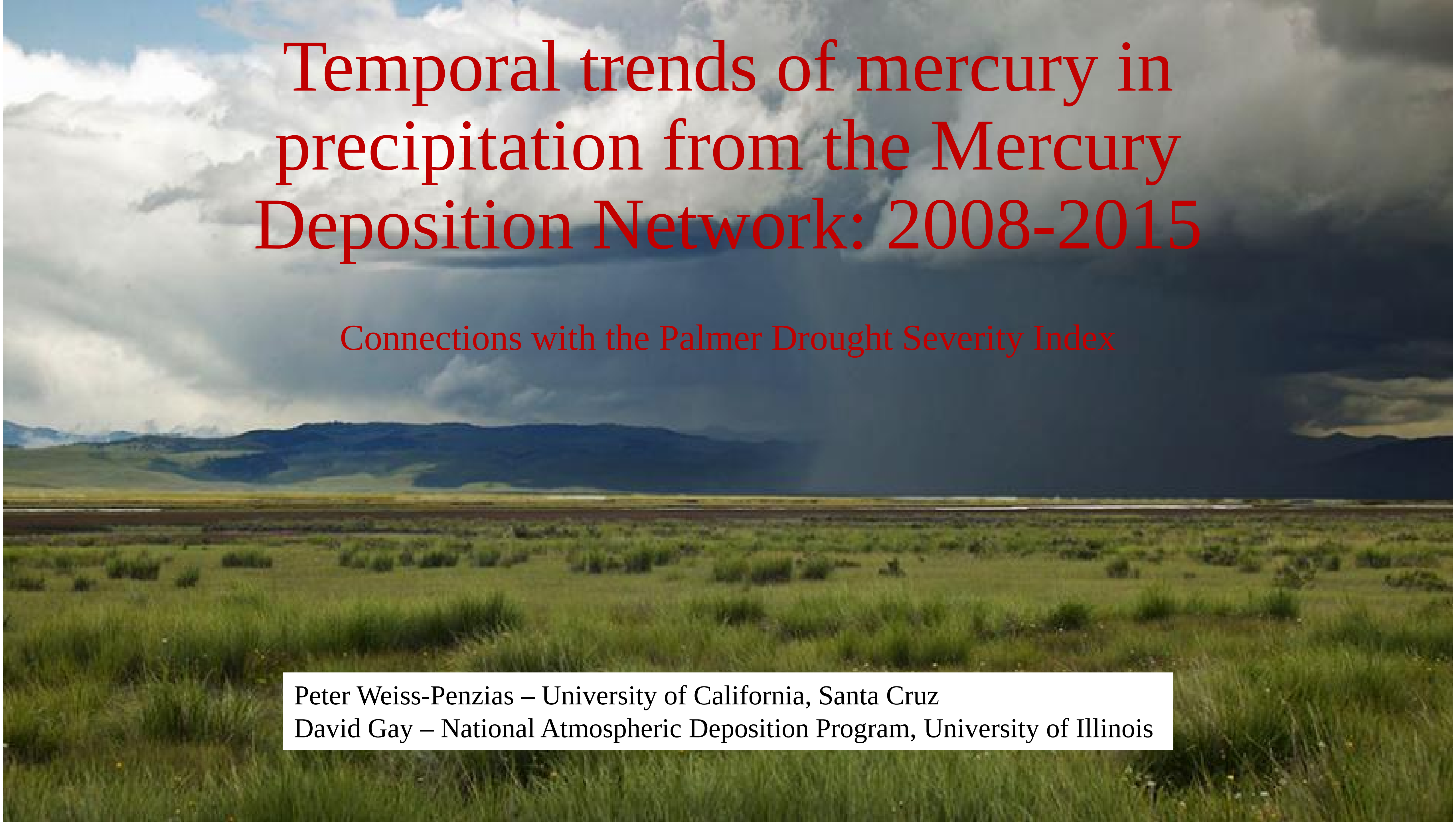


Temporal trends of mercury in precipitation from the Mercury Deposition Network: 2008-2015

Connections with the Palmer Drought Severity Index

Peter Weiss-Penzias – University of California, Santa Cruz
David Gay – National Atmospheric Deposition Program, University of Illinois

Outline

- Results from a study of **temporal trends** in Hg concentration in precipitation from MDN and Hg air concentrations from AMNet and other networks in North America using available data up through 2013.
  - How have trends in atmospheric Hg in North America been affected by decreasing anthropogenic Hg emissions from North America?
- Results of a comparison between 2008-2013 and 2008-2015 trends in Hg concentration in wet deposition.
  - Did the trends from the earlier term continue with the addition of the most recent data from the MDN?
- Results of a correlation analysis between monthly precipitation-weighted mean Hg concentration vs. the Palmer Drought Severity Index (PDSI) by region.
  - Can the relationship between Hg and PDSI by region tell us something about the reasons for the observed trends in Hg?

Trends in atmospheric Hg^0 and Hg^{II} wet deposition concentrations from a recent literature survey

PNAS

2016

Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions

Yanxu Zhang[a,1], Daniel J. Jacob[a,b], Hannah M. Horowitz[a], Long Chen[a,c], Helen M. Amos[a], David P. Krabbenhoft[d], Franz Slemr[e], Vincent L. St. Louis[f], and Elsie M. Sunderland[a,g]

In the U.S. and Europe, most trends are negative!

