, wind patterns)	2005 × 8	2005-2012

2.2 Analysis of observations

To assess historical trends in the Great Lakes region between 2005 and 2012, we use MDN measurements of weekly integrated precipitation depth (mm), mercury concentration (ng/L), and calculations of mercury wet deposition flux (ng/m^2) based on these measurements.[39] We define monitoring sites in the Great Lakes region broadly to include all sites in the eight states and the one province abutting the lakes (Minnesota, Wisconsin, Illinois, Indiana, Ohio, Michigan, Pennsylvania, New York, and Ontario), which are shown in Figure 1. For analysis of historical trends, and model-observation comparison, we consider only sites with >75% data for each year over this period.

The Seasonal Mann Kendall trend test (SMK) and Theil-Sen estimator of slope were used to assess the significance, sign, and magnitude of trends in monthly means of precipitation weighted concentration, precipitation depth, and wet deposition flux, across years (for both model and measurement data).[46–48] The SMK is a non-parametric test for the presence of a monotonic trend commonly used for environmental monitoring time series with seasonal variation, and the Theil-Sen estimator is a non-parametric method of estimating the slope of the linear trend.[46,47] In our analysis, each month is treated as a separate "season," yielding 12 test statistics which are then combined to yield an annual statistic.[46,48] In all statistical analyses in this work, we define the threshold for significance as $p<0.1$.

To supplement our evaluation of model performance, we also compare modelled atmospheric mercury concentrations of GEM to NADP Atmospheric Mercury Network (AMNet) measurements, aggregated as monthly means.[39] We use GEM as opposed to total gaseous mercury (TGM) as there remains uncertainty about the extent to which current methods to measure speciated mercury at these monitoring sites may underestimate gaseous oxidized mercury (GOM).[49–53] Further, measurements of GOM have been found to be affected by environmental and meteorological variables (such as O_3 and relative humidity),[53] making a quantitative, regional-scale comparison of seasonal variations and trends in GOM between measured and simulated values more difficult. We consider AMNet sites in our study region using speciation units, and so interpret reported GEM measurements as explicitly Hg(0).[49]

2.3 Chemical Transport Modelling

2.3.1 Model description

To model mercury deposition, we use the GEOS-Chem (version 10-01; http://acmg.seas.harvard.edu/geos/) coupled atmosphere-ocean-land mercury simulation, which includes a 3-D atmosphere,[54] and 2-D land and ocean modules.[55,56] Globally, we use a horizontal resolution of 4° latitude × 5°longitude, while over North America (10° to 70° latitude, -140° to -40° longitude), we also use a finer 1/2° × 2/3° resolution, using a one-way nested-grid simulation developed by Zhang et al.,[57] with boundary conditions from the global simulation. The atmosphere is modelled with 47 vertical layers in both the global and nested simulations. In the atmosphere and ocean, the model tracks inorganic mercury in two forms: elemental mercury, Hg(0), and divalent mercury, Hg(II), which in the atmosphere is modelled with equilibrium partitioning between gas and particle-bound phase based on temperature and aerosol concentration.[58] Oxidation of Hg(0) to Hg(II) in the atmosphere, and in-cloud reduction of Hg(II) to Hg(0) follow the mechanisms described in Holmes et al.,[54] with the rate for reduction scaled to NO_2 photolysis following Zhang et al.[57] (see standard code for GEOS-Chem v. 10-01). Bromine is assumed to be the primary oxidant in a two-step process.[59,60] Bromine concentrations are taken from a full-chemistry GEOS-Chem simulation described in Parrella et al.[61] Wet deposition, the primary metric of interest in this study, results from large-scale washout and rainout, and scavenging in moist convective updrafts of Hg(II), as described in Holmes et al.,[54], Amos et al.,[58] and Liu et al.[62]

2.3.2 Meteorology

In this work, GEOS-Chem mercury simulations are driven by assimilated meteorological fields from the NASA Goddard Earth Observing System, Version 5 (GEOS-5.2.0; http://gmao.gsfc.nasa.gov/GEOS/). The temporal coverage of GEOS-5.2.0 is 2004 to 2012, with a native resolution of 1/2° × 2/3°. GEOS-Chem mercury simulations using this meteorological data have been extensively compared to wet deposition and concentration measurements over the region of interest.[8,57,58] Yu et al.[63] have recently reported that the use of offline meteorological archives to drive chemical transport simulations such as GEOS-Chem may lead to vertical transport errors that bias surface concentrations of chemical tracers high, and upper troposphere concentrations low. Consequently, these simulations may underestimate the extent of global mercury transport and its contribution to Great Lakes mercury deposition. In the remainder of the text, we use the term "meteorological year" to refer to the year with which meteorological data is associated (see Table 2). All simulations are initialized with a three-year spin-up.

2.3.3 Emissions

Global anthropogenic mercury emissions are based on data from the Emission Database for Global Atmospheric Research (EDGAR) v4.tox2 inventory, which provides a time-series of spatially-resolved, speciated emissions at 0.1° × 0.1°, from 1970 to 2012.[64] This sectorally disaggregated inventory combines international activity data statistics, emissions factors, and data on control technology performance and adoption through the following equation:

$$E_{i,j,k}(y)=AD_{i,j,k}\times EF_{i,j,k}\times EOP_{i,j,k}$$

where *E* represents emissions (in mass Hg), *y* represents the year, *AD* represents activity data, *EF* represents emissions factor (mass Hg/unit of activity), *EOP* represents mercury removal from

end-of-pipe controls (%), and *i, j, k* are indices for country, sector, and technology.[64,65] Although the US National Emissions Inventory (NEI) and Canadian National Pollutant Release Inventory (NPRI) may offer better validated inventories for these specific regions (particularly with the use of test data in some sectors in recent years), the EDGAR inventory was selected for this study because: 1) its bottom up methodology allows us to treat EOP controls (both magnitude of removal and speciation) as a noisy variable in our experiments; 2) its yearly temporal resolution, using a consistent methodology, enables us to consider interannual variability in emissions as a source of noise (the NEI is released on a three year cycle). In our modelling experiments, we apply our policy scenarios to the electricity generation sector by modifying the *EOP* term, according to the technology standard policy change scenario described in Section 2.4.1 and Table 1. *AD* and *EF* are not changed. In the remainder of the text, we use the term "emissions year" to refer to the year with which activity data, emissions factors, and end-of-pipe control technology specifications are associated. For the electricity generation sector, "emissions year" corresponds to activity data and emissions factors only, as technology specifications are set in the policy scenario.

2.4 Model experiment

2.4.1 Policy scenarios

In our experiment, we model hypothetical policy change scenarios targeting the electricity generation sector, summarized in Table 1. We use a simplified policy treatment, assuming homogenous technology standards applied as a step change, to more easily diagnose the signal and noise dynamics that arise from introducing variability in policy implementation—which in our analysis is limited to variability in technology performance and resulting speciation from the sector-wide technology standard (see Section 2.4.2).

