Mercury Deposition in the Great Lakes Region

Angela Dickens
LADCO Data Scientist

MPCA 2023 Statewide Mercury TMDL Meeting
September 21, 2023

LADCO | LAKE MICHIGAN AIR DIRECTORS CONSORTIUM

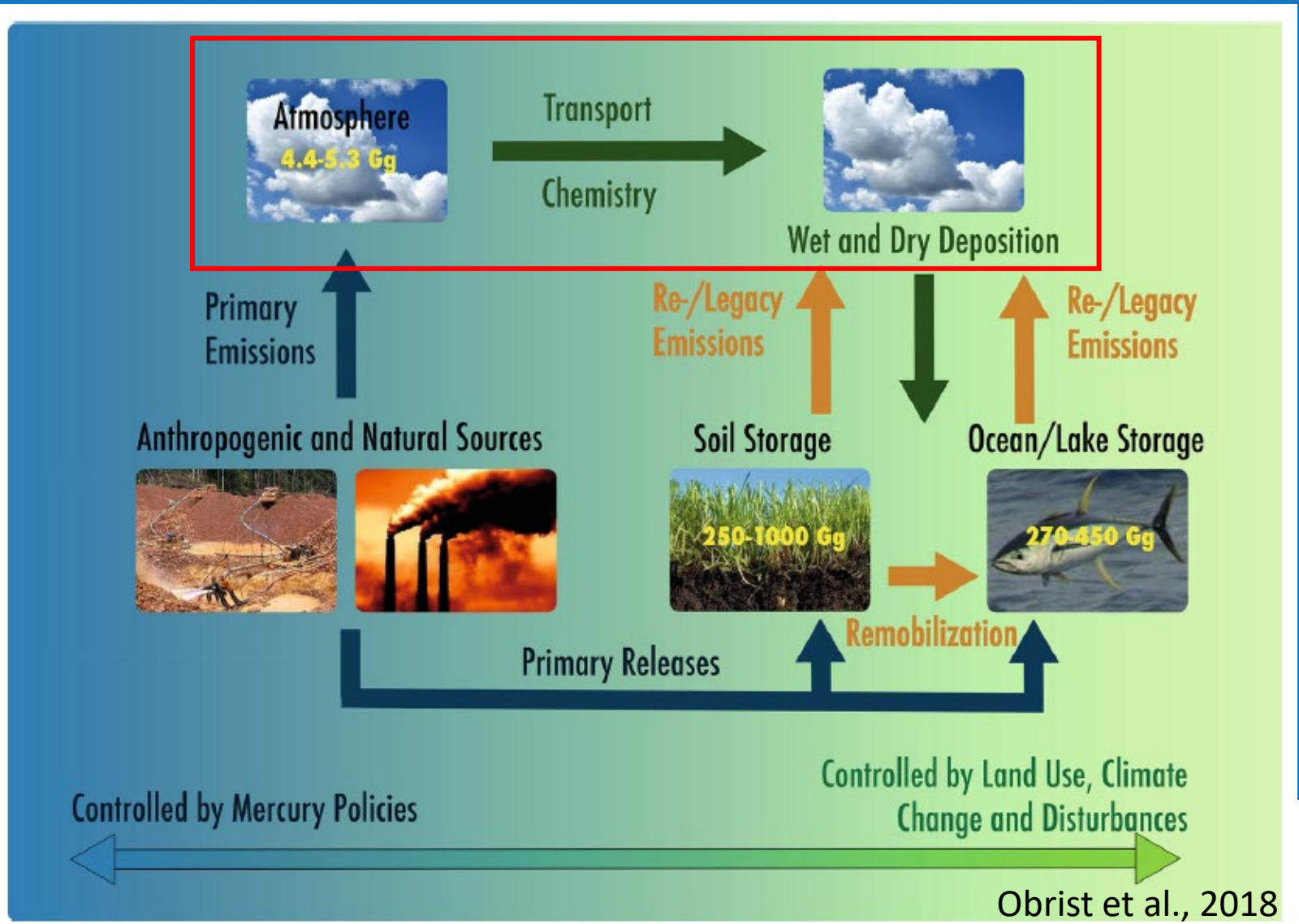
LADCO Report on Mercury

- Technical report: Mercury Deposition in the Great Lakes
  - Released June 2023
  - https://www.ladco.org/wp-content/uploads/Projects/Mercury/Mercury-deposition-in-the-Great-Lakes-Report-2023_FINAL-CLEAN.pdf
- Examines amounts and trends in wet and dry (litterfall) deposition of mercury in the Great Lakes states
  - MN, WI, MI, IL, IN, OH
  - Also looks at emissions trends and trends in atmospheric concentrations (where available)
  - Based on data from the National Atmospheric Deposition Program (NADP)
  - Interprets data using published research studies