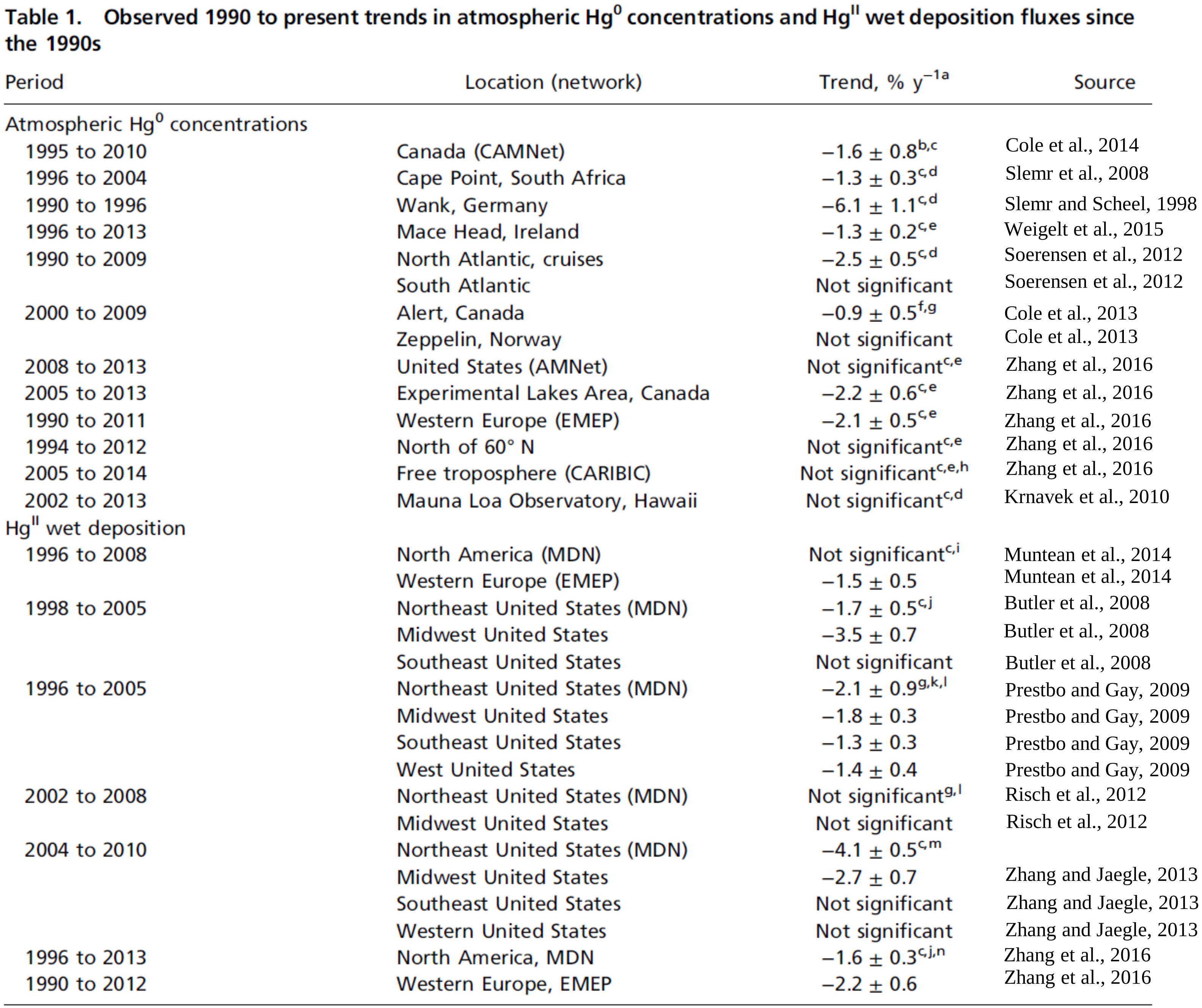


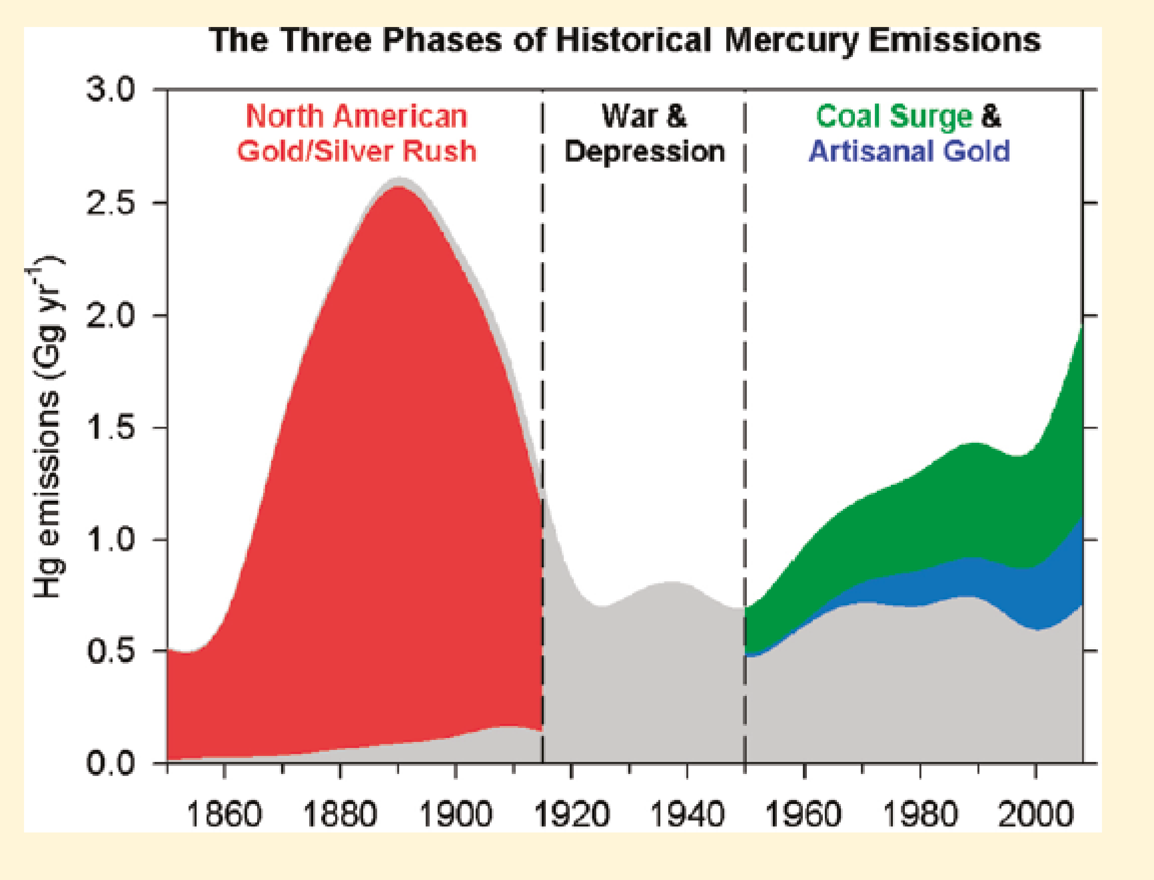
Table 1. Observed 1990 to present trends in atmospheric Hg^0 concentrations and Hg^{II} wet deposition fluxes since the 1990s

Period	Location (network)	Trend, % y^{-1}[a]	Source
Atmospheric Hg^0 concentrations			
1995 to 2010	Canada (CAMNet)	−1.6 ± 0.8[b,c]	Cole et al., 2014
1996 to 2004	Cape Point, South Africa	−1.3 ± 0.3[c,d]	Slemr et al., 2008
1990 to 1996	Wank, Germany	−6.1 ± 1.1[c,d]	Slemr and Scheel, 1998
1996 to 2013	Mace Head, Ireland	−1.3 ± 0.2[c,e]	Weigelt et al., 2015
1990 to 2009	North Atlantic, cruises	−2.5 ± 0.5[c,d]	Soerensen et al., 2012
	South Atlantic	Not significant	Soerensen et al., 2012
2000 to 2009	Alert, Canada	−0.9 ± 0.5[f,g]	Cole et al., 2013
	Zeppelin, Norway	Not significant	Cole et al., 2013
2008 to 2013	United States (AMNet)	Not significant[c,e]	Zhang et al., 2016
2005 to 2013	Experimental Lakes Area, Canada	−2.2 ± 0.6[c,e]	Zhang et al., 2016
1990 to 2011	Western Europe (EMEP)	−2.1 ± 0.5[c,e]	Zhang et al., 2016
1994 to 2012	North of 60° N	Not significant[c,e]	Zhang et al., 2016
2005 to 2014	Free troposphere (CARIBIC)	Not significant[c,e,h]	Zhang et al., 2016
2002 to 2013	Mauna Loa Observatory, Hawaii	Not significant[c,d]	Krnavek et al., 2010
Hg^{II} wet deposition			
1996 to 2008	North America (MDN)	Not significant[c,i]	Muntean et al., 2014
	Western Europe (EMEP)	−1.5 ± 0.5	Muntean et al., 2014
1998 to 2005	Northeast United States (MDN)	−1.7 ± 0.5[c,j]	Butler et al., 2008
	Midwest United States	−3.5 ± 0.7	Butler et al., 2008
	Southeast United States	Not significant	Butler et al., 2008
1996 to 2005	Northeast United States (MDN)	−2.1 ± 0.9[g,k,l]	Prestbo and Gay, 2009
	Midwest United States	−1.8 ± 0.3	Prestbo and Gay, 2009
	Southeast United States	−1.3 ± 0.3	Prestbo and Gay, 2009
	West United States	−1.4 ± 0.4	Prestbo and Gay, 2009
2002 to 2008	Northeast United States (MDN)	Not significant[g,l]	Risch et al., 2012
	Midwest United States	Not significant	Risch et al., 2012
2004 to 2010	Northeast United States (MDN)	−4.1 ± 0.5[c,m]	
	Midwest United States	−2.7 ± 0.7	Zhang and Jaegle, 2013
	Southeast United States	Not significant	Zhang and Jaegle, 2013
	Western United States	Not significant	Zhang and Jaegle, 2013
1996 to 2013	North America, MDN	−1.6 ± 0.3[c,j,n]	Zhang et al., 2016
1990 to 2012	Western Europe, EMEP	−2.2 ± 0.6	Zhang et al., 2016

Observed trends don’t agree with most anthropogenic Hg emissions inventories – which are increasing due to expanding gold mining and coal-fired utility operations worldwide.

The Zhang et al., 2016 emissions inventory shows that current emissions have actually *decreased* due to:

1) Phasing out of Hg use in commercial products
2) Artisinal Scale Gold Mining (ASGM) emissions not being as high as reported earlier
3) Hg speciation changes that resulted from installation of emissions controls (less Hg^{II} more Hg^0).



Streets et al., 2011

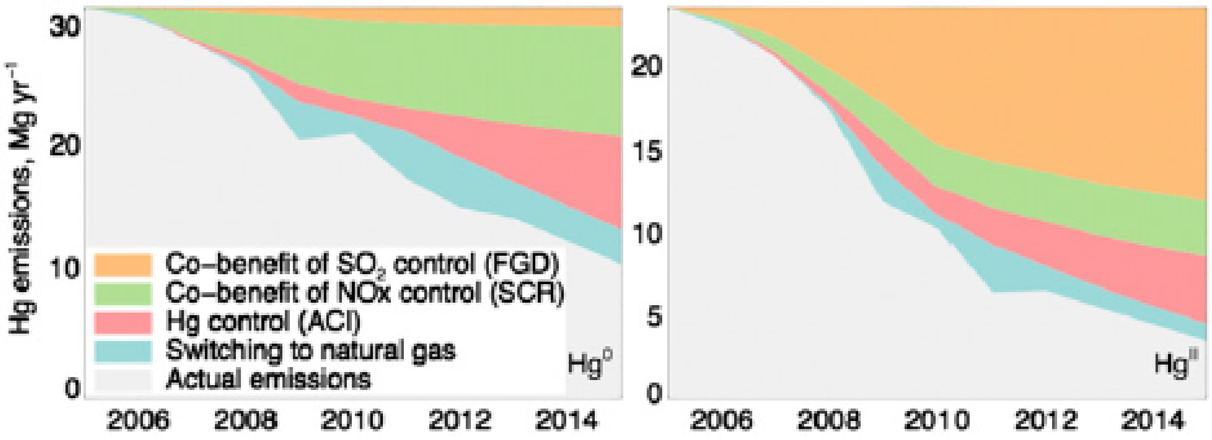
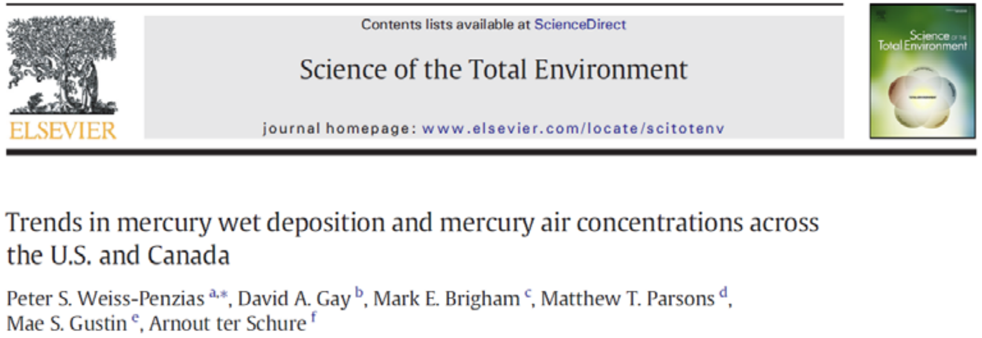


Fig. 1. Major factors driving declines in Hg emission from US coal-fired utilities between 2005 and 2015. Trends were inferred from data on the implementation of different types of emission control technologies.

Zhang et al., 2016

Assessing trends in Hg concentrations in precipitation across the U.S. and Canada with improved spatial coverage