Our policy change scenarios for the electricity generation sector are based on existing policy efforts. To distinguish regional and global policy influences on the Great Lakes region, we consider NA and ROW policies separately. We apply a homogenous technology standard to the sector, requiring 100% adoption. For NA, in the pre-policy period, we assume the use of PM and SO_2 controls—specifically cold-side electrostatic precipitators (ESP) and wet flue gas desulfurization (FGD) in pulverized coal boilers. In the post-policy period, we assume a configuration of SO_2, PM, and NO_x controls with higher mercury removal—specifically, spray dry absorber (SDA), fabric filter (FF), and selective catalytic reduction (SCR). This shift is similar to the actions that some plants would undertake to comply with the Mercury and Air Toxics Standards.[17] For ROW, in the pre-policy period, we assume the use of PM controls only, in the form of ESP. In the post-policy period, we assume the use of PM and SO_2 controls, through ESP and FGD. This technology shift is similar to the actions that plants have undertaken in China to comply with air quality regulations.[21,22]

The removal fractions and speciation profiles resulting from these technology standards are listed in Table 1. These values are based on emissions testing data collected by Bullock and Johnson for the US EPA.[45] Normal distributions, truncated between 0 and 1, for removal fraction and fraction Hg(0) were fit for each configuration, with goodness of fit evaluated using the Kolomogorov-Smirnov test at 5% significance. Values shown in Table 1 represent the mean of the distribution.

Our simplified policy scenarios do not reflect the real-world complexity of the power generation sector, which is globally heterogenous and time-varying in fuel type, and plant and air pollution control technologies. Moreover, many air pollution policies targeting this sector use performance standards/emission limits or market mechanisms, that allow for some flexibility in pollution control approach, accounting for local context.[66] However, a simplified policy scenario allows us to more easily evaluate the influence of changes in specific factors (e.g., the removal efficacy of end-of-pipe controls), providing quantitative insight into the behaviour of the human-natural system.

2.4.2 Policy scenarios

We conducted seven simulations—one for model-observation comparison, and six model experiments. The policy scenarios described in Section 2.4.1 and Table 1 are applied in all model experiments (see Table 2 for a summary). The first experiment evaluates the impact of the policy change alone, while the subsequent five introduce different sources of noise—trends and variability in factors that are exogenous to the policy action. Emissions by simulation are summarized numerically in Tables S1 and S2 and visually in Figures S2 and S3.

Historical time varying emissions and meteorology (HIS). For the purposes of model-observation comparison, we conduct a simulation with historical time varying emissions (using the unmodified EDGAR inventory, and underlying data on activity, emissions factors, and end-of-pipe controls) and meteorology (GEOS-5) between 2005 and 2012.

Policy only simulation (PO). We evaluate the strength of the policy signal—the difference between pre- and post-policy period wet deposition—holding all else constant except the technology standard in the power generation sector. Meteorological year 2005 and emissions year 2005 are therefore repeated throughout the eight-year period. As shown in Figures S2 and S3, the resulting emissions are constant in each four year period, with a step change occurring between simulation years 4 and 5. For NA policy, ROW emissions remain at pre-policy levels in the post-policy period, and vice versa.

Energy and economic trends simulation (EET). We consider the effect of trends in emissions due to changes in underlying energy and economic activity, which are exogenous to the policy that targets end-of-pipe emissions controls, on the strength and significance of the policy signals in the Great Lakes region. Between 2005 and 2012, global anthropogenic emissions are estimated to have increased, due to increased activity in power generation, cement production, metals production, and artisanal and small-scale gold mining (though there is substantial uncertainty associated with this source category).[64,65,67] These global inventories indicate that industrial activity in Asia in particular was a key driver of this growth.[65,67] In North America, emissions were estimated to be relatively stable between 2005 and 2008, while a combination of macroeconomic trends and regulation that affected energy and industrial activity contributed to lower emissions overall between 2009 and 2012.[32,64,65] We use "emissions years" 2005-2012 from EDGAR, while repeating "meteorological year" 2005 throughout the eight-year period.

Product emission trend simulation (PET). The use of mercury in commercial products has been hypothesized to be an often unaccounted for source of Hg(0) to the atmosphere, with emissions peaking in the 1970s and declining since then.[68] We evaluate the impact of a large,

declining source of Hg(0) on policy signals, using product emission magnitudes and spatial distributions from Zhang et al.[8] Because Zhang et al.[8] provide inventories for 2000 and 2010, we linearly interpolate a decreasing trend in each of the geographic regions they define between 2005 and 2010, and extend this trend to 2012. Because this product emissions inventory was harmonized with a base inventory from Streets et al.,[69] it is possible that some emissions are double-counted when combined with the EDGAR inventory (for instance, from waste incineration). However, given the purpose of this simulation—to investigate the impact of a large, and declining source of Hg(0) emissions—we do not expect these inconsistencies to change our interpretation. "Emission year" and "meteorological year" 2005 are repeated throughout the eight years.

Removal variability simulation (RV). Variability in the performance of air pollution control devices can be due to variabilities in fuel characteristics and operating conditions.[22,70,71] To investigate the potential impact of such variability on the policy signal, we treat the removal fraction of each air pollution control configuration probabilistically each year. Rather than assuming a static removal fraction for each air pollution control configuration, we bootstrap a normal distribution for the population mean from the sample data from Bullock and Johnson,[45]described in Section 2.4.1, and randomly select the removal fraction for each year from this bootstrapped distribution. The distributions from Bullock and Johnson are shown in SI Figure S4, and the parameterizations for distributions of the resulting population means are listed in Table S3. Speciation is deterministic in this simulation. We hold "emission year" and "meteorological year" constant at 2005.

Speciation variability simulation (SV). The same procedure used to probabilistically generate removal fraction for each year and air pollution control configuration is applied to % Hg(0), using data from Bullock and Johnson.[45] (Note: % Hg(II) = 100% - % Hg(0)) Removal fraction is deterministic in this simulation. "Emissions year" and "meteorological year" are held constant at 2005.

Interannual meteorological variability simulation (MV). Interannual variability in meteorology—including in temperature, precipitation volume, and precipitation type—can impact mercury chemistry and transport, with implications for wet deposition.[37,41] We simulate "meteorological years" 2005-2012 using GEOS-5 data (i.e., historical variability), while holding the "emissions year" constant at 2005, resulting in an identical emissions trajectory as in the "Policy Only" case (i.e. emissions only change due to policy).

2.4.3 Statistical analysis

For each modelled grid cell in the Great Lakes region, we evaluate the magnitude and statistical significance ($p<0.1$) of the difference in wet deposition (precipitation weighted concentration, flux, precipitation) between pre- and post-policy periods using the seasonal Hodges-Lehmann (HL) estimator of difference and the seasonal Mann-Whitney-Wilcoxon (MWW) rank sum test.[47,72] The MWW rank sum test and the HL estimator are non-parametric equivalents of a two sample t-test and difference of means, modified for seasonality.[47,72] The HL estimator is the median value of all possible differences between observations from the first and second period. As in our use of the SMK trend test, each month is considered a separate season in our analysis. For the PO and MV simulations, we repeat this analysis for GEM.

