Time-Series Trends of Mercury Deposition Network Data

Peter Weiss[1], David Gay[2], Mark Brigham[3], Matthew Parsons[4], Arnout Ter Schure[5]

[1]University of California, Santa Cruz, [2]National Atmospheric Deposition Program, [3]U.S. Geological Survey, [4]Environment Canada, [5]Electric Power Research Institute

Acknowledgements to: Leonard Levin, Udaysankar Nair, Greg Weatherbee, Dave Lorentz, and Electric Power Research Institute for funding

Presented at the 2014 NADP Fall Meeting, Indianapolis, IN

Objectives

- Initiate work on analyses of spatial gradients and temporal trends in mercury deposition and atmospheric concentrations using data from the Mercury Deposition Network.

- The primary question addressed is “Do the reductions in mercury emissions from EGUs (and other sources) in the United States, driven by MATS and other regulations, especially since 2007, translate into observed changes in: a) mercury concentrations in precipitation and/or b) total annual mercury wet deposition?

MDN Data – QA/QC

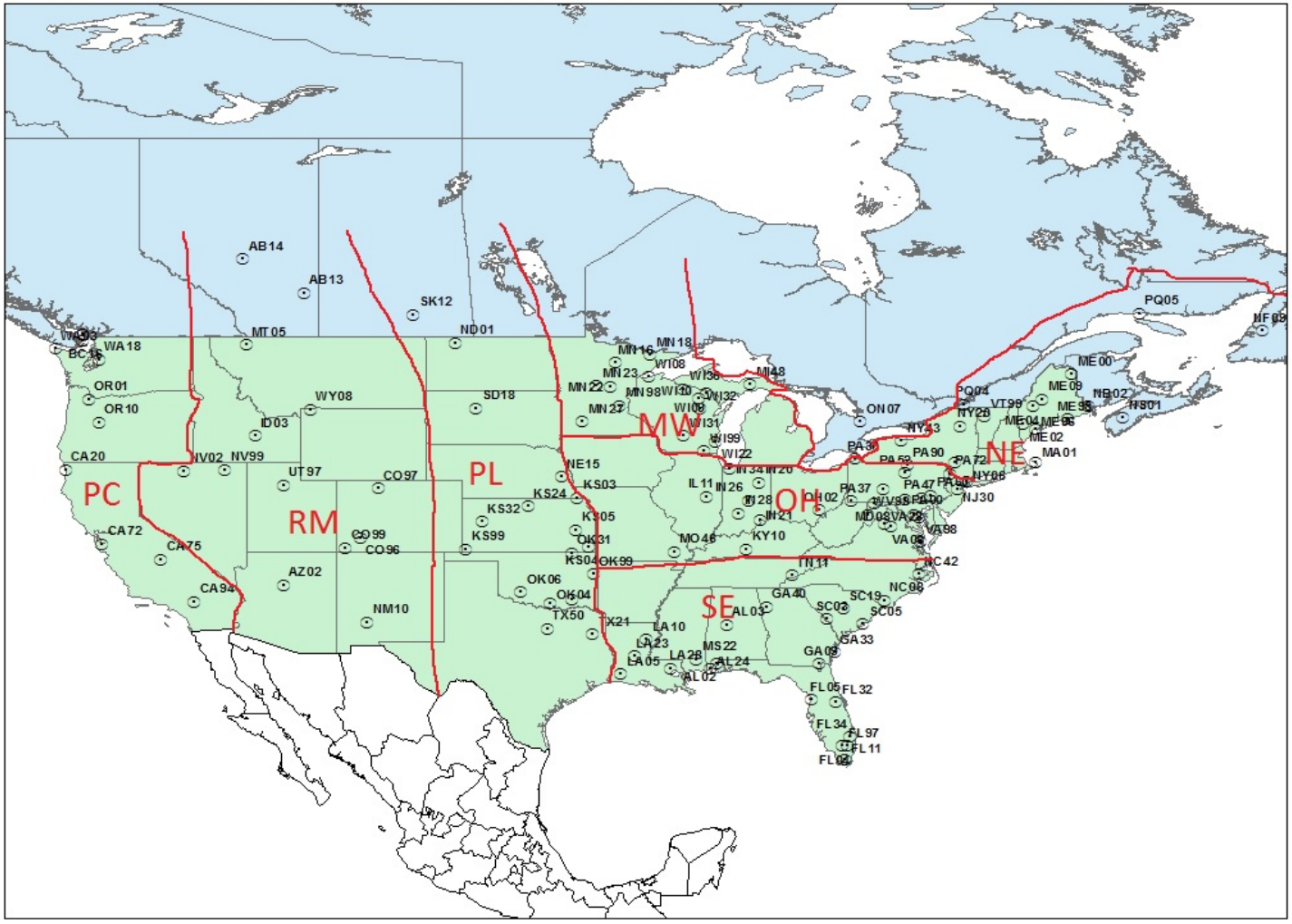
- Dry-week and collector-rinse samples, and laboratory quality control (QC) data, supplied by the Mercury Analytical Laboratory (HAL) and USGS Branch of Quality Systems, were evaluated for Hg recovery trends and compared to environmental trends, 1996-2011.

    - Trends in QC data are null or not of sufficient magnitude to explain precipitation Hg concentration temporal trends.