- Trends in Hg concentration and deposition fluxes were calculated for six time periods
- 19 sites in 1997-2013 (mostly in north/northeast regions)
- 81 sites in 2008-2013 with better coverage in the central and western U.S.
- Compare trends between regions to assess the effects of changing domestic emissions.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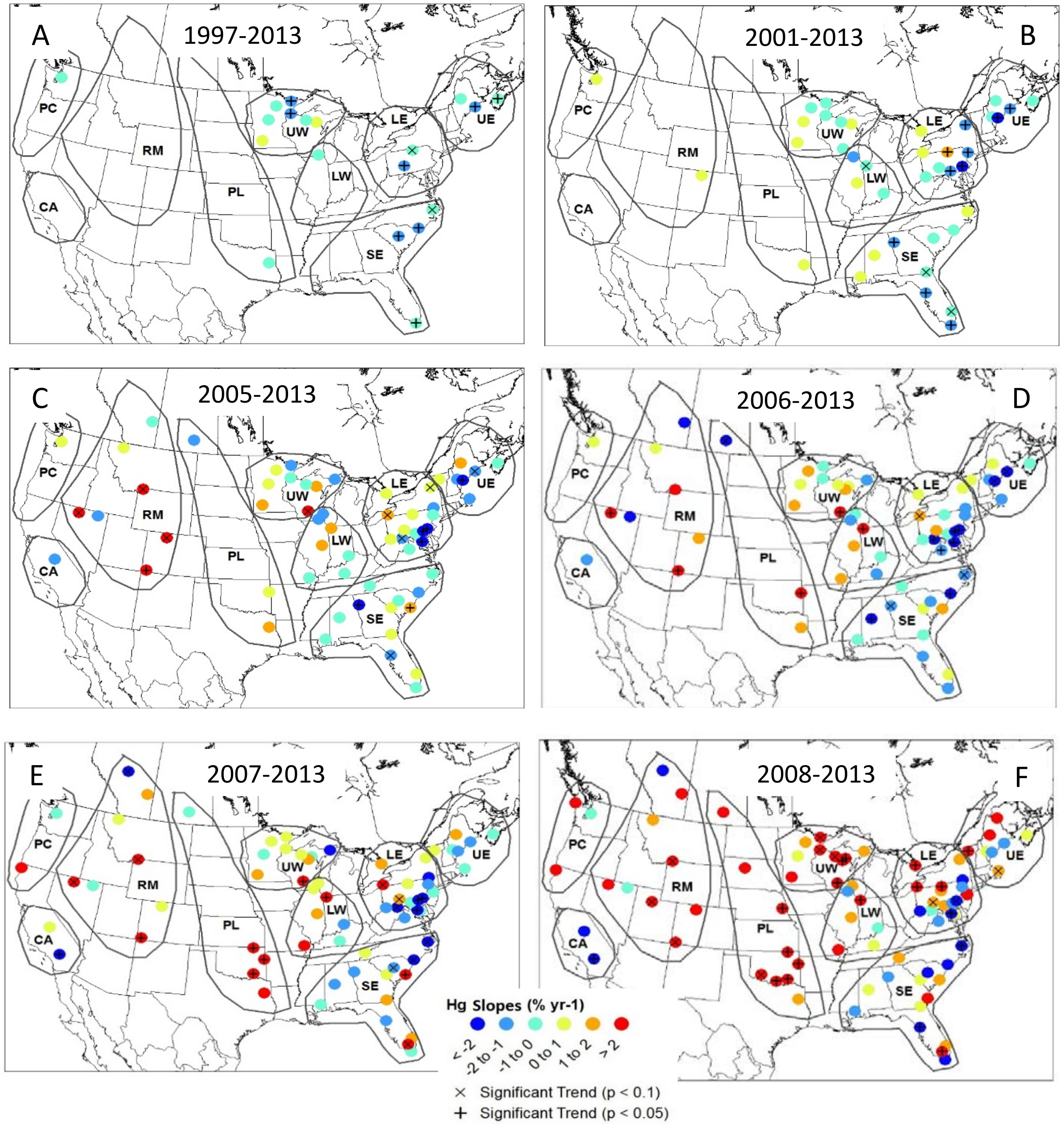
Trends in mercury wet deposition and mercury air concentrations across the U.S. and Canada

Peter S. Weiss-Penzias [a,*], David A. Gay [b], Mark E. Brigham [c], Matthew T. Parsons [d], Mae S. Gustin [e], Arnout ter Schure [f]

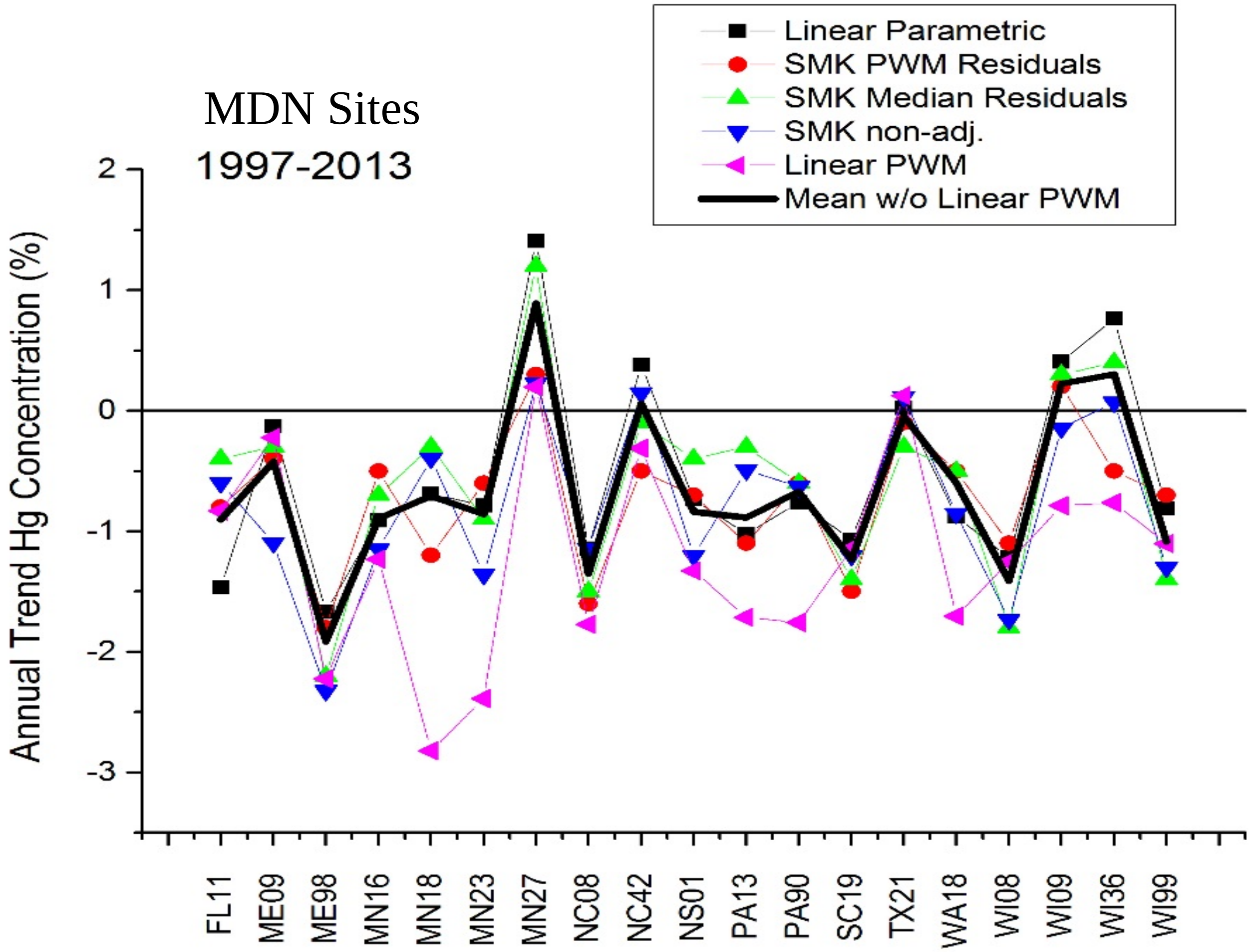
MDN sites operational during each time period



Trend calculation methods

- Due to the washout effect, Hg concentrations in precipitation follow a logarithmic relationship with precipitation depth. This complicates trend calculations and we sought to compare methods.
- Method 1: Seasonal Mann-Kendall trend modeling
  - Input data 1: precipitation-weighted monthly mean residual Hg concentration
    - $\log[Hg] = \beta_0 + \beta_1 \log(\text{precip depth}) + \varepsilon$
    - where ε is the weekly residual Hg concentration, i.e., the precipitation-depth adjusted log[Hg]
  - Input data 2: monthly median residual Hg concentration
  - Input data 3: monthly precipitation-weighted mean non-adjusted Hg concentration
- Method 2: Linear Parametric trend modeling
  - Input data: weekly Hg concentration
    - $\log[Hg] = \beta_0 + \beta_1 \log(\text{precip}) + \beta_2 \text{sine}(2\pi t) + \beta_3 \text{cosine}(2\pi t) + \beta_4 \text{sine}(4\pi t) + \beta_5 \text{cosine}(4\pi t) + \beta_6 \pi$
- Method 3: Linear Regression
  - Input data: precipitation-weighted annual mean Hg concentration
    - $[Hg] = mt + b$

Comparison between trend methods

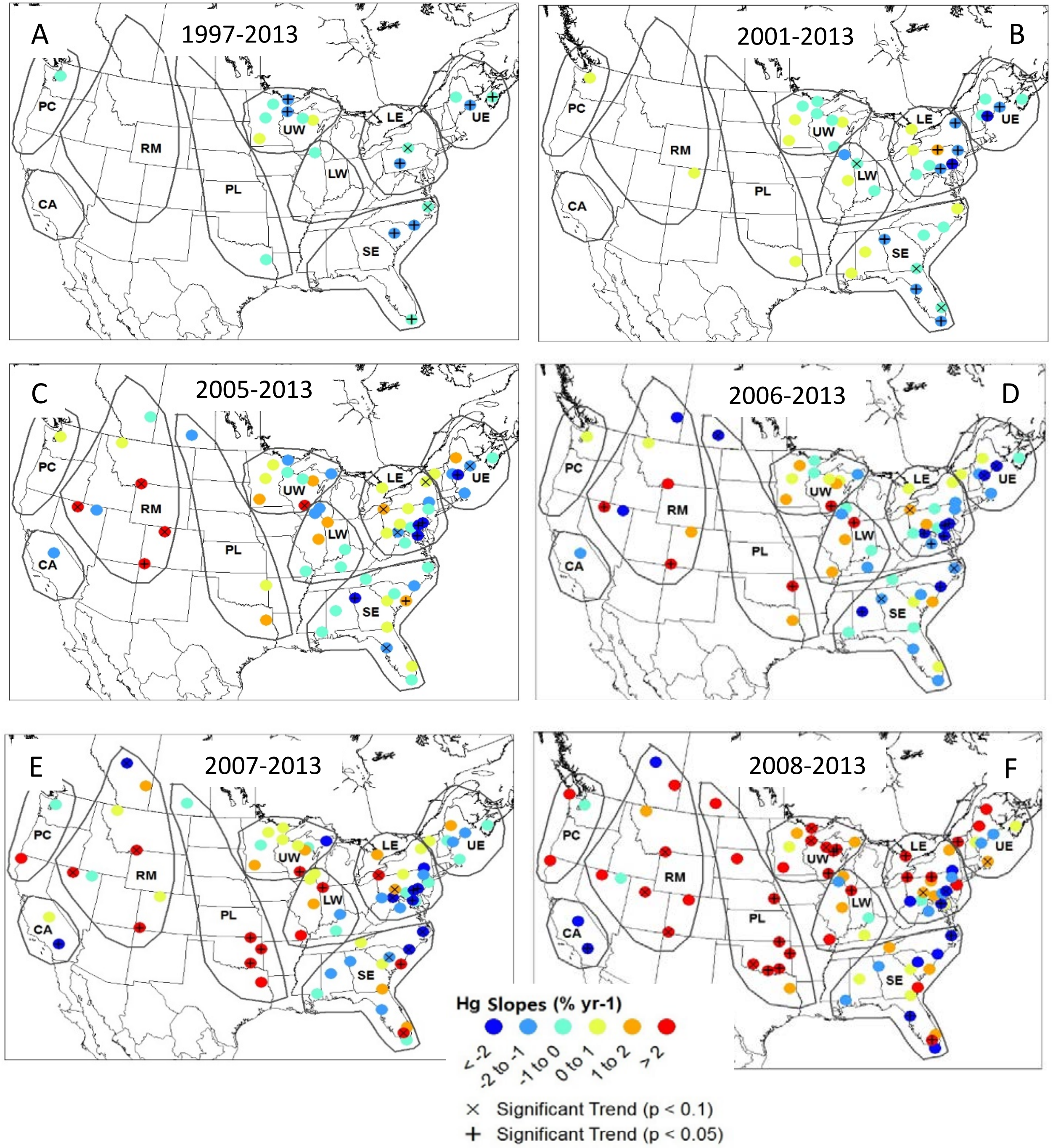


With the exception of the Simple Linear method (on annual PW means), agreement was good between methods.