3.0 Results

3.1 Observed trends

Figure 1 shows 2005-2012 trends in wet deposition, precipitation weighted concentration, and precipitation for MDN monitoring sites in the Great Lakes region, expressed as Δ% per year. We find few significant trends in wet deposition over this period: significant negative trends at two sites in Pennsylvania are on the order of 2 to 3% per year—in one case, driven by a significant decrease in concentration—and a significant positive trend at the site in Ontario is greater than 3% per year. Some non-significant decreasing trends in precipitation weighted concentration are observed downwind of major US emission sources in Ohio and Pennsylvania. Non-significant increasing trends in wet deposition around the upper lakes during this period may be due to both increasing precipitation and increases in concentration. The spatial pattern of these results is consistent with findings in Risch et al.[38] and Weiss-Penzias et al,[32] though direct comparison is difficult due to differences in statistical methods and time periods.

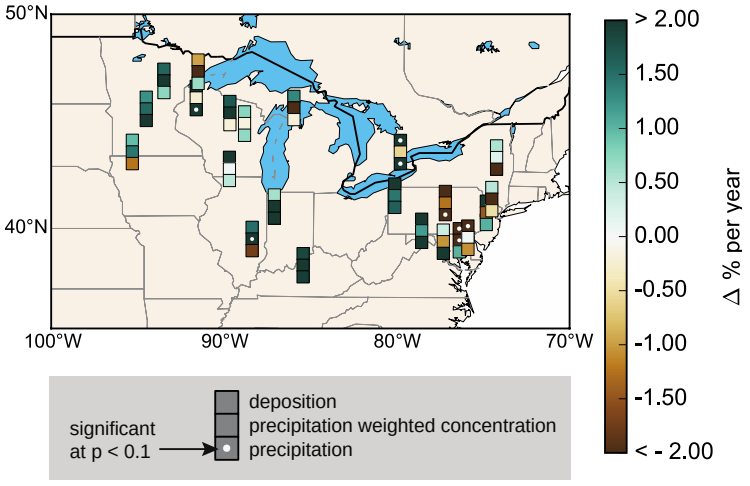


Figure 1 Observed 8 year (2005-2012) trend at MDN monitoring sites with ≥75% data availability. For each site, the trend in wet deposition (top), precipitation weighted concentration (middle), and precipitation (bottom) are shown. Trends significant at $p < 0.1$ are indicated with a dot. We evaluate significance of trends using the Seasonal Mann-Kendall trend test, and quantify the magnitude of the trend using Theil-Sen's estimator of slope.

3.2 Model evaluation

Although this study is motivated by puzzles in historical data, its goal is not to explicitly explain the magnitude and timing of historical trends. Rather, we aim to provide qualitative insight into the relative strength of drivers of variability in wet deposition that may affect trend detection. Consequently, our evaluation of model performance focuses on the ability of the model to capture the range of real-world spatial and temporal variability, and qualitative patterns of Great Lakes wet deposition.

Figure 2 shows a comparison of modelled (HIS) and observed annual wet deposition averaged from 2010-2012, when the spatial coverage of observational data is greatest,[39] and when underlying data in the EDGAR emissions inventory is most detailed.[64,65] The model reproduces

the spatial pattern of annual wet deposition, with the highest values in the Ohio River Valley. Magnitudes are underestimated in the central US region (e.g. Nebraska and Kansas), contributing to lower average modelled wet deposition at MDN sites in the depicted region of 9.4 $\mu g/m^2$ compared to the MDN average of 10.3 $\mu g/m^2$. SI Figure S7 compares modelled and observed trends for 2005-2012, for sites with > 75% data. The model predicts increasing trends South of the Great Lakes, and decreasing trends to the North and East. MDN observations indicate more sites with decreasing trends to the Southeast of the lakes, though regions with the strongest increasing trends are generally in agreement.

An aggregated time series of monthly modelled and observed values is provided in SI Figure S6. The Pearson correlation coefficient for the modelled and observed time series across the monthly site averages, *r*, is 0.41, with an individual site maximum of r=0.70 and minimum of r=0.13. Temporally, correlation between model and observations is stronger in the recent period of 2009-2012, when r=0.65, while model predicted wet deposition magnitudes are biased low between 2005-2008. The root mean squared error (RMSE) and normalized mean bias (NMB) for the aggregated data over the full period are 11.5 $\mu g/m^2$ and -4%, respectively. Lower estimates of emissions from key North American emission source categories, like coal combustion, in EDGAR compared to other inventories may contribute to this discrepancy.[65] While the model reproduces the general seasonal cycle at most sites (and broadly matches the observed between-site variability), it underestimates summertime peaks, while wintertime values tend to be larger than those observed. Similar model biases, particularly summer underestimates of wet deposition, have been reported in several modelling studies in this region.[25,33,73] Underestimates of precipitation in GEOS-5 in the Midwest, summertime prevalence of deep convective thunderstorms that more effectively scavenge upper-troposphere Hg(II) (and which are not resolved in a global-scale models like GEOS-Chem), and model underestimates of upper tropospheric Hg(II) may contribute to these wet deposition underestimates.[33,73,74] Lower snow collection efficiency, compared to rain, of MDN samplers has also been hypothesized to contribute to wintertime differences between observed and modelled values.

SI Figure S8 shows a comparison of the seasonal cycle of modelled and observed monthly mean surface GEM concentrations. The model is biased low (NMB=-9.6%, RMSE=0.17 ng/m^3), however captures key features of the seasonality at most sites. A key exception is two urban sites, where the model cannot reproduce elevated summertime concentrations.