Approach

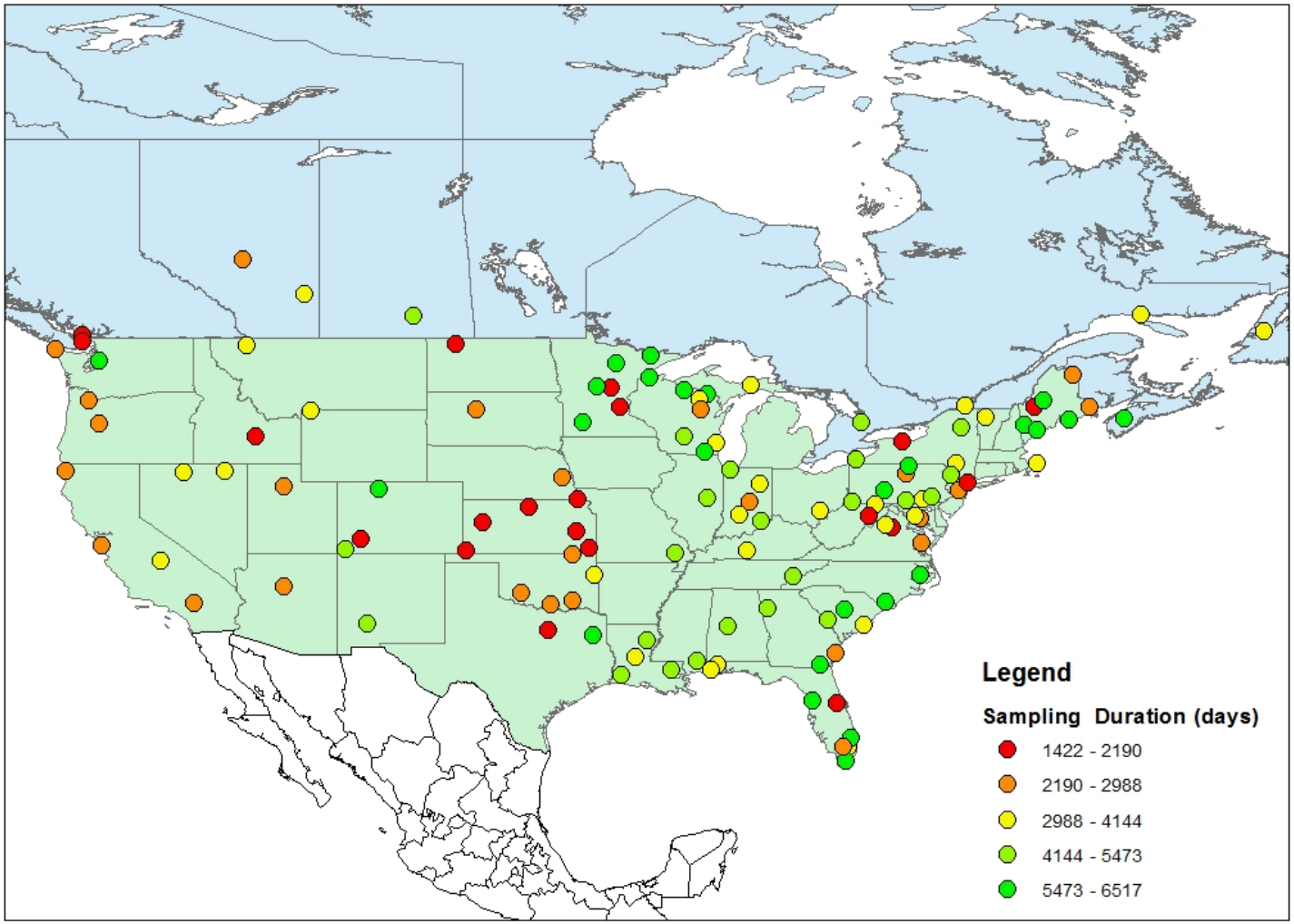
- 127 out of 185 Mercury Deposition Network sites selected with data records > 5 years.
- Trend analyses performed:
  1. Seasonal Mann-Kendall – ranking system of monthly sums for Precipitation and Hg Deposition, and monthly medians for Hg Concentration. Non-parameteric, does not require normality of input data (Prestbo and Gay, 2009).
     - Calculations independently derived by D. Gay and M. Parsons and results verified.
  2. Linear Regression Model – Includes precipitation and season to model the variation in [Hg] and uses a Fourier transform to make the data more normally distributed.
     - log[Hg] is a function of log[precip] + sine(2*pi*time) + cosine(2*pi*time) + sine(4*pi*time) + cosine(4*pi*t)
  3. Linear Regression on Annual Totals and Means – Simple linear fit of annual total deposition and precipitation weighted mean concentration data.