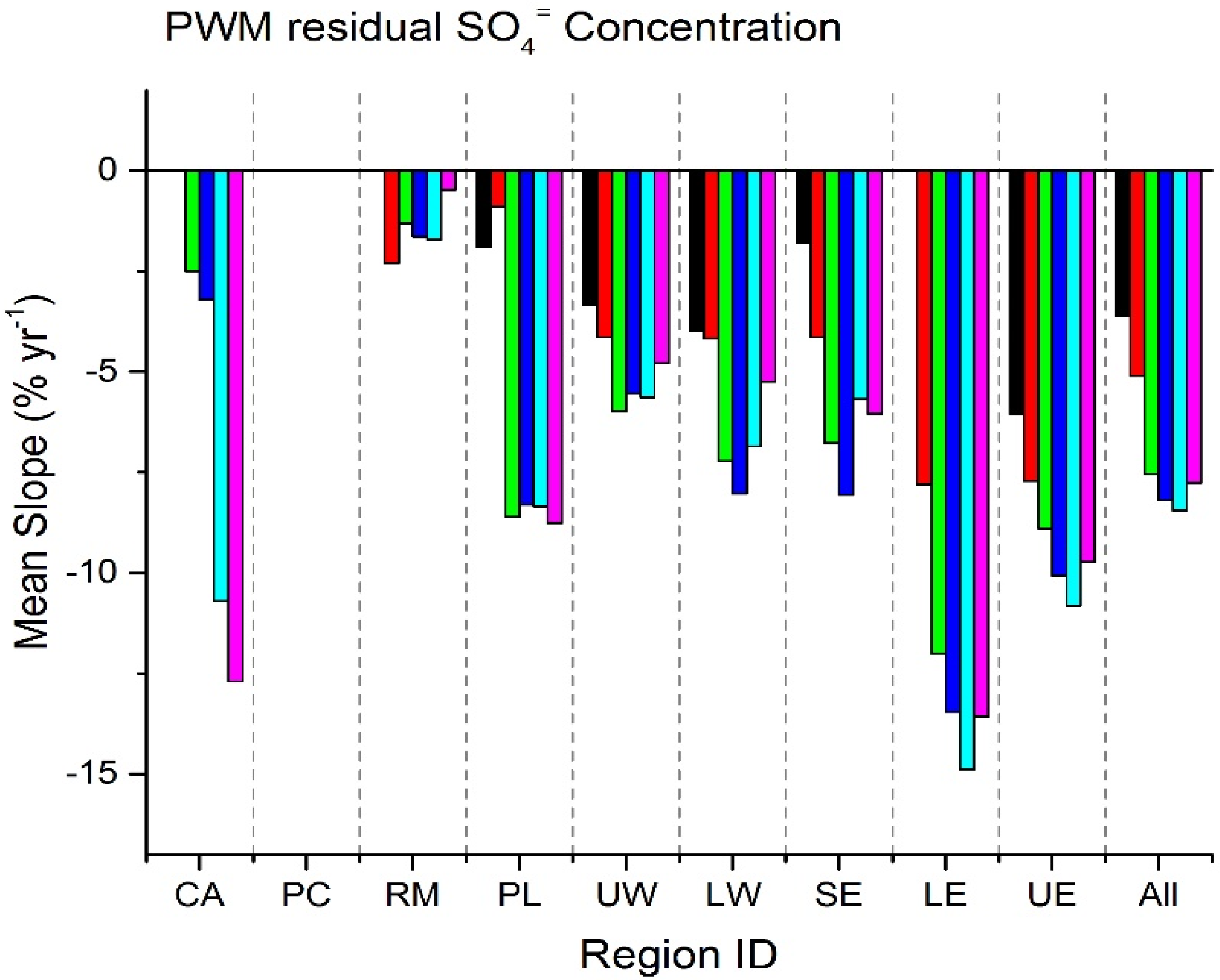
Hg concentration in wet deposition, annual rates of change.

(PWM Residual Hg Concentration)

- 53%/0% of sites had significant negative/positive trends in mercury wet deposition for 1997–2013.
- 6%/30% of sites had significant negative/positive trends for 2008–2013.
- The positive trends in mercury wet deposition were primarily found in the middle/west of continent.



Trends of $SO_4^=$ concentrations at collocated MDN/NTN sites



- No increasing trends in $SO_4^=$ concentration in recent time perids.
- All regions showing negative trends for all time periods.
- Coal combustion in North America not the likely cause of increasing trends in Hg concentration

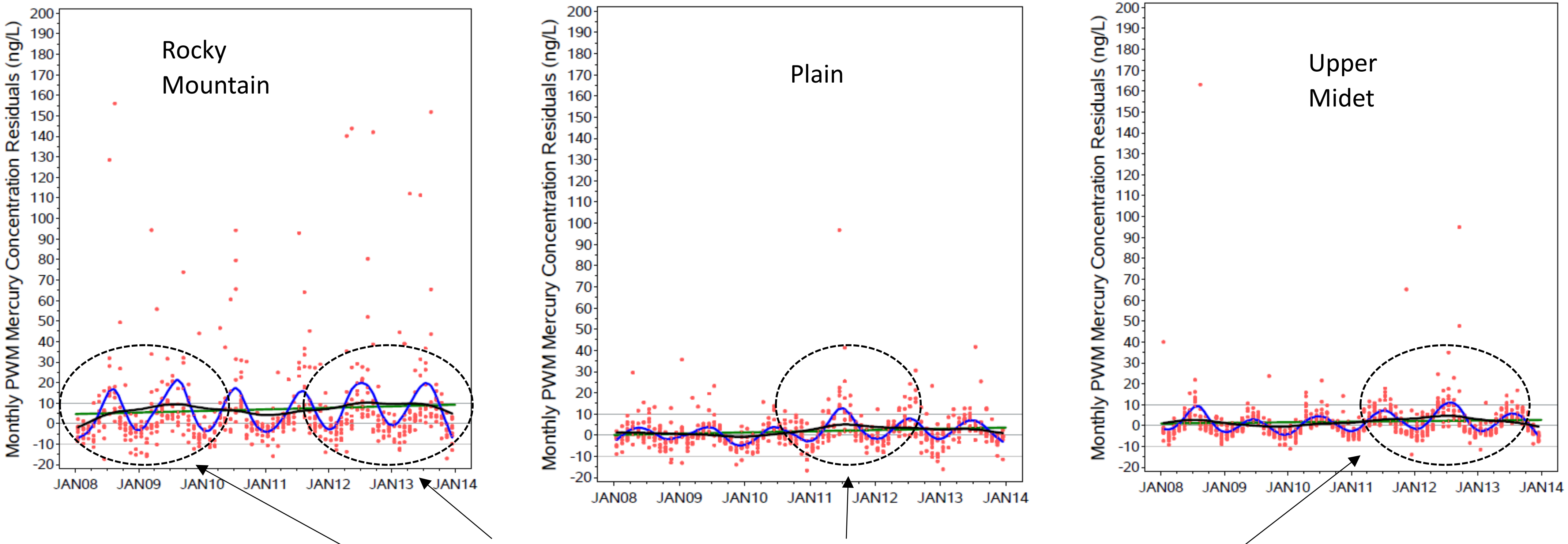
Inspection of the weekly data, smoothing curves, and linear fits for three regions with positive trends

Slopes of linear fit (2008-2013):