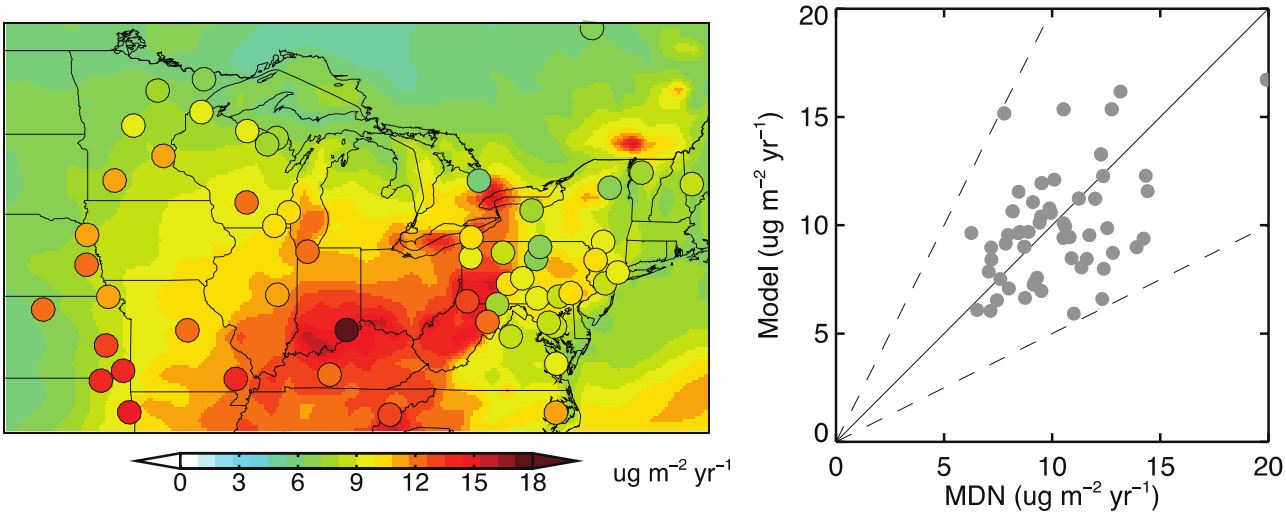


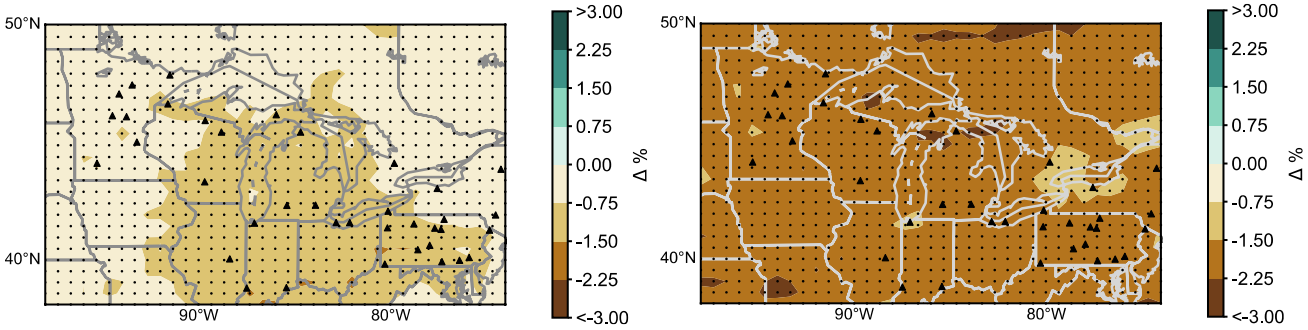
Figure 2 Comparison of modelled (background) and observed (filled circles) 2010-2012 average annual wet deposition.

3.3 Policy Only Simulation

Figure S5 maps the pre-and post-policy emissions difference resulting from the NA and ROW technology standards. For the NA policy, the 30% decrease in emissions (25 Mg/y) occurs predominantly in the Northeastern US, where many coal-based power generating units are located to the South of the Great Lakes in the Ohio River Valley. Due to the nature of the prescribed air pollution control configuration, which promotes the oxidation of Hg(0) to Hg(II) and facilitates capture of this soluble form of mercury,[71] these reductions are predominantly in the form of Hg(0) rather than Hg(II) (seen in Figure S2). Because Hg(0) is long-lived in the atmosphere, with an estimated lifetime of 0.5-1 year compared to a lifetime of days to weeks for Hg(II),[75,76] these speciation differences have important implications for transport.[77,78] The 18% emissions decrease (276 Mg/y) under ROW policy is predominantly in the form of Hg(II), as the adoption of FGD in addition to PM control increases the removal of gaseous oxidized mercury.[71] These decreases are largest over East and South Asia, and Western Europe.

Simulated deposition differences in the PO simulation are shown in Figure 3. Figure S9 plots differences in precipitation weighted concentration, with numerical results at MDN sites summarized in Table S5. Note that here, Δs refer to the pre- and post-policy period difference, rather than a percent change per year. Table 3 gives a numerical summary of differences sampled at MDN site locations (that were active at any time during this period) for this and all subsequent simulations. Holding all else constant, the NA policy results in statistically significant decreases in deposition at all simulated grid cells in the region ranging from -0.3 to -2.0%. The regions with strongest decreases trace the footprint of local power generation emission sources in the Ohio River Valley and the western edge of Lake Erie. The average difference in deposition at MDN sites due to policy is -0.85%. The relative spatial homogeneity of the policy difference is due to the speciation of the modelled emissions decrease (predominantly elemental mercury), resulting in a more diffuse impact on deposition.

The ROW policy also leads to statistically significant decreases in simulated wet deposition at all grid cells ranging from -0.8 to -2.7%, with an MDN site average of -1.78%. The magnitude of this decrease is even more spatially homogenous than for the NA policy, reflecting the influence of ROW emission decreases on global background concentrations of mercury. The effect of ROW policy, in terms of Δ% deposition is therefore weakest where the contribution of local emission sources to deposition is strongest—for instance, around metal smelting facilities near Lakes Ontario, Erie, and Michigan.



(a) NA Signal **(b)** ROW Signal

Figure 3 Change in deposition (%) between pre-policy and post-policy period, for 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.

3.4 Emission Trend Simulations

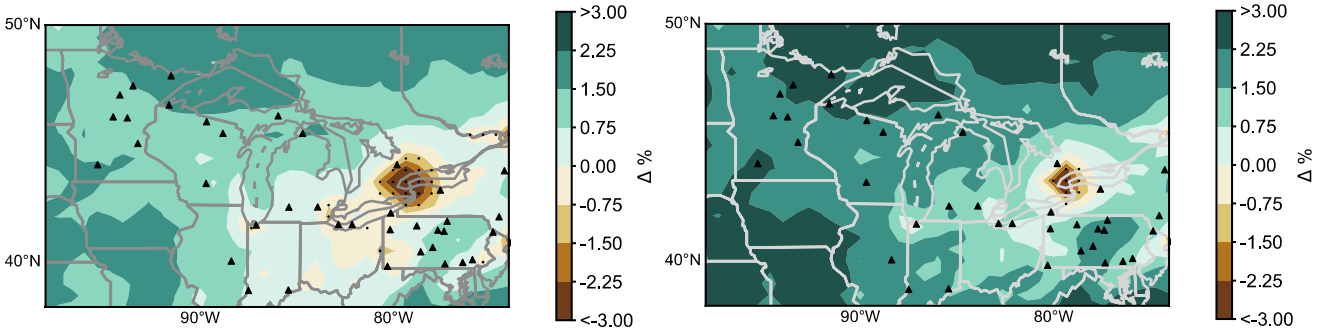
We consider two categories of emissions-related trends exogenous to the policy that may act as "noise" in detecting the policy signal in wet deposition: energy and economic activity trends (EET) that lead to globally increasing emissions, and product emission trends (PET) leading to globally decreasing emissions.

In our PET simulations, we add a linearly decreasing source of Hg(0) emissions from commercial products. These products result in an additional 488 Mg of Hg(0) in 2005, 15% of which is located in NA. Global emissions from products decrease by 150 Mg over the eight-year period.