MDN Sites with > 5 Years of Data and Assigned Regions



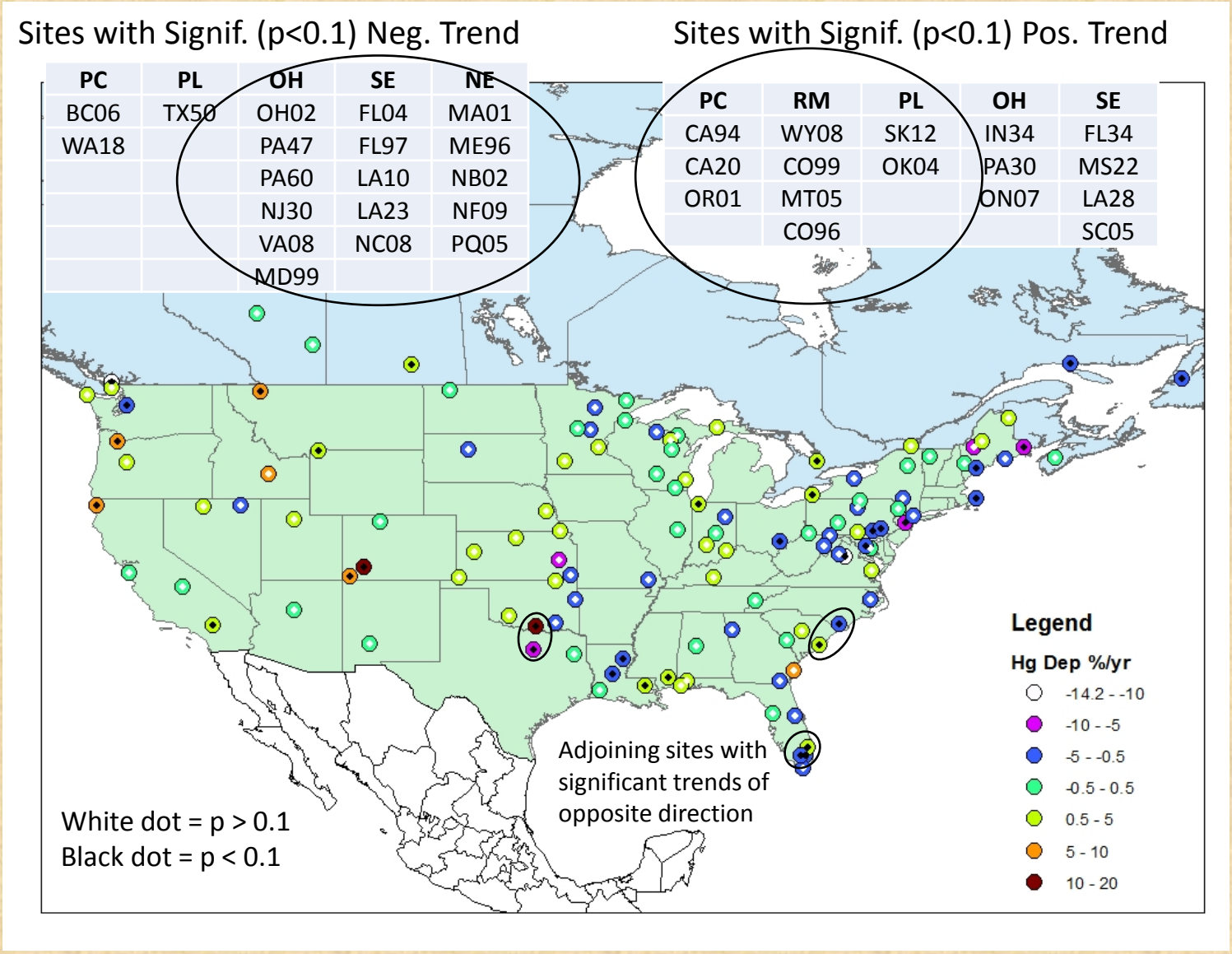
Region	Number of Sites
PC	10
RM	13
PL	15
MW	15
OH	27
SE	23
NE	19

Duration of Data Set for Each Site > 5 Years



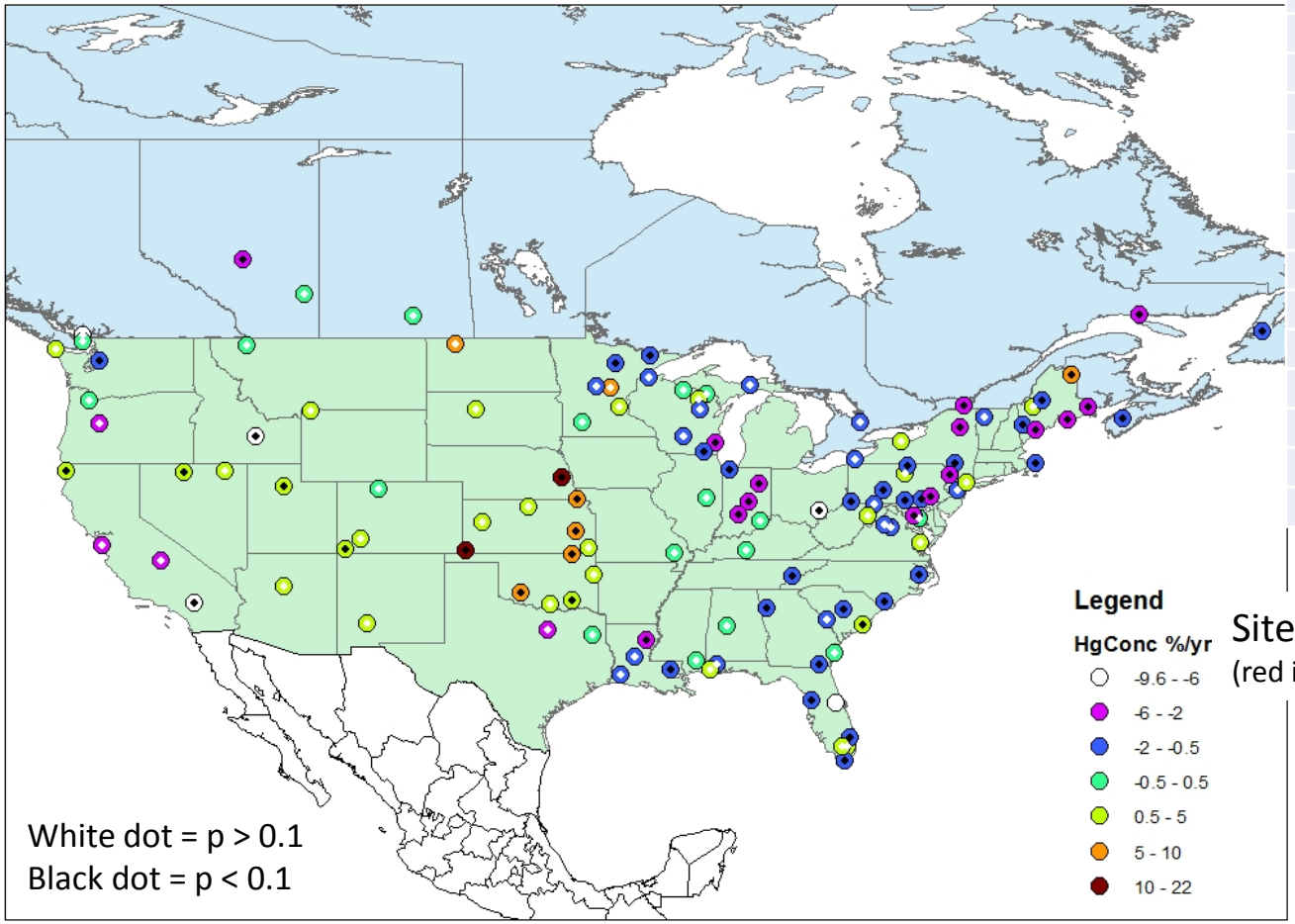
Trends in Hg Deposition with Significance