+3.6 %/yr

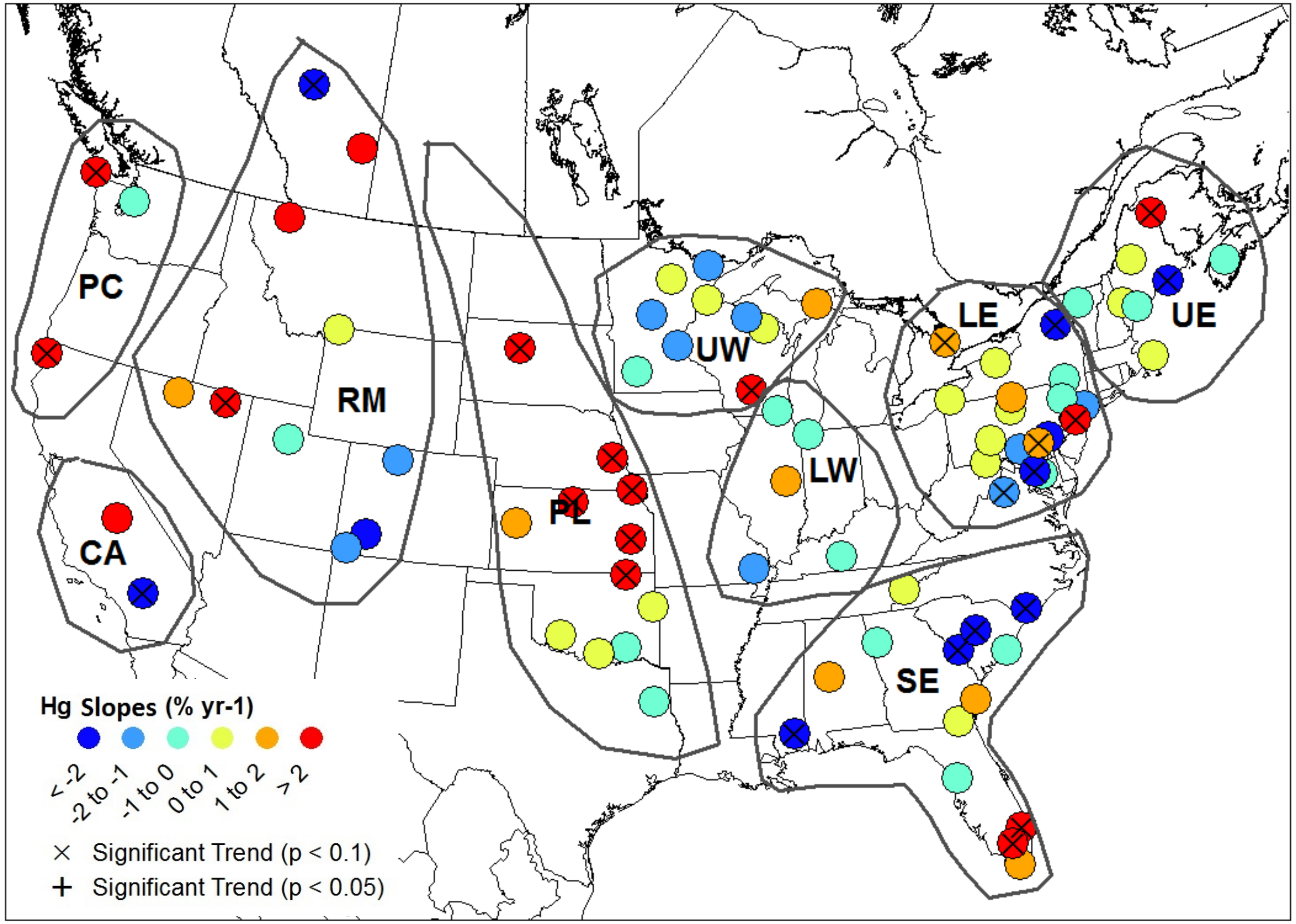
+4.9 %/yr

+2.7 %/yr



Times when PWM residual Hg concentrations were elevated

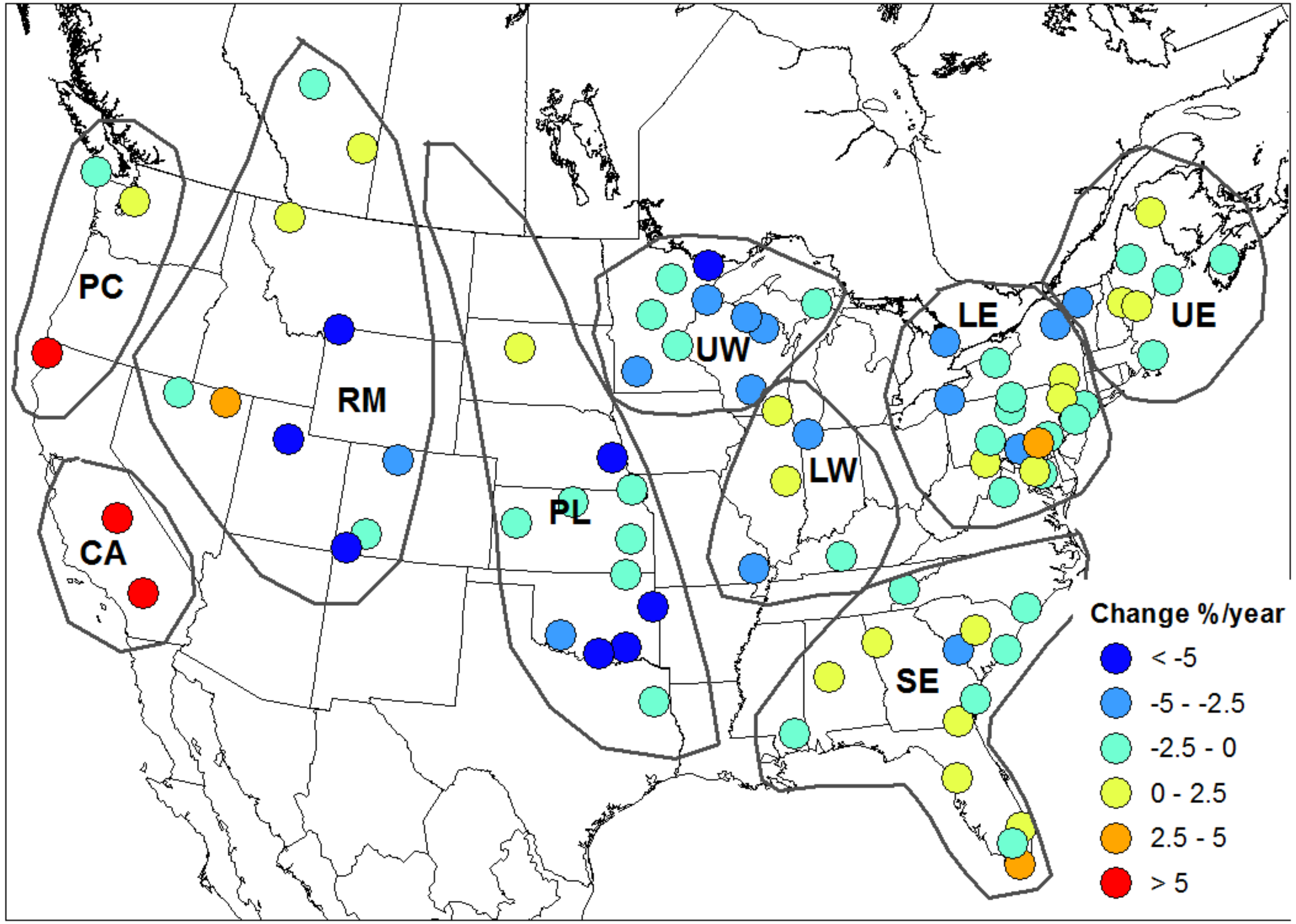
Update: Hg concentration trends* on 2008-2015 MDN data



*SMK non-adjust method

- 12%/19% of the sites had negative/positive trends in Hg concentration in precipitation.
- Compare with 2008-2013: 6%/30% of sites had significant negative/positive trends.
- However, the western half of the continent still contained the majority of sites with annual rates of change > +2% per year (12 out of 27) compared to the sites in the East (5 out of 57).

Percent change in the 2008-2015 slopes vs. the 2008-2013 slopes

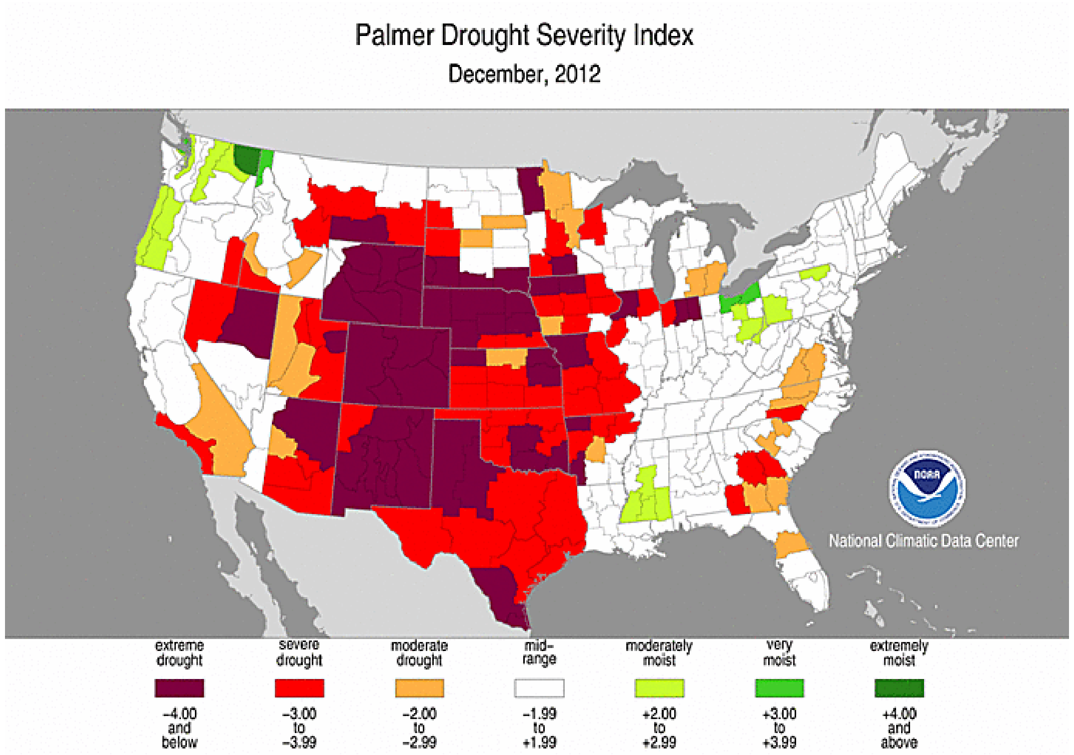


Hot colors indicate the slopes for the later term were more positive/less negative than for the earlier term

Positive trends seen in the early term in most regions appear to have been caused by a temporary phenomenon

The relationship between Hg concentration in precipitation and the Palmer Drought Severity Index (PSDI)

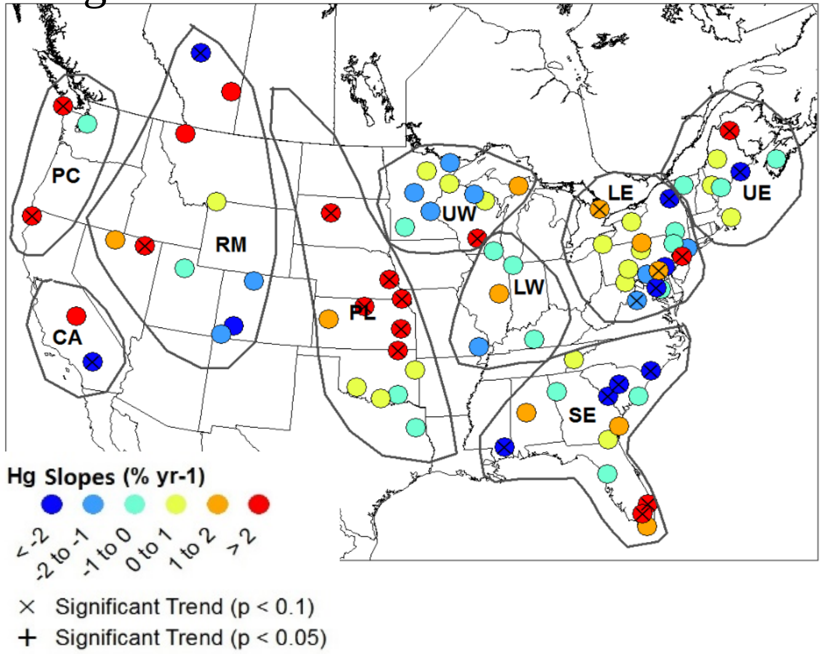
- The PDSI was originally developed by *Palmer* (1965) with the intent to measure the cumulative departure in surface water balance.
- It incorporates antecedent and current moisture supply and demand into a hydrological accounting system.
- The PDSI ranges from about -10 (dry) to +10 (wet) with values below -3 representing severe to extreme drought.
- The PDSI depicts droughts on time scales greater than 12 months.
- The PDSI was designed to be strongly auto-correlated in order to account for the impact of land memory on drought conditions.
- PDSI monthly values are given by county and were aligned with monthly PWM Hg concentrations by the county of the MDN site.