In our EET simulation with NA policy, ROW emissions monotonically increase between Simulation Year 1 and Simulation Year 8 from 1520 to 1976 Mg/y (see Table S1). In NA, trends in energy and economic activity drive emissions decreases overall between 2005 and 2012 (with a large drop between 2008 and 2009 due to the economic recession), leading to a larger emissions gap between pre- and post-policy periods, compared to the PO simulation. With the ROW policy (Table S2), total emissions decrease sharply between 2008 and 2009 due to decreases in Hg(II) emissions from the technology standard, however, by 2012, total emissions exceed the highest emissions year in the pre-policy period (1675 Mg/y in 2012 compared to 1607 Mg/y in 2008). For NA, even without policy, reduced activity in energy and other sectors leads to a decrease in emissions in the post-ROW-policy period.

In the EET simulation, the area over which a statistically significant decrease is detected between periods is limited to the eastern portion of the Great Lakes region. The area surrounding Lake Superior is highly influenced by increasing global emissions, as seen in Figure 4, showing the Δ% in wet deposition. Figure 4 also highlights the large impact of local emission sources on Great Lakes deposition: though total global emissions are monotonically increasing in the NA policy simulation, we find statistically significant decreases in Indiana, Michigan, Ohio, Pennsylvania, New York, and Southern Ontario (an average -2.23% decrease at the 3 MDN sites with significant differences) driven by NA emission reductions. Comparison between the ROW and NA plots in Figure 4 suggests that the large differences simulated in Southern Ontario and upstate New York under both NA and ROW policies are due to decreasing activity in metals production (rather than the simulated NA power generation policy) that substantially reduce Hg(II) emissions from iron production facilities adjacent to Lake Ontario. The additional benefit of the NA policy targeting the power generation sector occurs in Indiana, Ohio, and the Northern areas of Kentucky and West Virginia (where fewer MDN sites are located). Results for precipitation weighted concentration are shown in Figure S10 and Table S5.

In the PET simulations, we see statistically significant decreases in wet deposition and precipitation weighted concentration at all grid cells in the modelled region between the pre- and post-policy period, exceeding the magnitude of the PO simulation differences (Figures S11 and S12). For the NA policy, differences range from -0.7 to -2.5%, while for the ROW policy, differences range from -0.8 to -2.2%. The modelled differences are more reflective of the trend in commercial product emissions, which for the ROW case, represents a less aggressive % decrease than the Policy Only simulation at some locations. This influence is also seen in the increased spatial homogeneity of differences in the NA policy simulation. The smaller fluctuations in emissions due to our simulated policy are harder to discern against a higher global background of atmospheric mercury, except in the immediate vicinity of local emissions sources.



(a) NA Signal **(b)** ROW Signal

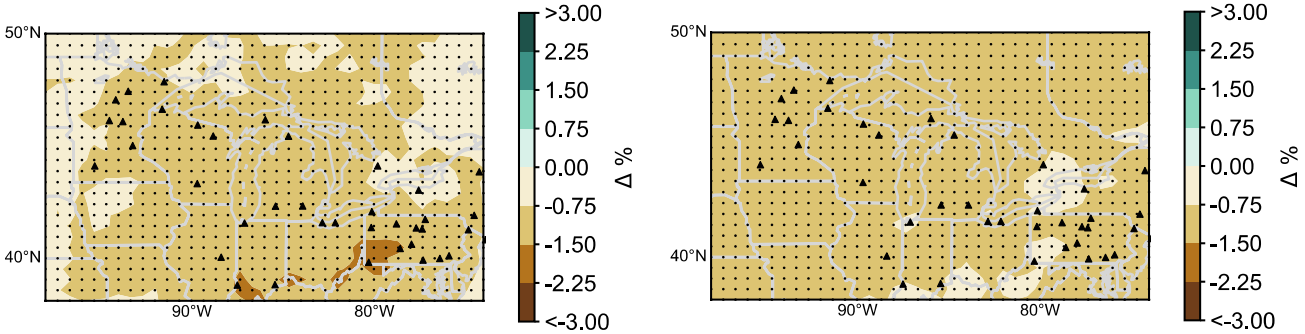
Figure 4 Change in wet deposition (%) between pre-policy and post-policy period, for Energy and Economic Trends simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.

3.5 Air Pollution Control Variability Simulations

We conduct two simulations that explore how variability in the performance of air pollution control devices in the regulated sector affects the pre- and post-policy wet deposition difference: one treats the removal fraction of pollution control as a probabilistic variable while holding speciation constant (RV), while the other considers the fraction of flue gas emissions that are in

the form of Hg(0) probabilistically, while holding removal fraction constant (SV). For removal fraction, this variability is primarily due to ESP and ESP+FGD, while for fraction Hg(0), SDA+FF+SCR is the primary driver (see Figure S4). The resulting variability in emissions *within* pre- and post-policy periods can contribute to increases or decreases in median difference *between* the periods compared to the PO simulation, depending on chance. For example, in our NA policy simulation, Removal Variability resulted in a larger difference compared to the PO simulation, while in our ROW policy simulation, Removal Variability resulted in a smaller difference (see Tables S1 and S2). These simulations represent just one realization of a probabilistic phenomenon. As an example of the potential magnitude of real-world interannual variability in emissions, we include an analysis and comparison of the US EPA's Toxics Release Inventory and National Emissions Inventory in the Great Lakes region in the Supplemental Information (Section S4).

Figure 5 shows resulting wet deposition differences for the RV simulation. Results for RV concentration and SV deposition and concentration are shown in Figures S13-S16. For both policies, introducing removal variability does not change the sign of the pre- and post-policy differences, or their statistical significance, but does affect their magnitude. For NA policy, removal fraction variability slightly increases the modelled difference at MDN sites to -1.04% (compared to -0.85% in the PO simulation, an increase of 22%), while for the ROW policy, removal variability reduces the difference at MDN sites to -0.85% (a 52% decrease from -1.78% in the PO simulation). Speciation variability has a larger dampening effect than removal variability on the NA policy signal (the difference at MDN sites is -0.15%, which is 82% smaller than in the PO simulation), while the opposite is true for the ROW policy signal (Figure S14). This result further emphasizes the importance of local emissions of Hg(II) to wet deposition in the Great Lakes region—even small variations in divalent mercury can weaken the effect of overall policy-related emissions decreases. In contrast, ROW policy affects Great Lakes wet deposition primarily through contributions to total atmospheric burden.



(a) NA Signal **(b)** ROW Signal

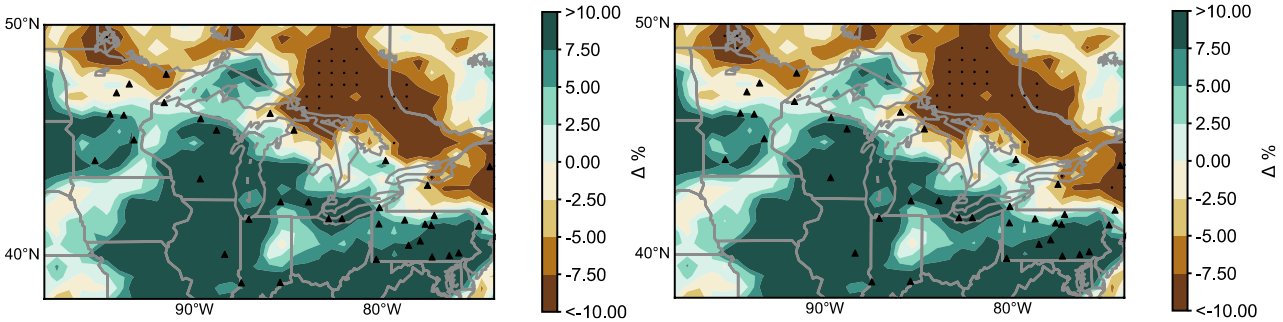
Figure 5 Change in wet deposition (%) between pre-policy and post-policy period, for removal fraction variability simulation. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.

