

Controlling Factors of Long-Term Trends in Mercury Wet Deposition and Precipitation Concentrations at Huntington Wildlife Forest

Final Report

September 2016

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Controlling Factors of Long-Term Trends in Mercury Wet Deposition and Precipitation Concentrations at Huntington Wildlife Forest

Final report

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Preferred Citation

NYSERDA. 2016. "Wet Deposition and Precipitation Concentrations at Huntington Wildlife Forest" NYSERDA Report 16-29. Prepared by Huiting Mau (State University of New York College of Environmental Science and Forestry, Syracuse NY). www.nyserda.ny.gov/publications

Abstract

As a toxic pollutant, mercury (Hg) is concerned due to the neurotoxic human health effects of its methylated form in aquatic environment (Mason et al., 2006; Miller et al., 2007; Rolfhus et al., 2003), and has been a critical environmental concern for human and wildlife in New York State, as well as other regions in the U.S. Atmospheric Hg deposition is the most important Hg input to water bodies. Observations from the Mercury Deposition Network (MDN) at Huntington Wildlife Forest (HWF) suggested that a significant decline in mercury (Hg) concentrations in precipitation was linked to Hg emission decreases in the U.S., especially in the Northeast and Midwest, and yet Hg wet deposition has remained fairly constant over the past two decades. It is critical to characterize the long term trends in atmospheric Hg wet and dry deposition and understand the factors controlling such trends. The present study aimed to investigate the climatic and anthropogenic factors that influenced the decadal variation in Hg wet deposition in Upstate New York. To achieve this, measurement data were utilized to identify the long term trend in Hg wet deposition and the meteorological parameters controlling Hg wet deposition during 2000–2015. The Community Multiscale Air Quality (CMAQ) model was employed coupled with our own state-of-the-art Hg and halogen chemistry mechanisms constrained with global model output. Sensitivity simulations were conducted to quantify the contributions to Hg wet deposition from changing meteorological conditions and anthropogenic emissions reductions in and outside NYS.

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1 Project Objectives

The objectives of this project were to:

- Identify mercury trends.
- Investigate meteorological impacts on Hg wet deposition at HWF.
- Quantify in- and out-of-state source contributions to Hg wet deposition at HWF.

2 Methods

2.1 Observational data

Observations of bi-weekly Hg wet deposition flux, precipitation, and Hg concentration in precipitation at HWF (Figure 1) were obtained from NADP MDN. Ambient speciated Hg concentrations were from Atmospheric Mercury Network (AMNet), and data of meteorological factors (e.g., wind speed, wind direction, North Atlantic Oscillation (NAO) index) were obtained from the Clean Air Status and Trends Network (CASTNet), and the National Oceanic and Atmospheric Administration (NOAA). These data were used to characterize trends in Hg wet deposition and to evaluate model simulations.

Figure 1. Study site



2.2 Atmospheric Hg model

The CMAQ Modeling system (Byun and Schere, 2006) with the mercury component (denoted as default CMAQ-Hg) (Bullock Jr. and Brehme, 2002) and our improved CMAQ-Hg were used. Our simulations were focused on the warm season, i.e., spring and summer months from March to August, since Hg wet deposition flux was strongly correlated with precipitation in spring and summer and hence scavenging of Hg is most efficient and GOM production is most active during those months. Domain 1 was set to include the northeastern U.S. at 12 km x 12 km spatial resolution (Figure 2).

Figure 2. Domain 1 is the CMAQ modeling domain. The "GEOS-Chem Domain" indicates the one with GEOS-chem output to be used as CMAQ initial and boundary conditions



The default CMAQ-Hg modeling system does not capture diurnal – seasonal variations in ambient Hg (Holloway et al., 2012). Our improved CMAQ-Hg includes three major modifications:

- 1. Chemical mechanisms: Our most up-to-date, detailed Hg and halogen chemical mechanisms (Ye et al., 2016) were implemented.
- 2. Constraining Br: Br is one of the most important oxidants in GEM oxidation, and yet Br is absent in the default CMAQ-Hg model, and its simulation is generally missing in current models. In our improved version, not only Br chemistry was implemented, but also BrO mixing ratios were constrained using observed BrO vertical profiles from Volkamer et al. (2015).
- 3. Boundary (BC) and initial conditions (IC): The IC and BC in default CMAQ-Hg were timeindependent. In this improved CMAQ-Hg, monthly GEOS-Chem global model output (Zhang and Jaeglé, 2013; Zhang et al., 2012) for the U.S. domain was used as BC and IC.