Mann-Kendall Procedure Using Monthly Sums of Weekly Data



Trends in Hg Concentration with Significance

Mann-Kendall Procedure Using Monthly Medians of Weekly Data



Sites with Signif. (p<0.1) Neg. Trend

(green indicates decreasing trend in Hg Dep)

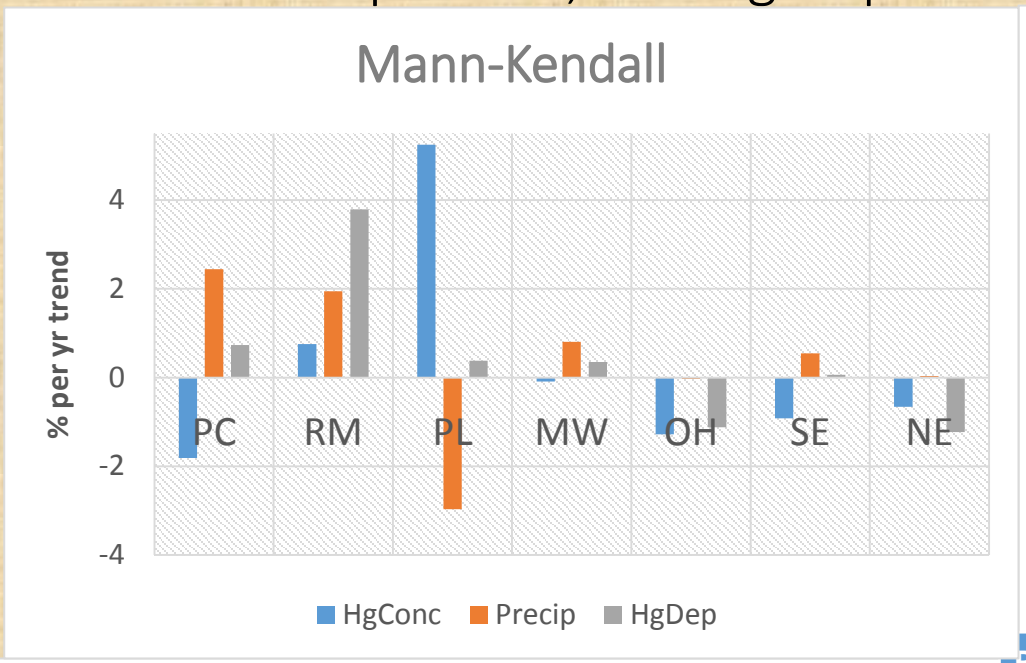
PC	RM	MW	OH	SE	NE
CA94	ID03	WI22	OH02	LA10	NB02
WA18	AB14	MN18	IN20	NC08	PQ04
		WI99	IN26	GA40	ME96
		MN16	MD99	SC19	PQ05
			PA72	GA09	NY20
			PA60	FL11	ME98
			IN28	TN11	NY68
			PA00	FL05	ME02
			PA47	FL34	MA01
			PA13	LA28	NS01
			IN34	NC42	NF09
			PA37		ME09
			PA90		