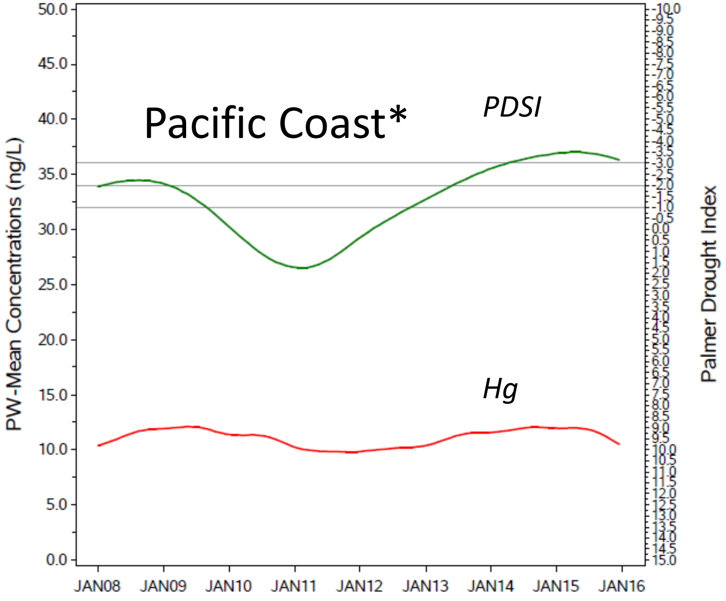


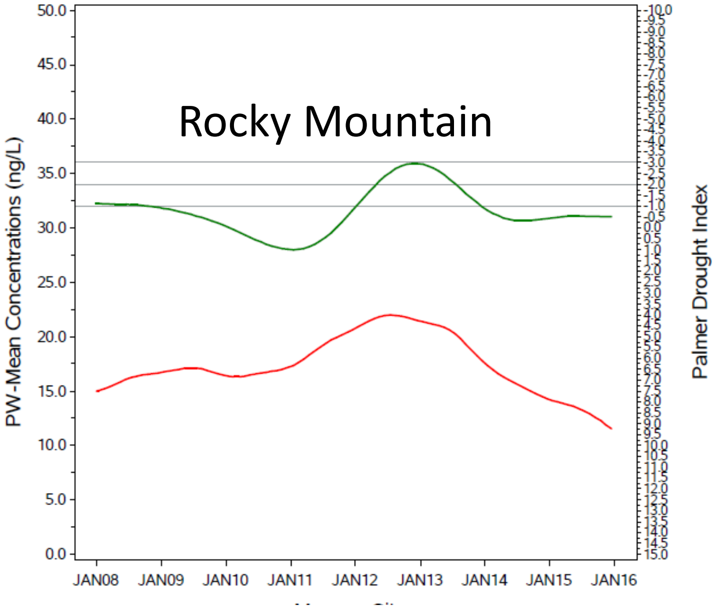
Map corresponds to a time when Hg concentrations were elevated in RM, PL, and UW regions.

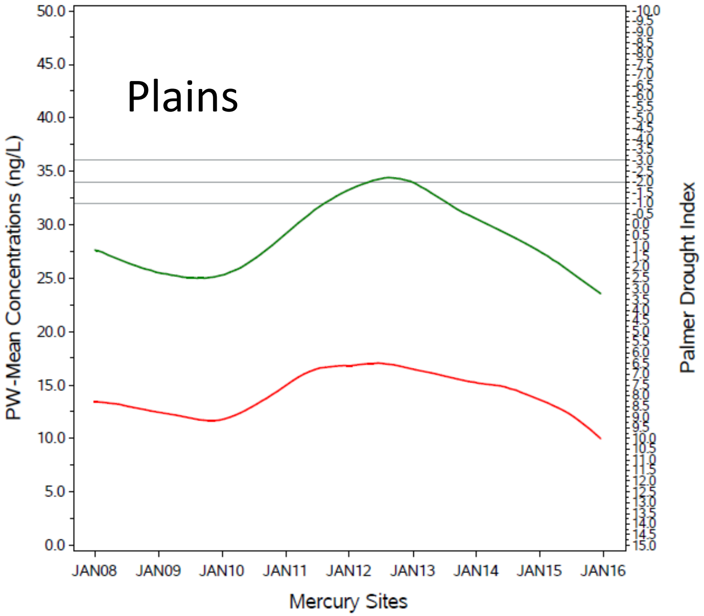
Comparison of mean regional PWM Hg concentration and mean regional PDSI

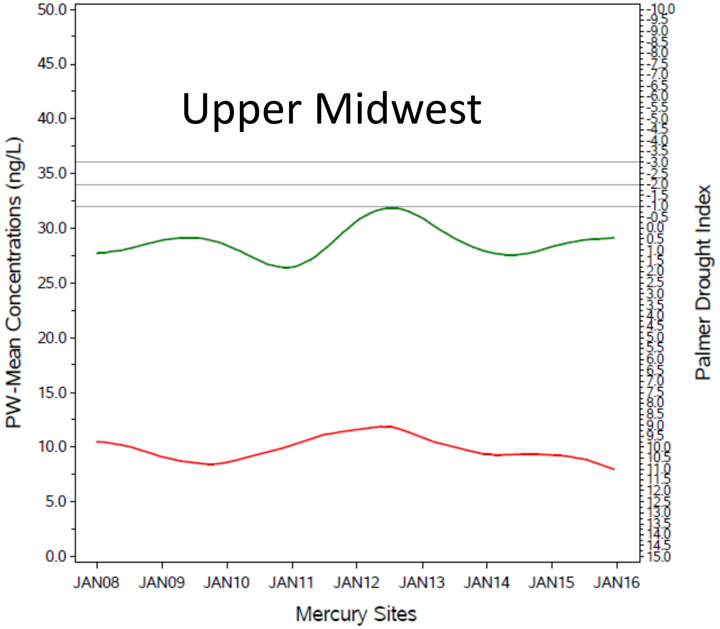
2008-2015 Annual Rates of Change, Hg Concentration MDN sites







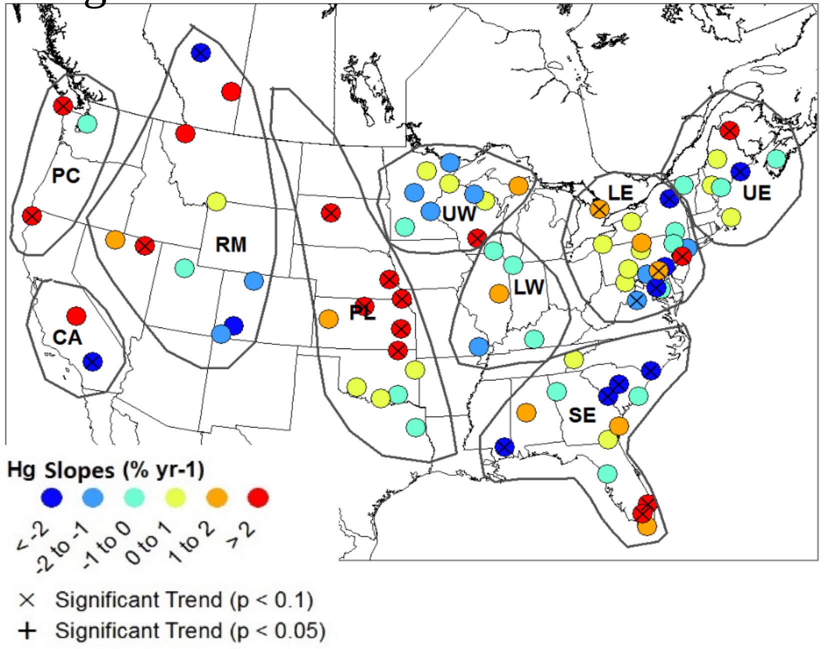


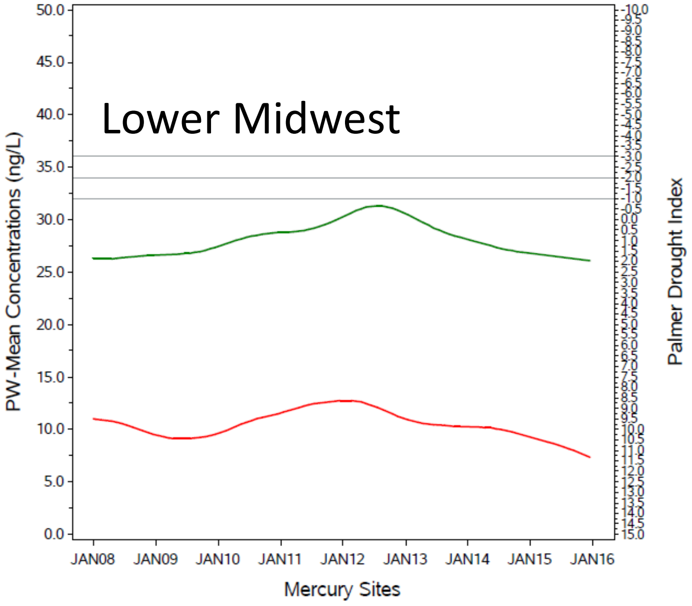


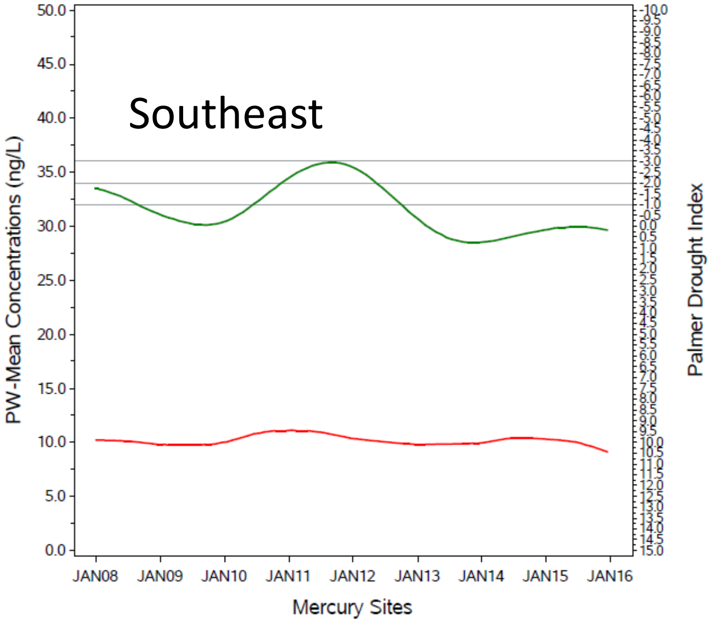
*PC region redefined to include CA region as well