The emission input data for CMAQ were preprocessed by the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (Coats, 1996) using National Emission Inventory (NEI) modeling platform version 2005, Hg version. Meteorological input data were obtained from the United States Environmental Protection Agency (EPA), which were simulated using The Weather Research and Forecasting (WRF) Model (information available online at <u>http://www.wrf-model.org/index.php</u>. Here are the simulations conducted in this study:

- 1. S_d (default CMAQ-Hg scenario): using the default CMAQ-Hg model, with its default IC/BC, 2010 meteorological data, NEI 2011 emission inventory.
- 2. S₀ (base scenario): using our improved CMAQ-Hg model, IC/BC from GEOS-Chem output, 2010 meteorological data, NEI 2011 emission inventory.
- 3. S_{ei} (NYS emission sensitivity scenario): Same as S_0 , but assuming no anthropogenic Hg emission inside NYS.

3 **Results and Discussions**

3.1 General Characteristics and long term trends of Hg wet deposition and Hg concentrations in wet deposition

Bi-weekly precipitation, annual volume weighted mean (VWM) Hg concentrations and total wet deposition at HWF during 2000–2014 are shown in Figure 3. Three key results are: 1) The mean VWM Hg concentration for 2000–2014 was 5.89 ± 0.47 ng L⁻¹, the mean annual Hg wet deposition was $6.84 \pm 0.67 \mu \text{g m}^{-2} \text{ yr}^{-1}$, and the mean annual precipitation amount was 1.16 ± 0.08 m; 2) Annual VWM Hg concentrations declined slightly at a rate of 0.12 ng L⁻¹ yr⁻¹ from 2000 to 2014 (r² = 0.34, *p* = 0.03); 3) No significant trends were found in annual Hg wet deposition and precipitation during 2000–2014.





Annual cycles of Hg wet deposition and VWM Hg concentrations showed annual maximums in summer and minimums in winter (Figure 4). For precipitation, there were three peaks occurring in May–June, October, and December.



Figure 4. Annual cycles of Hg wet deposition (HgDep in black), VWM Hg concentrations (HgConc in red) and precipitation (SUBPPT in blue) averaged for the period of January 2000 – May 2014

In examining the data by season, the interannual variations of Hg wet deposition, VWM Hg concentrations, and precipitation varied for different seasons (Figure 5). In spring and summer, significant correlations ($r^2 = 0.89$, p < 0.0001 for spring; $r^2 = 0.58$, p = 0.002 for summer) were found between Hg wet deposition and precipitation. In fall and winter, significant correlations ($r^2 = 0.61$, p = 0.0009 for fall; $r^2 = 0.33$, p = 0.026 for winter) were found between Hg wet deposition and VWM Hg concentrations. This implies different predominant mechanisms for Hg wet deposition in different seasons.





3.2 CMAQ Model evaluation

Our improved CMAQ-Hg simulated ambient Hg concentrations and Hg wet deposition much better than the default CMAQ-Hg as suggested by comparisons with observations. Moreover, significant diurnal and seasonal variation of GEM was captured as opposed to the generally flat patterns simulated using the default model. More detailed model evaluation results were provided as follows.

3.2.1 Ambient Hg concentrations

Simulated ambient speciated Hg mixing ratios were evaluated using measurements of GEM, GOM, and PBM at HWF for March–November 2010. Four statistical performance measures (Chang and Hanna, 2004; Hanna, 1988; Hanna et al., 1991, 1993), including model bias, fractional bias, normalized mean square error (NMSE), and the partition of NMSE due to systematic errors (NMSEs) were used to quantify the model performance. These statistical measures (Table 1) showed significant improvement of GEM, GOM, and PBM simulations using the improved CMAQ-Hg, especially for GOM and PBM simulations.

In addition to HWF, ambient Hg observational data were available at Thompson Farm (TF) in New Hampshire, Cannan Valley Institute in West Virginia (WV), Brigantine in New Jersey (NJ), Bronx (BX), and Rochester (RC) within the model domain. The first three sites are rural, BX urban, and RC sub-urban. Simulations of GEM at these six sites using both default and improved CMAQ-Hg were compared with observations at these six sites (Figure 6). The new model simulations showed significantly enhanced diurnal variation and seasonal variation of GEM. These enhancements were due to the addition of Br chemistry and the use of GEOS-Chem IC and BC.