Outline

- The mercury cycle and monitoring networks
- Mercury emissions trends
- Atmospheric mercury concentrations
- Mercury deposition trends
  - Wet deposition
  - Dry deposition (litterfall)
- Insights into sources of mercury in the region

The Mercury Cycle

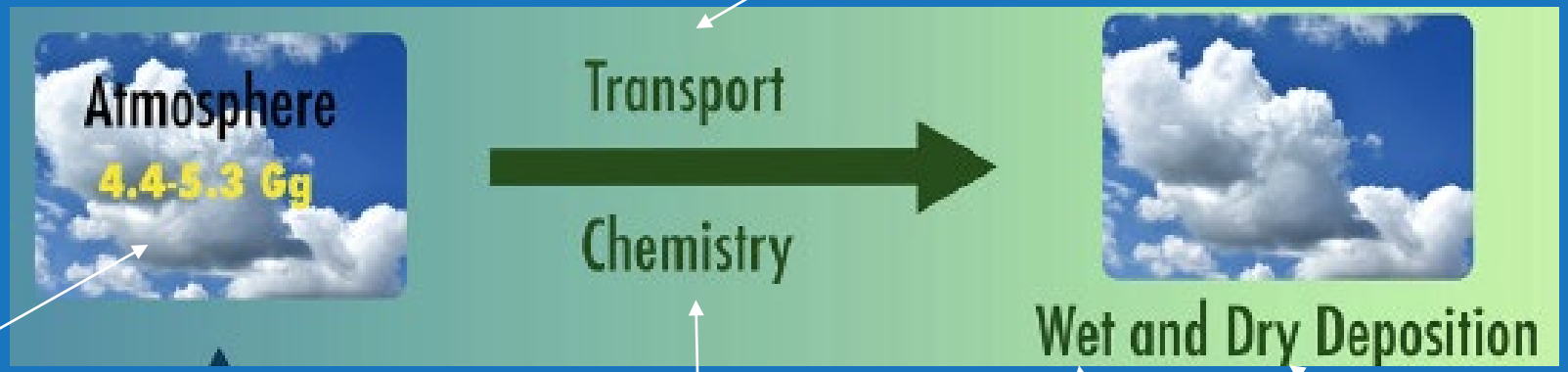


Obrist et al., 2018

LADCO

The Mercury Cycle

Local, regional, continental, global



Gaseous elemental mercury (GEM)
Gaseous oxidized mercury (GOM)
Particle-bound mercury (PBM)