3.6 Interannual Meteorological Variability Simulation

We simulate the policy change while including the historical interannual meteorological variability from 2005-2012. Figures 6, S16, and 7 show the pre-and post-policy period differences in wet deposition, concentration, and precipitation, respectively. Similar to the

Energy and Economic Trends simulation, the resulting pattern of deposition differences has regions of positive and negative difference, ranging from < -20 to > 20%. The resulting spread in values is much larger than for any other simulation (mean absolute deviation of 5.9 compared to 0.2 in the PO simulation, and 0.8 in the EET simulation). The number of grid cells showing statistically significant differences also decreases dramatically. These significant decreases are predominantly at higher latitudes and include areas in Ontario, Quebec, New York, and Northern Minnesota. Large, but not significant, increases in deposition are simulated South of the Great Lakes.

The similarity of the results for the NA and ROW policy simulations with meteorological variability indicate that meteorological influence is larger than that of emissions—at least for the magnitude of emissions changes considered here. For instance, comparison of Figure 6, showing deposition changes, and Figure 7, showing precipitation changes, demonstrates that variability in precipitation volume alone can account for much of the simulated pattern in deposition change. Moreover, that the spatial pattern of deposition change in this simulation captures many of the features in Figure 1, our analysis of 2005-2012 trends at MDN sites, speaks to the extent to which interannual meteorological variability is a driving force in observed wet deposition.



(a) NA Signal **(b)** ROW Signal

Figure 6 Change in wet deposition (%) 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.

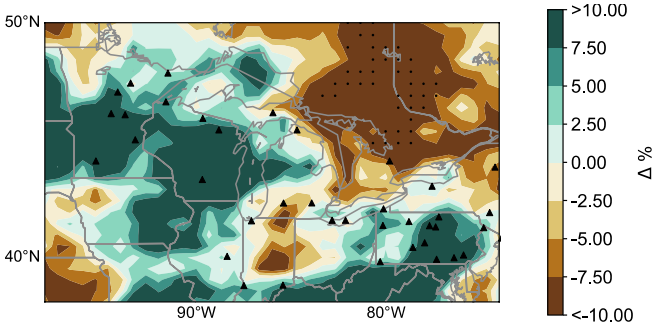


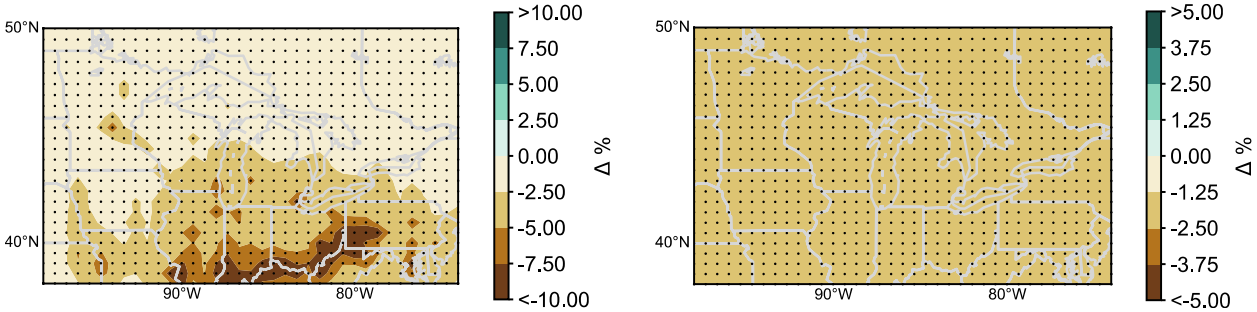
Figure 7 Change in precipitation volume (%) between pre-policy and post-policy period, for interannual meteorological variability simulation. Note the larger color bar range of -10 to 10%, compared to the other plots. Grid cells with a significant ($p<0.1$) change are indicated with a dot. For reference, locations of monitoring stations are indicated with triangles.

Table 3 Step change in Hg wet deposition (Δ%) 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 Only	Removal Variability	Speciation Variability	Meteorological Variability	Energy and Economic Trends	Product Trends
NA	all sites	-0.85%	-1.04%	-0.15%	6.79%	0.48%	-1.68%
	sig. sites	-0.85% (100%)	-1.04% (100%)	- (0%)	-9.80% (2%)	-2.23% (7%)	-1.68% (100%)
ROW	all sites	-1.78%	-0.85%	-0.94%	6.64%	1.34%	-1.81%
	sig. sites	-1.78% (100%)	-0.85% (100%)	-0.94% (100%)	-10.27% (2%)	-1.93% (5%)	-1.81% (100%)

3.7 Changes in Atmospheric Concentrations

To evaluate whether atmospheric mercury concentrations are equally sensitive to these sources of noise, we analyze surface GEM in the PO (policy signal) and MV (largest source of noise in wet deposition experiments) simulations. Figure 8 shows the simulated pre- and post-policy change in surface GEM concentrations for the NA and ROW policies (note the different scales). Holding all else constant, the NA policy leads to statistically significant decreases at all grid cells in the region of interest, ranging from -19% to -0.9%. Similar to the spatial pattern for wet deposition differences, the largest decreases occur downwind of local power generation emissions sources, though the maximum strength of the GEM signal is close to ten times as large as that for wet deposition. This more spatially concentrated effect is due to the nature of the air pollution control technology adopted under the NA policy—as noted previously, the 30% NA emissions decrease is predominantly as Hg(0). In contrast, the ROW policy results in very homogenous decreases in GEM concentrations across the study region, ranging from -2.0% to -1.4%, as the 18% decrease in ROW emissions has a more diffuse effect on the global background GEM concentration.