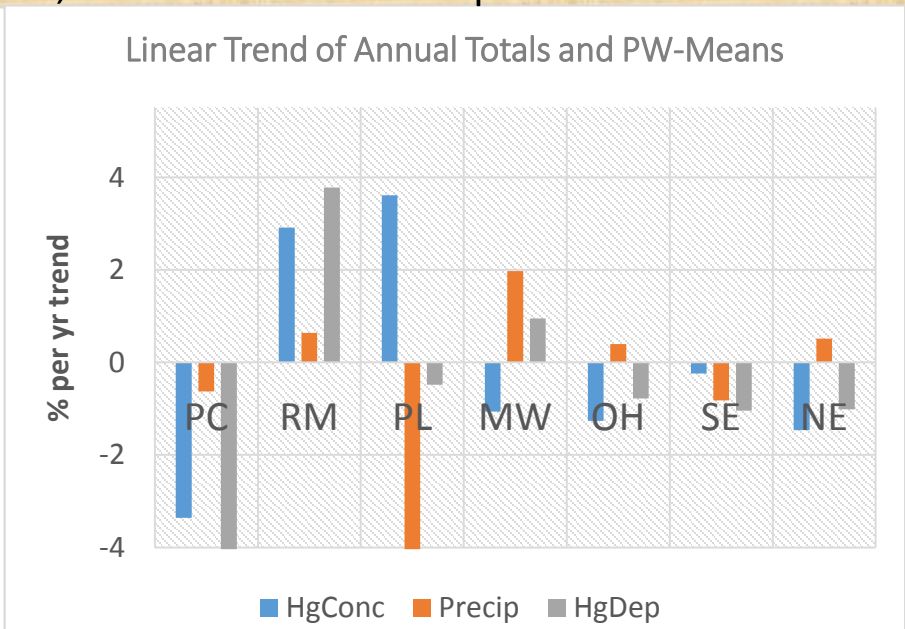
Sites with Signif. (p<0.1) Pos. Trend

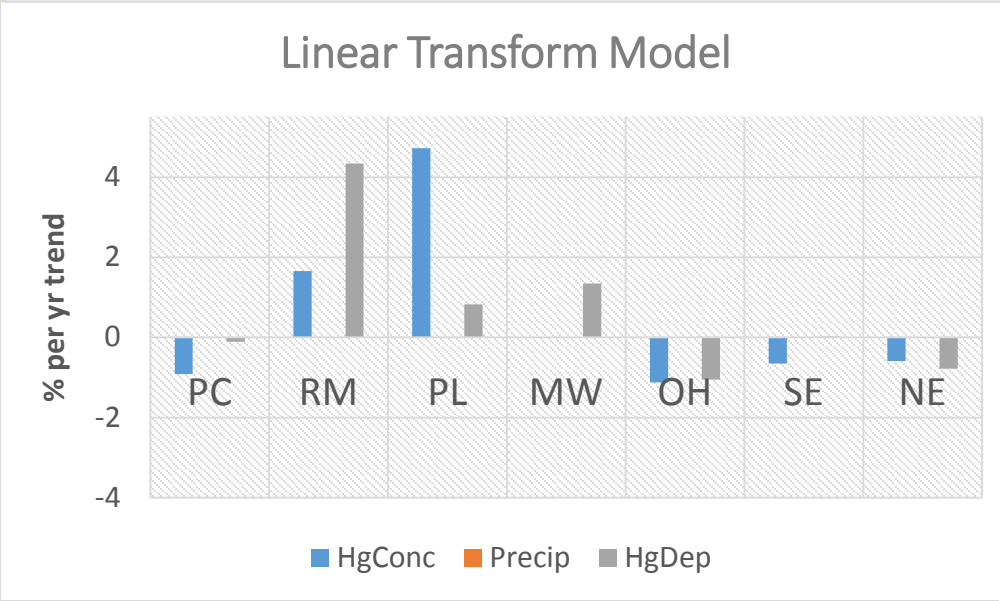
(red indicates increasing trend in HgDep)

PC	RM	PL	SE	NE
CA20	NV02	OK01	SC05	ME00
	CO99	OK06		
	UT97	OK31		
		KS03		
		KS05		
		NE15		
		KS99		

Mean Regional Trends in PWM-Hg Concentration, Precipitation, and Hg Deposition; 3 Method Comparison





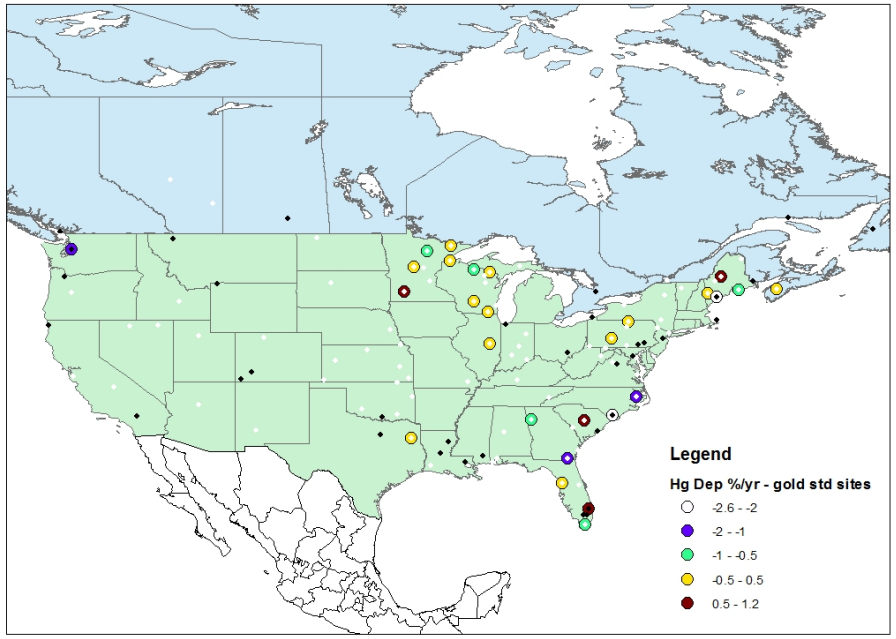


Region	HgConc	Precip	HgDep
PC	-	unclear	unclear
RM	+	+	+
PL	+	-	unclear
MW	-	+	+
OH	-	unclear	-
SE	-	unclear	unclear
NE	-	unclear	-