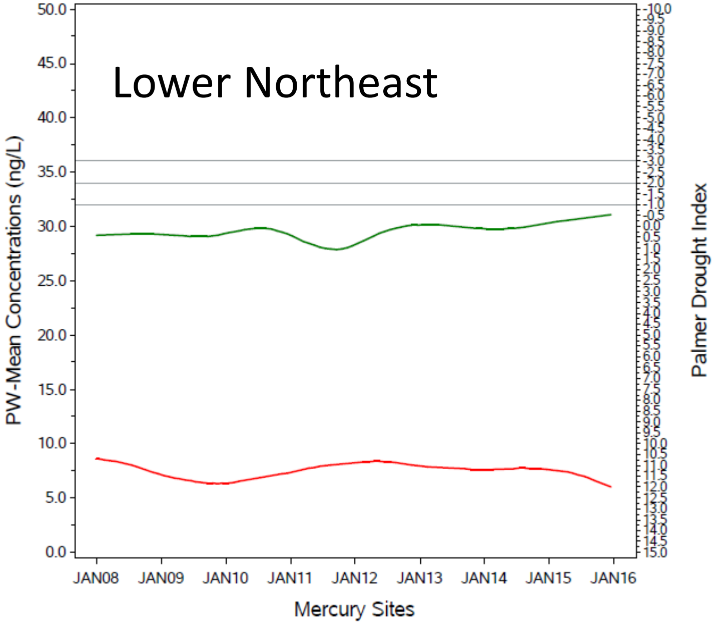
Comparison of mean regional PWM Hg concentration and mean regional PDSI

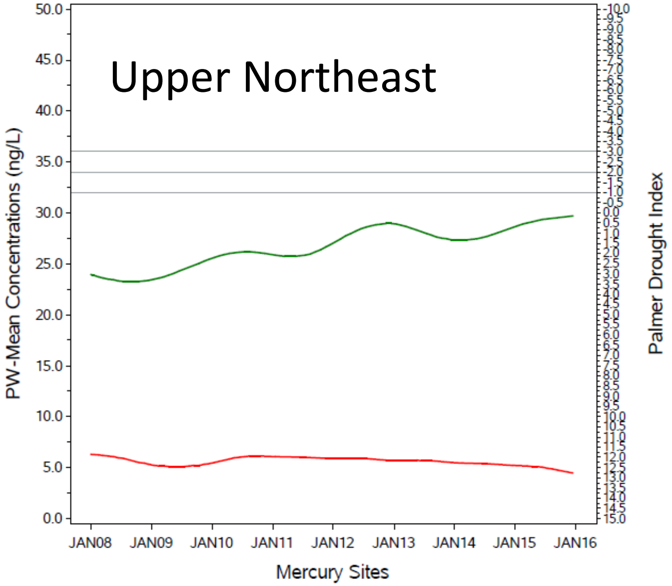
2008-2015 Annual Rates of Change, Hg Concentration MDN sites



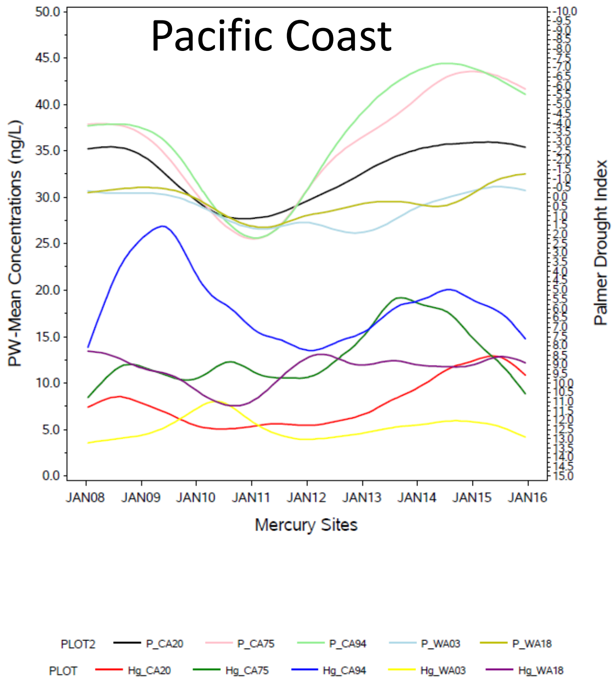


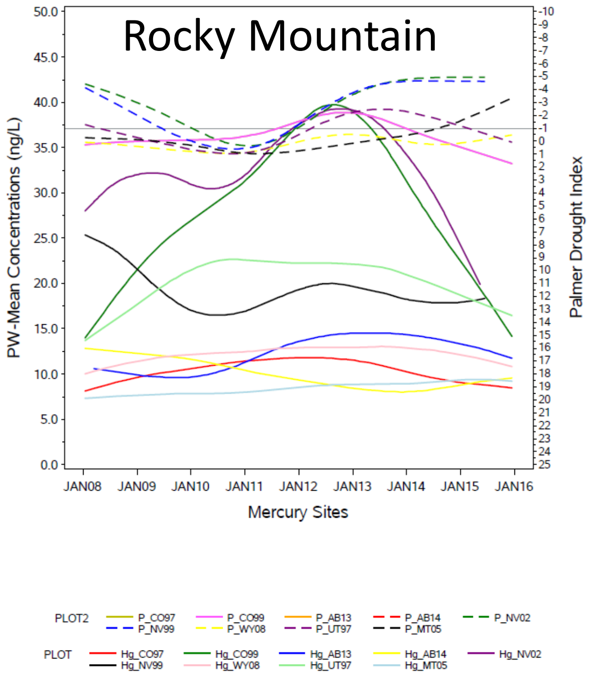


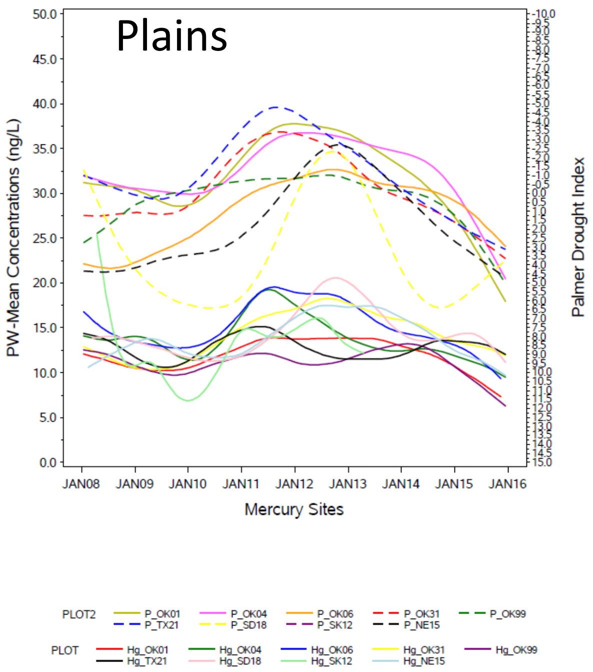


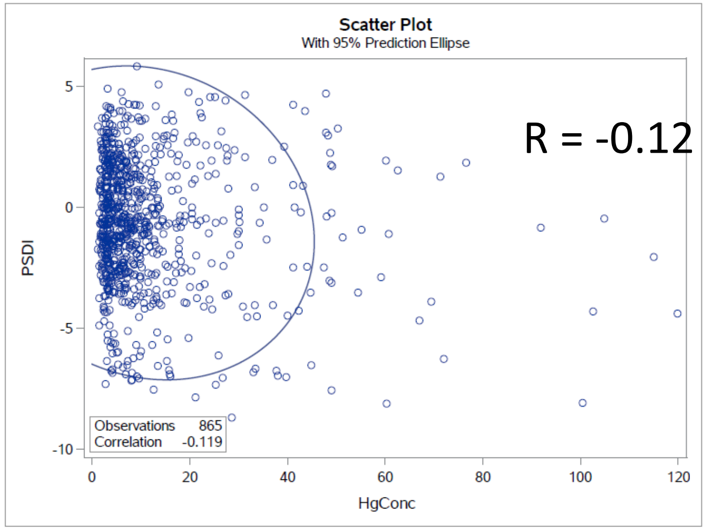


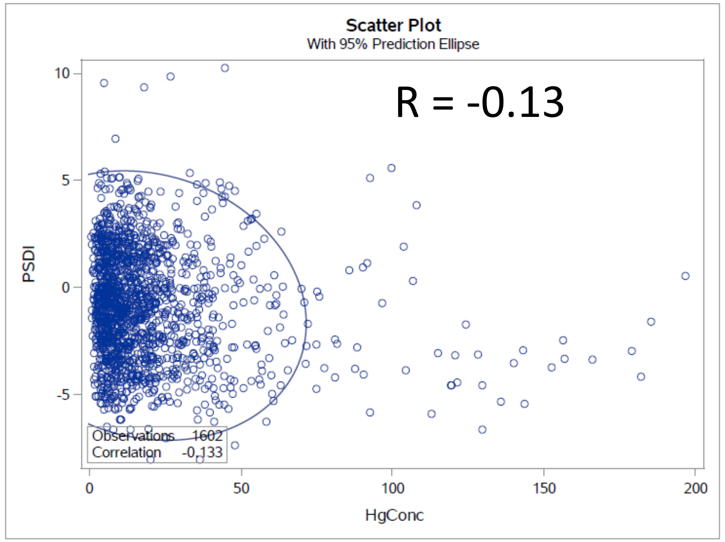
Detail for three regions: PDSI and Hg concentration