Species	Scenarios	Model bias	Fractional bias	NMSE	NMSE _s
GEM (ppgy)	Default	-26.67	0.18	6.03%	3.26%
GEM (ppqv)	Improved	-20.78	0.13	3.22%	1.62%
COM (nngu)	Default	1.58	-1.41	654%	393%
GOM (ppqv)	Improved	0.10	-0.39	295%	16%
DDM $(n\alpha/m^3)$	Default	29.64	-1.52	949%	549%
r Divi (lig/lil ⁻)	Improved	-0.11	0.02	53%	0.04%

Table 1. Model bias index for the simulations using Default and Improved CMAQ-Hg at HWF

Figure 6. Observed (blue dots) and simulated GEM mixing ratios using default CMAQ-Hg (Simulation_new in red line) and improved CMAQ-Hg (Simulation_old in black line) at 6 sites: a) HWF; b) TF; c) WV; d) NJ; e) BX; f) RC



3.2.2 Hg wet deposition

Within the modeling domain, Hg wet deposition observational data at 31 NADP MDN sites were used for model evaluation over March–November 2010 (Figure 7). As a result, the simulation of Hg wet deposition appeared to be in reasonable agreement with observations at 7.2 percent NMSE_s



Figure 7. Simulated vs. observed Hg wet deposition at 31 NADP sites over March–November 2010

3.3 Controlling factors of Hg wet deposition

3.3.1 Precipitation and Dynamic Processes

Over 2000–2014, Hg wet deposition was significantly correlated with precipitation ($r^2 = 0.45$, p < 0.00001) with the best correlation in spring and summer as mentioned in section 3.2. A link between Hg wet deposition and climate could be established if a link could be found between precipitation and large scale dynamic processes.

Amongst all climate indices, representing large scale processes, significant positive correlation was found between precipitation at HWF and the North Atlantic Oscillation (NAO) index ($r^2 = 0.35$, p = 0.02) in spring. During a positive NAO phase, the mean North Atlantic storm track parallels the eastern North American coastline before extending northeastward (Rogers, 1990), which may give rise to relatively wet conditions near the eastern U.S. coast (Hurrell, 1995). This explains why springtime precipitation at HWF was correlated with the NAO index. Furthermore, this indicates that the long term trend in springtime Hg wet deposition was associated with that in NAO.





Of the entire study period the largest peak (3.8 μg m⁻³) of Hg wet deposition occurred in spring 2011, corresponding to the maximum precipitation (587 mm) of all years (Figure 5). This peak was attributed mostly to the Hg wet deposition and precipitation in April (1552 ng m⁻², 167 mm) and May (1675 ng m⁻², 242 mm) 2011. The average NAO index in spring 2011 is 1.01, the highest of the 2000–2014 springs. Besides, previous studies suggested that a positive NAO phase was usually associated with above-average precipitation in the northeastern U.S., due possibly to relatively strong warm-air advection during such regimes (Archambault et al., 2008). Over April–May of 201–2014 positive geopotential height anomalies were found near the northeastern U.S. coast over the North Atlantic Ocean and negative geopotential height anomalies over the Midwest U.S. (Figure 8) suggesting weakened westerly wind resulting in more warm and moisture air from the ocean forced into the studied site. To corroborate this point, hourly precipitation, wind speed, and wind direction observations from the Clean Air Status and Trends Network (CASTNET) were used to examine the precipitation distribution vs. wind speed and direction (not shown). For all the seasons, larger precipitation amounts were associated with east to southeasterly wind transporting warmer air masses with increasing moisture from the ocean.

In summary, warmer and moister air masses coming from east or southeast of HWF in the positive NAO phase regime was likely one of causes for above-average precipitation at HWF in spring. Furthermore, the significant correlation of Hg wet deposition with precipitation suggested that NAO was likely one of the important factors controlling springtime Hg wet deposition at HWF.

3.3.2 Anthropogenic Hg emissions in NYS

A sensitivity case was conducted using the same emissions as the base case but assuming no anthropogenic Hg emissions in NYS to investigate the role of domestic anthropogenic Hg emissions in controlling ambient Hg concentrations and Hg wet and dry deposition in NYS.