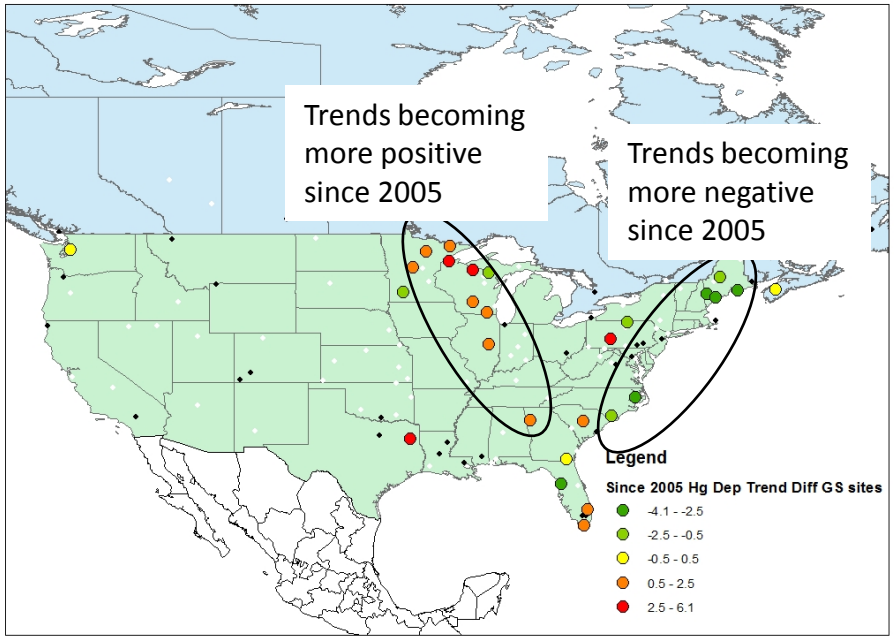
Agreement by 3 Methods

Hg Deposition Trends Since 2005 Compared Against the Trend Since 1998

1998 – 2013 Trends



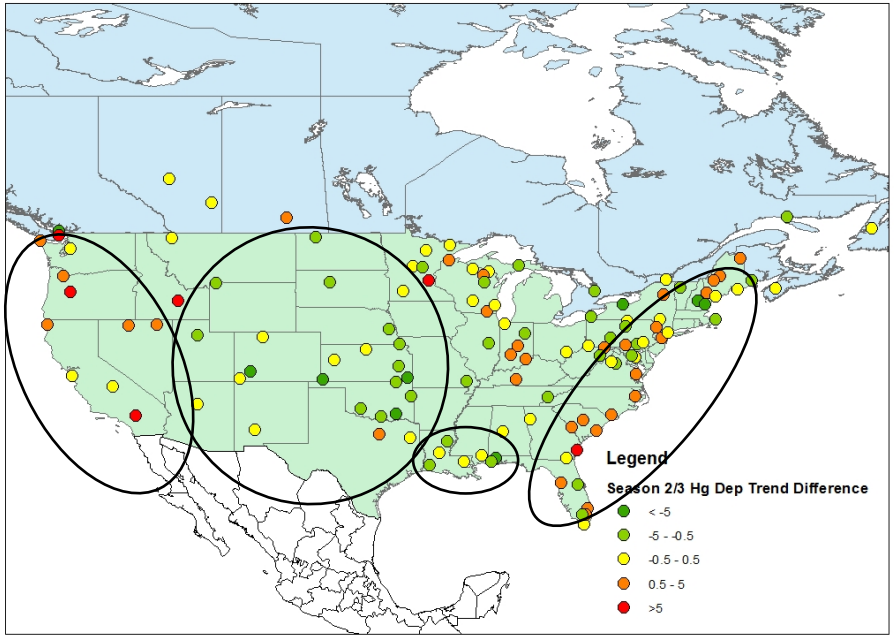
Difference between 2005-2013 and 1998-2013 Trends



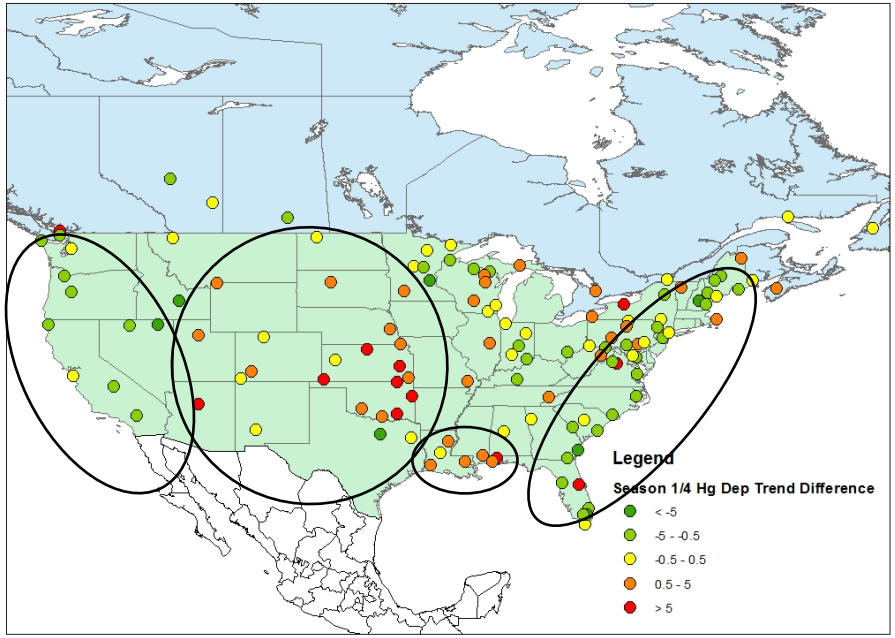
Sites shown are those with the longest (1998 – 2013) data records

Hg Deposition Trends as a Function of Season

Difference between spring/summer trend and the all data trend



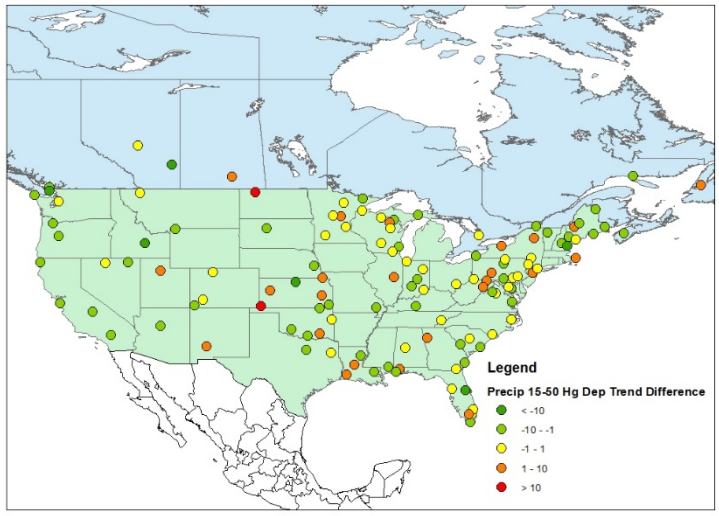
Difference between the fall/winter trend and the all data trend



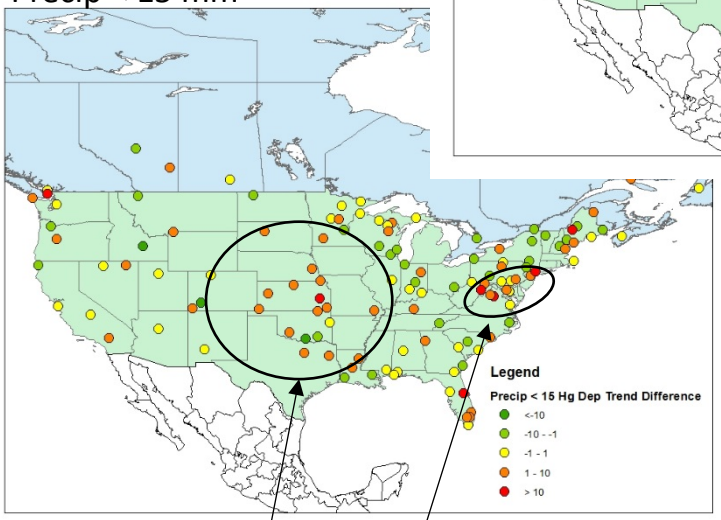
Spring/Summer trend increases: NE, eastern SE, and PC.
Fall/Winter trend increases: RM, PL, and western SE.