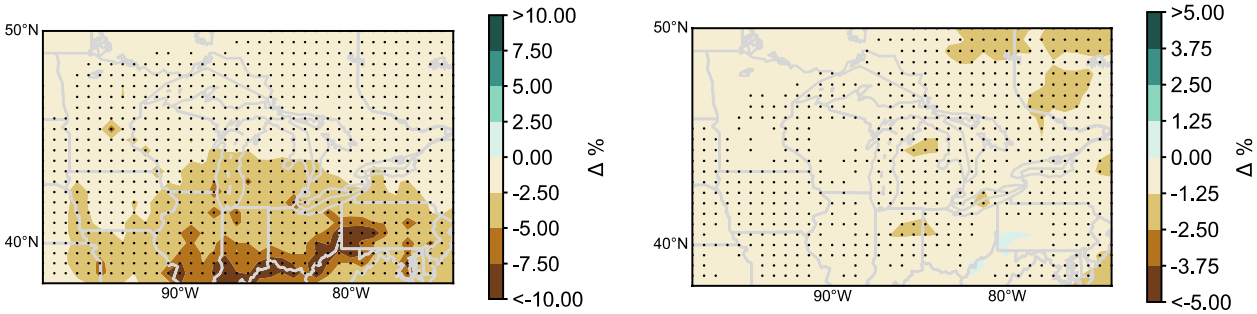


(a) NA Signal **(b)** ROW Signal

Figure 8 Change in surface GEM concentration (%) between pre-policy and post-policy period, for Policy Only simulation. Note the larger color bar range of -10 to 10% for NA Signal (a). Grid cells with a significant ($p<0.1$) change are indicated with a dot.

Given the large influence of interannual meteorological variability on wet deposition described in Section 3.6, we also evaluate pre- and post-policy GEM concentration differences in the MV

simulation (Figure 9).For NA, the overall spatial pattern of differences remains the same as in the PO simulation, though the strength of the signal is reduced (range: -17.9% to -0.03%). Importantly, the differences in the Northwest of the study region are no longer significant. For the ROW policy, the interannual meteorological variability increases the range in concentration differences (-1.5% to +0.6%), compared to the PO case, and these differences are no longer significant across the domain. In contrast to the wet deposition results, meteorological variability has a noticeably larger impact on the ROW signal, compared to the NA signal. This result suggests that the meteorological drivers of surface concentration variability are less local (e.g., precipitation frequency and volume) and more connected with global circulation.



(a) NA Signal **(b)** ROW Signal

Figure 9 Change in surface GEM concentration (%) between pre-policy and post-policy period, for interannual meteorological variability simulation. Note the larger color bar range of -10 to 10% for NA Signal (a). Grid cells with a significant ($p<0.1$) change are indicated with a dot.

4.0 Discussion and Implications for Policy Monitoring and Evaluation

Reported trends (here and elsewhere) present a puzzle for connecting changes in anthropogenic mercury emissions and changes in wet deposition measurements. Our analysis of trends in measured mercury wet deposition between 2005 and 2012 is consistent with findings in the literature.[18,35,38,79] Collectively, these analyses suggest that the large (~50%) declines in regional (North American) emissions that occurred during this period were not statistically detectable in wet deposition measurements in the Great Lakes region as a whole, though statistically significant declines were observed in some select locations. However, negative trends in other Great Lakes media (air, water, fish) have been more consistently reported for this approximate period.[32,80–82] For instance, Zhou et al. find significant declines in predatory fish concentrations when data for all lakes but Erie are combined (though some lakes show increases or stable concentrations in the 2010s, following steep declines between 2005 and 2010).[80] Similar patterns of decreases and then stabilization or increase have been reported for GEM concentrations (though certain sites, particularly in the Northeast, show steady and statistically significant declines),[32,81] and other analyses of fish concentrations.[83]Although our analysis is not designed to explain these historical trends explicitly, it does aim to use modelling experiments to generate insight into the relative influence of factors that mediate the connection between anthropogenic emissions changes and wet deposition in this region.

Our modelling results highlight the potential challenges of detecting statistically significant policy-related changes in Great Lakes wet deposition on a sub-decadal scale, given the magnitudes of realistic emissions changes and sources of confounding “noise,” exogenous to the

policy change. Our simplified policy scenario for the electricity generation sector results in 30% (25 Mg) and 18% (276 Mg) step decreases in emissions from NA and ROW, respectively, over successive four-year periods. These emissions decreases translate into pre- vs. post-policy deposition decreases ranging from -0.3 to -2.0% and -0.8 to -2.7% in the Great Lakes region, holding all else constant. Notably, differences of this magnitude may be within the range of measurement uncertainty, which can limit our confidence in detecting these changes[38]: Wetherbee et al. found that sampling measurements could resolve wet deposition differences of ±8.5%, suggesting that many of the statistically significant wet deposition differences simulated in this study could be below the limits of measurement uncertainty.[40] The introduction of global trends in emissions with realistic magnitudes—based on energy and economic activity (increasing trend ~ +50 Mg/y) and commercial product emissions (decreasing trend ~ -20 Mg/y)—reduces the areas where the policy signal can be detected to the immediate vicinity of targeted emissions sources, as these global trends dominate elsewhere. We find that the introduction of variability in emissions and meteorology can also obscure policy signals. In our simulations with variability in the magnitude and speciation of emissions, based on air pollution control test data, even a relatively small amount of year to year variability within pre- and post-policy periods could shift the magnitude of the simulated deposition difference between periods at MDN sites compared to the PO simulation—by up to 80% for the NA policy, and up to 50% for the ROW policy. Even more influential, however, is interannual meteorological variability, which drove deposition differences of more than ±20% in some areas of the Great Lakes in our simulation, greatly exceeding the changes associated with both the regional and global modelled policy. In the real world, these sources of "noise" that we have treated separately here, from exogenous trends in emissions to variability in technical or natural systems, operate simultaneously, further complicating the task of attributing observed changes in deposition to specific policy-action.

Our emissions trend results point to the continued importance of North American policy for the Great Lakes region, even in the face of potentially increasing global background concentrations of mercury. As North American emissions represent a smaller fraction of the anthropogenic total with continued emissions growth elsewhere, global emissions may have a larger impact on regional wet deposition.[33] However, the results from our energy and economic trends simulation highlight the extent to which some areas of the Great Lakes region are influenced by local/regional sources, supporting results from monitoring campaigns.[36] The persistence of these areas is perhaps surprising: in our simulation of NA policy with global energy and economic trends, even though global emissions increases far outweigh regional policy-related emissions decreases (~+400 Mg vs. ~-30 Mg), statistically significant wet deposition decreases before and after policy on the order of -1% can still be detected in Michigan and Ohio. Our simulations indicate that strategic location of monitoring sites near emissions sources targeted by policy may compensate for noise from exogenous trends in emissions. For communities living in the Great Lakes then, where coal combustion, metals production, and incineration facilities are located, there remain opportunities to build on past progress in local/regional emissions decreases, to achieve further reductions in locally-driven wet deposition.

Another key finding from our work is that variability in emissions—potentially due to stochastic processes in social and technical systems—can greatly attenuate our ability to detect statistically significant trends or differences in wet deposition at monitoring sites. The large epistemic

uncertainties—that is, uncertainty due to imperfect knowledge[84–86]—in anthropogenic emissions inventories (in the range of ±30%) are widely acknowledged to be a challenge for mercury modelling, monitoring, and policy evaluation.[14] However, our simulations demonstrate that even if "true" emissions values are known, year-to-year variability in these emissions—in our simulations, driven by variability in air pollution control technology performance,[22,70,71] but potentially also from other sources, like fluctuations in economic activity—can dampen a policy effect. Because they are labor-intensive to produce, many emissions inventories are released at multi-year intervals, with users linearly interpolating between these years. However, these assumed linear changes between data points may elide true interannual variability, resulting in larger and more statistically significant predicted effects in environmental concentrations and fluxes than can be actually observed. In the absence of continuous emissions monitoring for mercury (which is now required in the US, but not Canada), there may be a tradeoff between ensuring more accurate point estimates (i.e. reducing epistemic uncertainty), and better capturing temporal variability (i.e. quantifying aleatory uncertainty) (see Ambrose et al.[87] for a comparison on TRI and NEI against plume measurements from six power plants). Our analysis indicates that both efforts are relevant for interpreting monitoring data.