Without NYS anthropogenic emissions, monthly accumulated Hg wet deposition for the whole NYS region was decreased by 0.9 percent to 4.3 percent, and monthly accumulated Hg dry deposition decreased by 7.2 percent to 17.4 percent. NYS emissions appeared to have a minimum influence on HWF wet and dry deposition in summer (Figure 9). The same seasonal pattern was found in the effect of NYS emissions on ambient GOM and PBM mixing ratios (Figure 10). The seasonal pattern of difference in Hg wet and dry deposition between the two cases was most likely determined by the minimal difference in GOM and PBM in summer and larger difference in spring and fall due to the high solubility and low vapor pressure of GOM and PBM. In contrast, the difference in GEM mixing ratio between the two cases showed a relatively flat seasonal pattern with slightly larger changes in spring (Figure 11). This indicates that the summertime minimal influence of NYS emissions on GOM and PBM was possibly due to more reactive photochemistry coupled with the most efficient wet removal in summer. This was corroborated by the summertime maximum of Hg wet deposition (Figure 4). If the summertime Hg wet deposition was mostly controlled by the photochemical environment and meteorological conditions, instead of emissions, it could explain why there was no decreasing trend in observed Hg wet deposition despite the decrease of anthropogenic emissions.

Of the three Hg forms, GOM mixing ratio was influenced the most (a \sim 37 percent decrease on average) by NYS anthropogenic Hg emissions, followed by GEM (\sim 2 percent) and PBM (\sim 0.6 percent). In addition, the NYS domain-mean GEM and GOM mixing ratios showed very similar diurnal patterns of the effect of NYS emissions with larger difference at night and minimal during daytime (not shown). The larger nighttime effect was likely due to more stable atmospheric conditions in the nocturnal boundary layer. However, the PBM diurnal pattern showed the largest decrease in the early morning (6–7 a.m.) for the entire NYS region. For urban areas, apart from the early morning peak, a secondary peak in late afternoon (6–8 p.m.) was also observed. These changes in PBM diurnal variation seemed to be related to the traffic peaks in urban areas, suggesting the relative important role of traffic emissions in PBM mixing ratios.





Figure 10. NYS averaged percentage difference in monthly mean GEM, GOM, and PBM concentrations between the base case and "no NYS emissions" case for the period of March–November 2010



4 Summary and Implications

The key findings from this project thus far are:

- A significant decreasing trend was found in Hg concentrations in precipitation and no trend in Hg wet deposition during the 15-year period from 2000 to 2014.
- A strong correlation of Hg wet deposition with precipitation was found in spring and summer and a significant correlation with Hg concentrations in precipitation in fall and winter.
- The large-scale dynamical forcing of NAO appeared to be a predominant factor influencing the interannual variation in springtime Hg wet deposition.
- Simulations of the seasonal and diurnal variations in GEM, GOM, and PBM significantly improved using our improved CMAQ-Hg model compared to using the default model.
- In-state anthropogenic Hg emissions contributed more to Hg dry deposition (13 percent) than to Hg wet deposition (2 percent) over the entire NYS region.
- In urban and suburban areas of NYS, the contribution of in-state anthropogenic Hg emissions could reach 26 percent and 76 percent of total Hg wet and dry deposition, respectively, and 63 percent of ambient GOM mixing ratios.
 - In summer, when Hg wet deposition accumulation peaked, Hg wet and dry deposition in NYS was likely influenced more by meteorological conditions and atmospheric photochemical conditions compared to in-state Hg anthropogenic emissions. This could explain why there was no trend in observed Hg wet deposition despite anthropogenic emission reductions.

The implications of our findings are:

- Anthropogenic Hg emission reductions in NYS appeared to be much more effective in reducing Hg dry deposition than wet deposition, and the effect of emission reductions appeared to be the largest for wet deposition in spring and for dry deposition in spring and fall.
 - In summer, meteorological conditions and atmospheric photochemistry seemed to affect Hg wet and dry deposition in NYS more than in-state anthropogenic emissions. This perhaps contributed to the overall lack of trend in Hg wet deposition, which was suggested by the HWF data, despite anthropogenic emission reductions over the past decades.

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