Hg Deposition Trends as a Function of Precipitation Amount

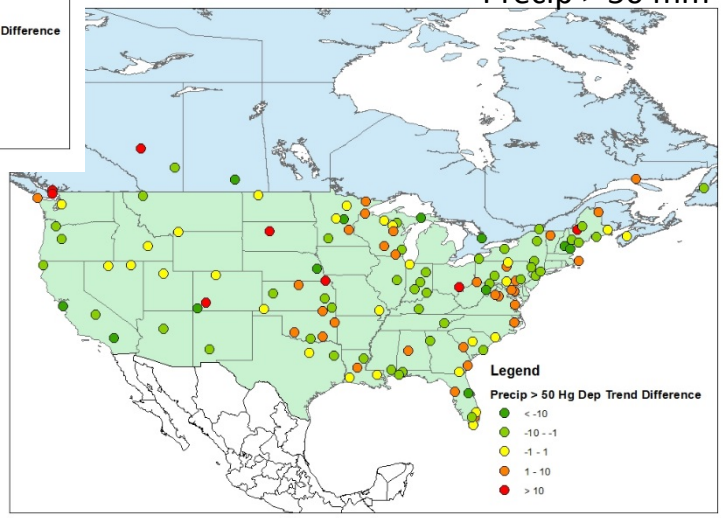
Precip between 15 and 50 mm



Precip < 15 mm



Precip > 50 mm

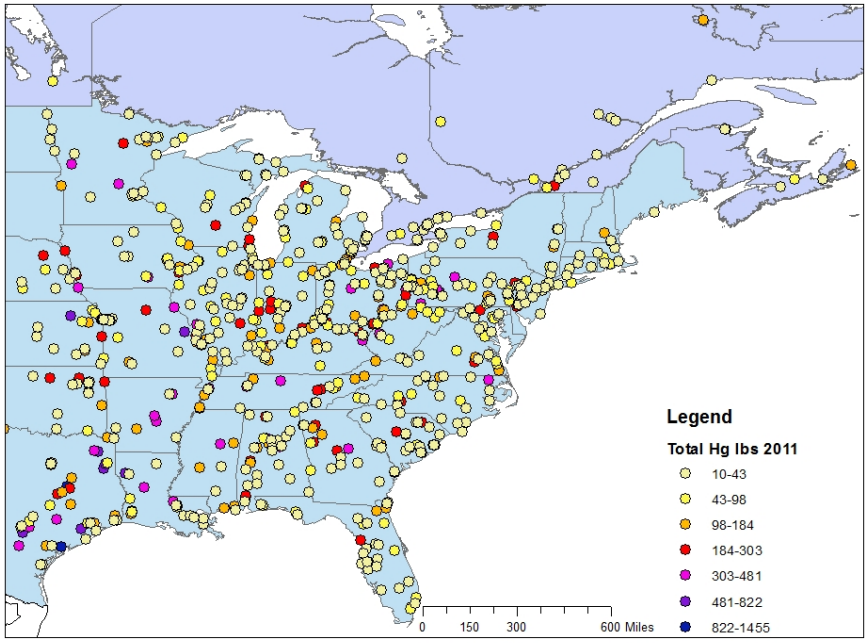


All Compared to
Trends Using All Data

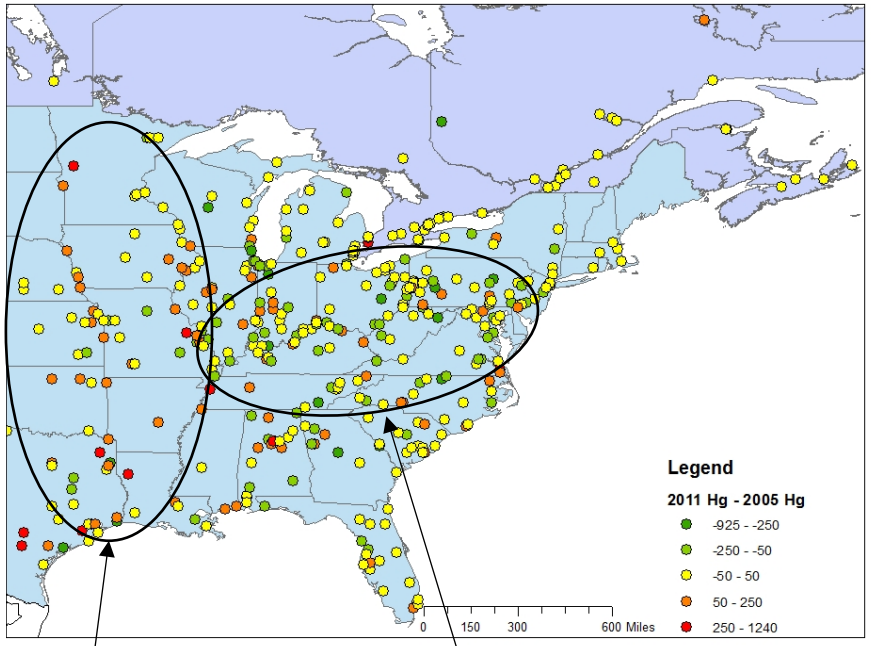
Increasing trends in low precip volume range