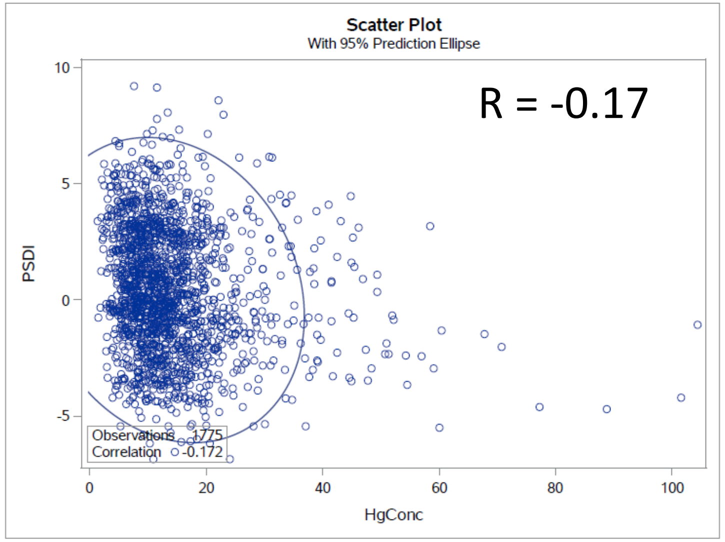












Correlation coefficients for monthly PDSI vs. monthly PWM Hg concentration and for monthly PDSI vs. monthly Hg deposition flux for all sites in each region, 2008-2015

Region	R (concentration)	R (deposition)
Pacific Coast	-0.12	0.19
Rocky Mountain	-0.13	0.11
Plains	-0.17	0.11
Upper Midwest	-0.08	0.20
Lower Midwest	-0.14	0.19
Southeast	-0.12	0.17
Lower Northeast	-0.16	0.15
Upper Northeast	-0.15	0.17

Plains is region with strongest (negative) PDSI correlation with Hg concentration and the weakest (positive) correlation with Hg deposition flux.

Conclusions

- Positive trends in Hg concentration observed in the central and western U.S. during 2008-2013, for the most part did not continue through 2015 indicating a temporary phenomenon.
- We suggest this was due to regional changes in precipitation and temperature, as indicated by the PDSI. We suggest the high Hg concentrationd of 2011-2013 and other positive trends seem to be at least a function of the drought.
- The Plains region showed the strongest correlation between Hg concentrations and PDSI and the weakest correlation between Hg deposition and PDSI.
- The cause of the correlation between Hg concentration and PDSI may be simply evidence of the washout effect or may indicate a different balance of sources and sinks under drought vs. normal conditions. Modeling may help answer these questions.
- Next: more work to do, other drought indexes, perhaps finer resolution, compare with global Hg models.

Acknowledgements

- Arnout ter Schure and Leonard Levin – Electric Power Research Institute
- Mark Brigham – US Geological Survey
- Matt Parsons – Environment Canada
- Mae Gustin – University of Nevada, Reno

Environment Canada

Thank you!!

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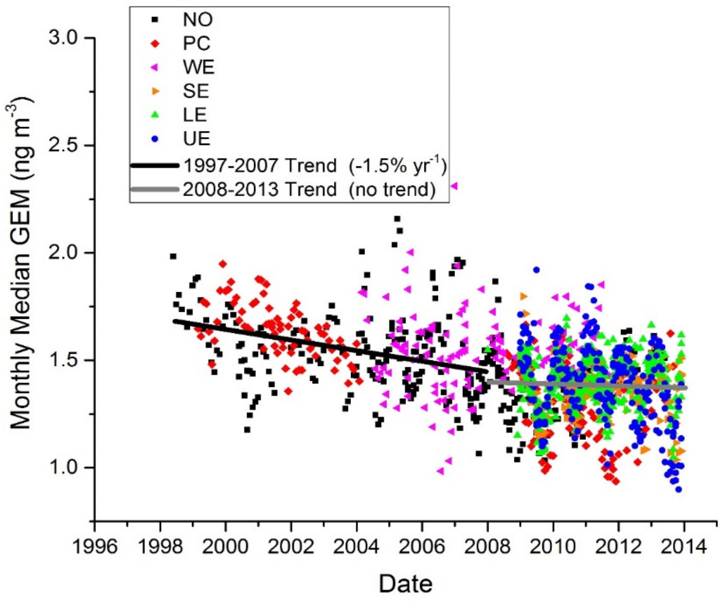
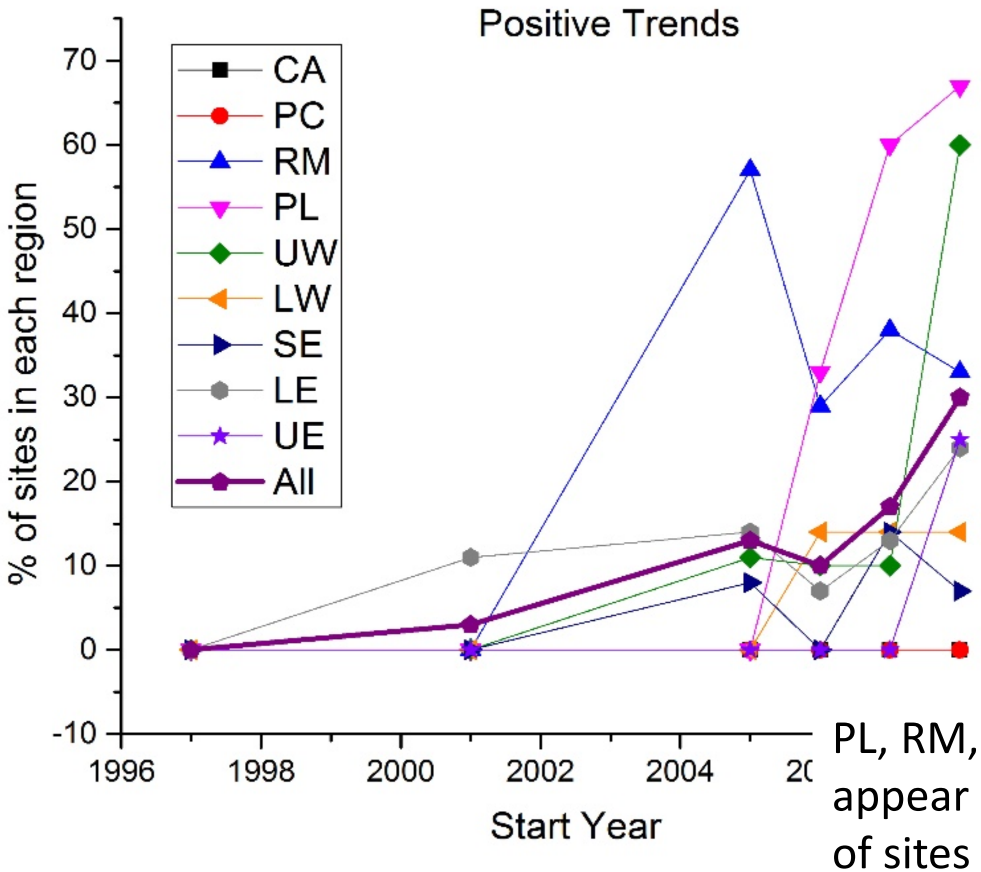
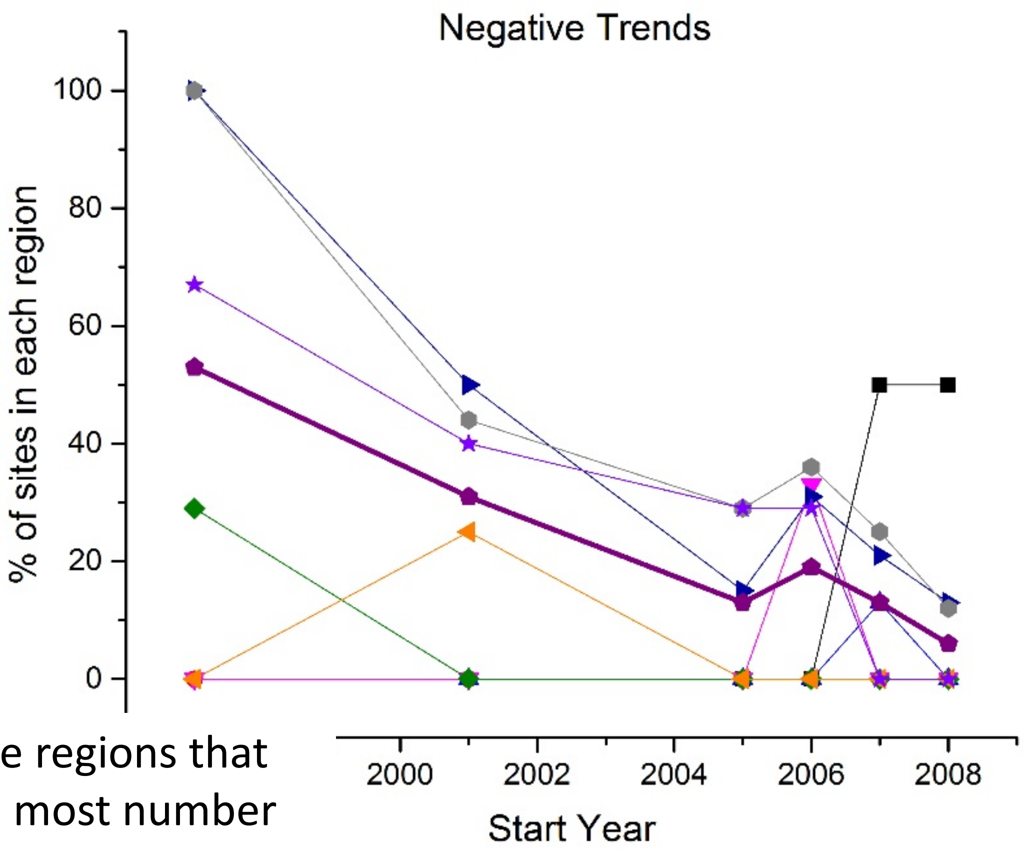
Extra slides



Percentage of sites in each region that displayed positive or negative trends ($p<0.1$) considering data with an end year of 2013





PL, RM, and UW are regions that appear to have the most number of sites with positive trends