These findings on emissions variability also have implications for chemical transport modelling. It is important to note that our simulations represent only single realizations of this emissions variability—these results therefore speak only to the ability to detect statistically significant differences, rather than quantify the full distribution of these differences. Although probabilistic emissions inventories for mercury have been developed (e.g., Wu et al.[70], Zhao et al.,[22] and Zhang et al.[71]), the computational resource intensity of Eulerian chemical transport modelling can be prohibitive to fully-coupled emissions-chemistry probabilistic simulation. The application of computationally efficient means to quantify the resulting uncertainty in wet deposition due to emissions variability—for instance, response surface modelling (e.g., Ashok et al.[88]), adjoint or other sensitivity methods (e.g., Sandu et al.,[89] Henze et al.[90]), and polynomial chaos expansion (e.g., Thackray et al.[91])—would be a valuable next step.

Our results emphasize the large role of meteorology in explaining spatial and temporal variability in wet deposition in the Great Lakes region on a sub-decadal scale, particularly in comparison to anthropogenic emissions. Similar to studies exploring anthropogenic signal detection with respect to climate change,[92] and O_3,[93,94] these results indicate that distinguishing policy signals over meteorological variability in an 8 year observation record requires substantially larger emissions decreases than those modelled here—or alternatively, distinguishing policy signals of the size modelled here requires a substantially longer observation record (for example, multi-decadal time scales considered in Zhang et al.,[8] Risch and Kenski,[18] and Zhou et al.[81]). Future work addressing this topic can further clarify the mechanisms through which meteorology drives wet deposition variability, on an interannual and decadal scale. For instance, in addition to precipitation volume, Shah et al. find that meteorological processes affecting oxidation of global pools of Hg(0) in the mid and upper troposphere explain spatial variability in MDN,[41] and Mao et al. highlight the role of decadal scale variability in circulation patterns.[42]

Compared to wet deposition, we find that simulated regional (NA) policy-related emissions decreases translate more strongly into changes in surface GEM concentrations in the Great Lakes

(-19% to -1%), and that these changes are more robust (in terms of magnitude, significance, and spatial pattern) to sources of "noise" like interannual meteorological variability. This finding is true, to a lesser extent, for the simulated global (ROW) policy. These results suggest that surface GEM concentrations may be a less noisy indicator for policy detection, particularly for local/regional changes in Hg(0). In addition, because ambient concentration is linked to dry deposition flux, atmospheric concentration can inform estimates of this significant pathway for mercury loading to the Great Lakes watershed.[28,95] (Indeed, decreases in ambient concentrations and dry deposition may help explain observed trends in fish concentrations, even when wet deposition trends have been inconsistent.) However, in the context of policy evaluation, several additional factors may need to be considered: wet deposition measurements can be less costly to deploy, maintain, and calibrate, and have larger (temporally and spatially) existing monitoring records which may facilitate the establishment of a baseline. Measurement uncertainty is an additional consideration: although our results suggest that larger signals are likely to be detected in GEM, these may in some cases be partially offset by larger measurement uncertainty (on the order of 10-20% for GEM,[96] and 10% for wet deposition[40]). Further, the generalizability of this result to other policy efforts that more strongly target Hg(II) (which would be the case in many non-North American regions[13,97]) is an important topic for future research. Nevertheless, our analysis indicates that atmospheric concentrations are an informative metric for attributing environmental changes to policy action.

In this work, we have evaluated several factors hypothesized in the literature to affect the translation of emissions mitigation policy into wet deposition changes, clarifying the nature and potential magnitude of their influence in the Great Lakes region in particular; however, there remain additional factors that merit further investigation. The results from our speciation variability simulation, and the large impact of this variability on the detection of regional policy in the Great Lakes region, suggest that a better understanding of mercury's atmospheric redox chemistry,[98–100] and potential meteorological and climatological drivers of its variability,[101] can aid in the interpretation of monitoring data and attribution of global vs. local/regional policy signals. While we focus on atmospheric emissions, trends and variabilities in discharges to terrestrial and aquatic systems may have important effects as well due to mercury biogeochemical cycling.[102] Finally, the endpoint of our analysis is atmospheric inputs into the Great Lakes ecosystem, yet the ultimate goal of much mercury mitigation policy is to prevent dietary mercury exposure from fish consumption.[103] Understanding sources of "noise" in the translation of decreases in atmospheric inputs of mercury into changes in fish tissue concentration, and ultimately human exposure, is therefore a critical next step in this line of inquiry. Given the complexities of mercury biogeochemical cycling, a full suite of metrics for policy effectiveness, ranging from upstream indicators based on emissions,[104] to intermediate indicators, such as wet deposition and atmospheric concentrations,[105] to human endpoints,[103] should be considered.

This work speaks to the severity of the signal-to-noise challenges for mercury monitoring in the Great Lakes, and provides support for taking them seriously in the design and evaluation of mercury policy. Our simulations illustrate the wide variety of wet deposition outcomes that could be consistent with policy adoption, given the influence of "noise." These results suggest that failing to see a decrease in wet deposition—for instance, in our interannual meteorological variability simulation or energy and economic trends simulation—does not indicate a failure in

implementation of policy (indeed, our simulations assume 100% compliance). However, although all of our simulated deposition outcomes are consistent with successful policy implementation, in areas where wet deposition is a large component of mercury loading, they may not all be consistent with successful policy outcomes. All things being equal, policy implementation will avoid increases in mercury wet deposition, however if the goal of policy is to reduce mercury inputs to vulnerable ecosystems within a decade—and ultimately, human exposure—further attention to the magnitude of noise, and how to design policy signals that overcome it, is necessary.

Conflicts of Interest

There are no conflicts to declare.

Acknowledgments

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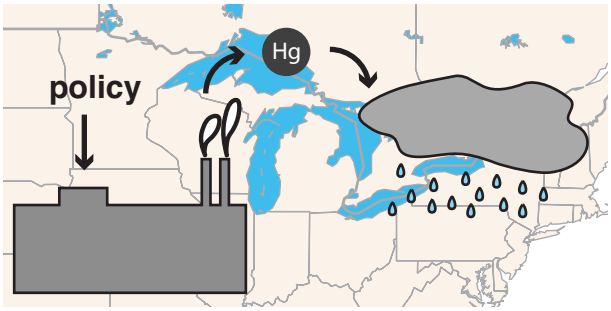
Table of Contents Entry



Challenges for detecting sub-decadal policy-related changes in mercury wet deposition in the Great Lakes are identified through modelling