Point Source Total Hg Emissions > 10 lbs/yr
U.S. – NEI Data; Canada – NPRI Data

2011 Emissions



2011 Emissions – 2005 Emissions

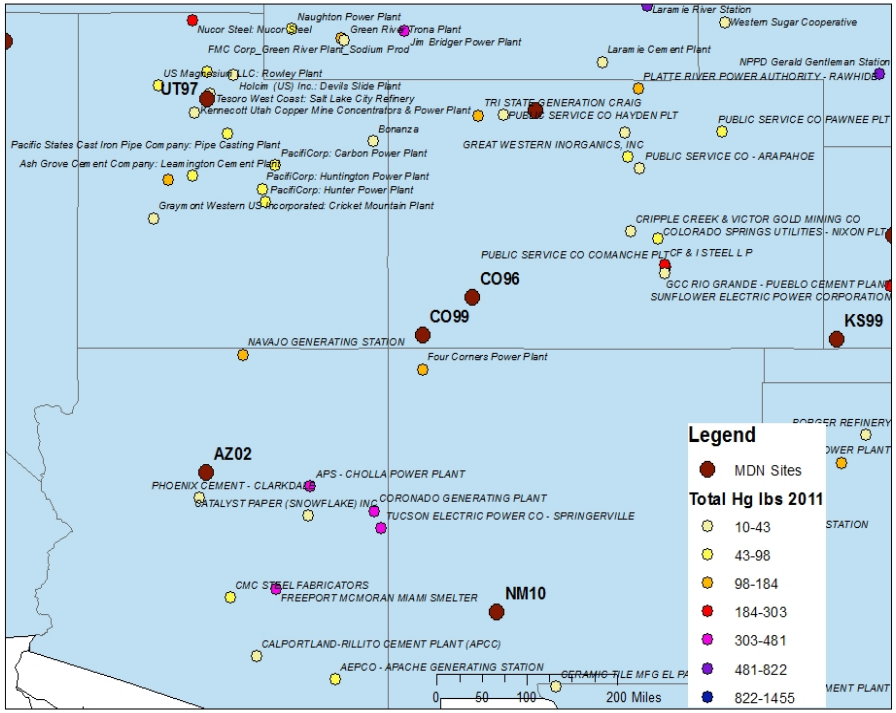


Some increasing emissions since 2005

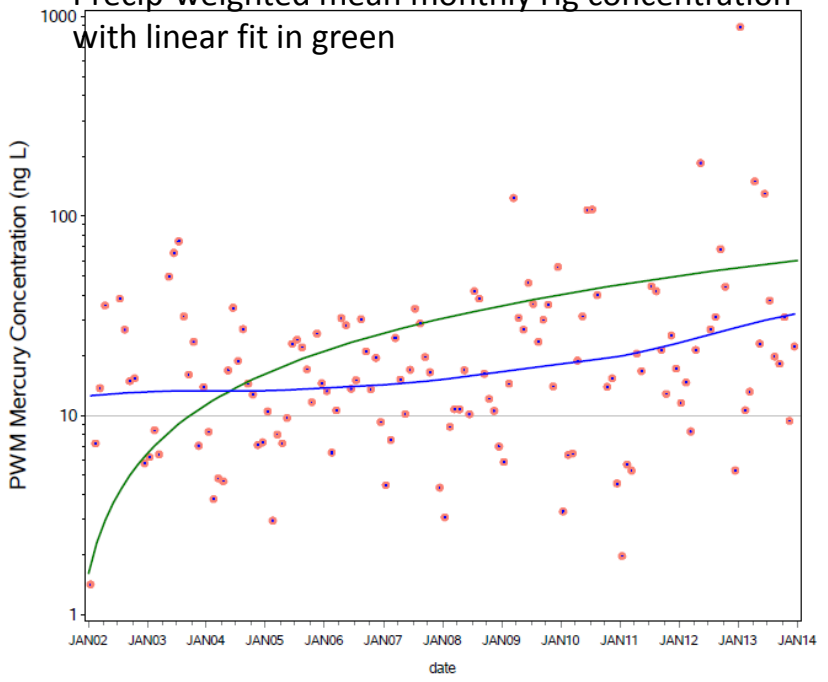
Mostly decreasing emissions since 2005

Increasing [Hg] and Hg Dep Trends at CO99

CO99	Mann-Kendall (sums) (%/yr)	Linear Transform Model (%/yr)	Linear, Annual (%/yr)
Hg Conc	4.1 (signif)	7.5 (signif)	7.2 (signif)
Hg Dep	7.0 (signif)	10.8 (signif)	9.6 (signif)



Precip-weighted mean monthly Hg concentration with linear fit in green



Year	4-Corners Power Plant Hg lbs/yr	Data Source
2005	544	NEI
2010	367	EPRI indirect
2011	164	NEI
2012	518	EPRI direct

Conclusions

- Trends in Concentration and Deposition are statistically significant at many sites across the U.S. and Canada.
- Mann-Kendall and Linear Regression models produced reasonably similar results.
- From a regional perspective:
  - Hg Deposition and Hg Concentration are decreasing in NE and OH regions.
  - Hg Deposition and Hg Concentration are increasing in RM region.
  - Hg Concentration is decreasing in SE region, but increasing Precipitation is causing Hg Deposition to be slightly increasing.
  - Hg Concentration is strongly increasing in PL region, but decreasing Precipitation is causing Hg Deposition to be only slightly increasing.
- Trends since 2005 compared to 1998-2013: generally more negative in NE and more positive in MW.
- Seasonality: Spr/Sum increases in NE, PC and SE; Fall/Win increases in PL, RM.
- Trends increasing in PL and decreasing in NE for lowest precip. volume samples.
- CO99 displays strongly positive trends not explained by emissions at local coal-fired power plant.

Hypotheses to be Tested

1. Domestic Hg emissions reductions has led to a measureable decrease in Precipitation [Hg] in the MW, OH, SE, and NE regions.
2. Downward trend in hemispheric background level of GEM in atmosphere is also contributing to the observed Precipitation [Hg] trends.
3. Increasing trend in oxidant concentration in Western North America is contributing to the increasing Precipitation [Hg] trends in the western regions.