GEM ↔ GOM
GOM dissolves in water
GOM ↔ PBM

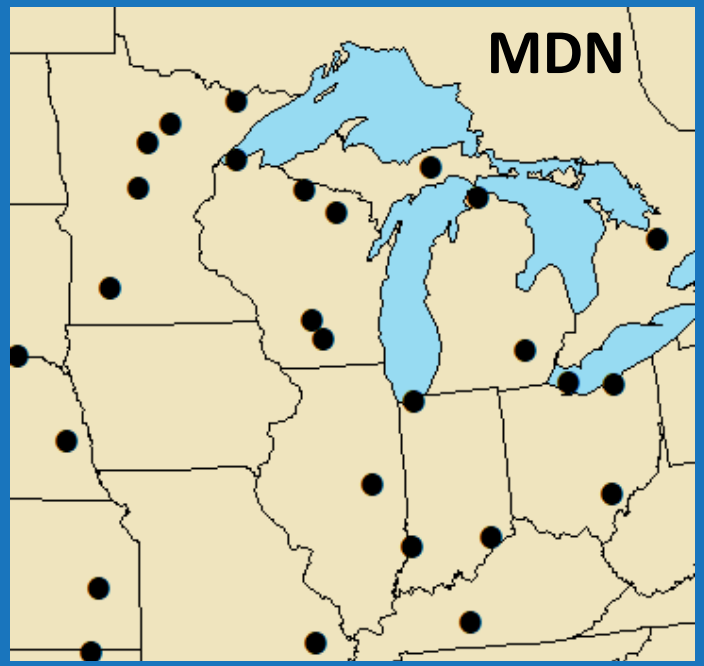
Wet deposition
- Mostly GOM & PBM

Dry deposition
- Mostly GEM
- 75% via litterfall

Obrist et al., 2018

LADCO

National Atmospheric Deposition Network (NADP) Sites

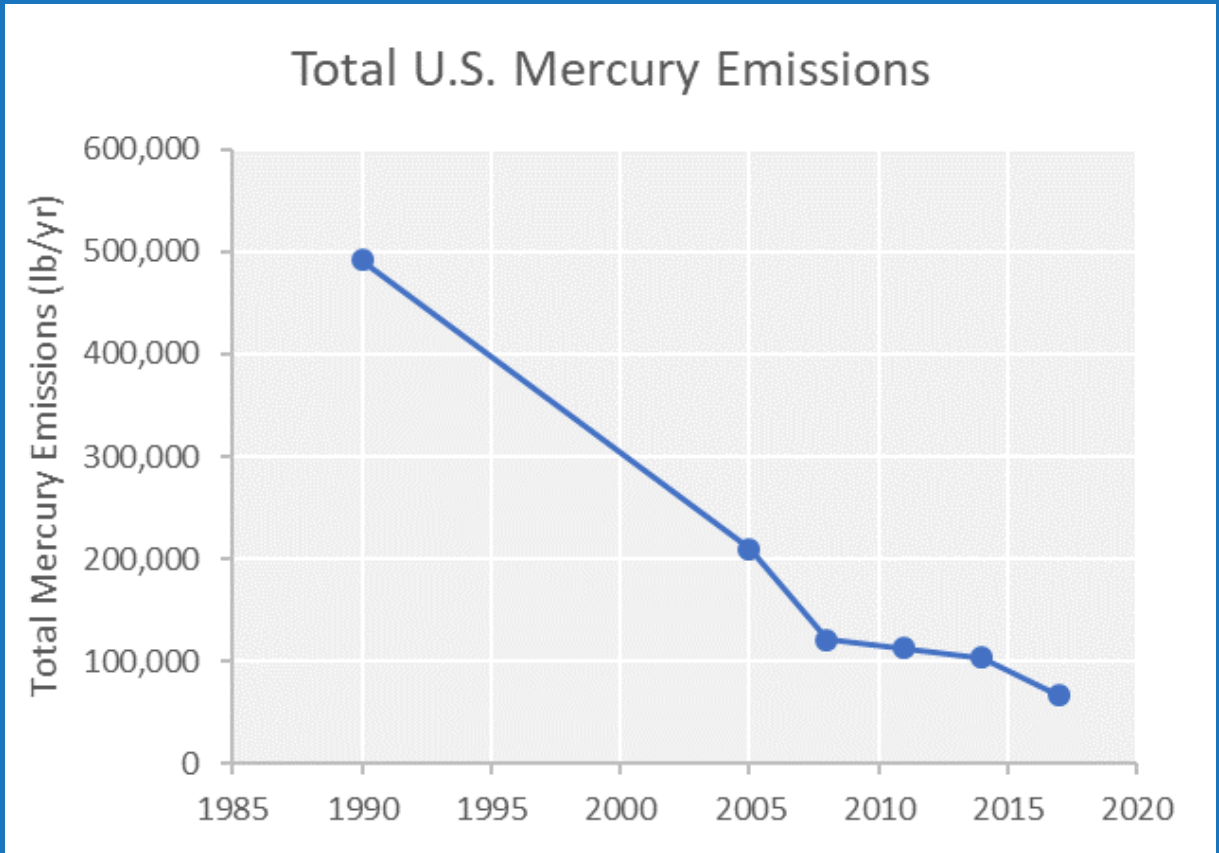


Mercury Deposition Network
- Measures wet deposition
- Most extensive network
- Longest record

Mercury Litterfall Network
- Measures dry deposition
- Intermediate coverage

Atmospheric Mercury Network
- Measures gaseous or particulate forms
- Very sparse network

Mercury Emissions



Decreased by 87% from U.S. sources

- Reductions from a variety of sources, particularly:
  - Chlor-alkali plants
  - Coal combustion

Global emissions trends are less certain

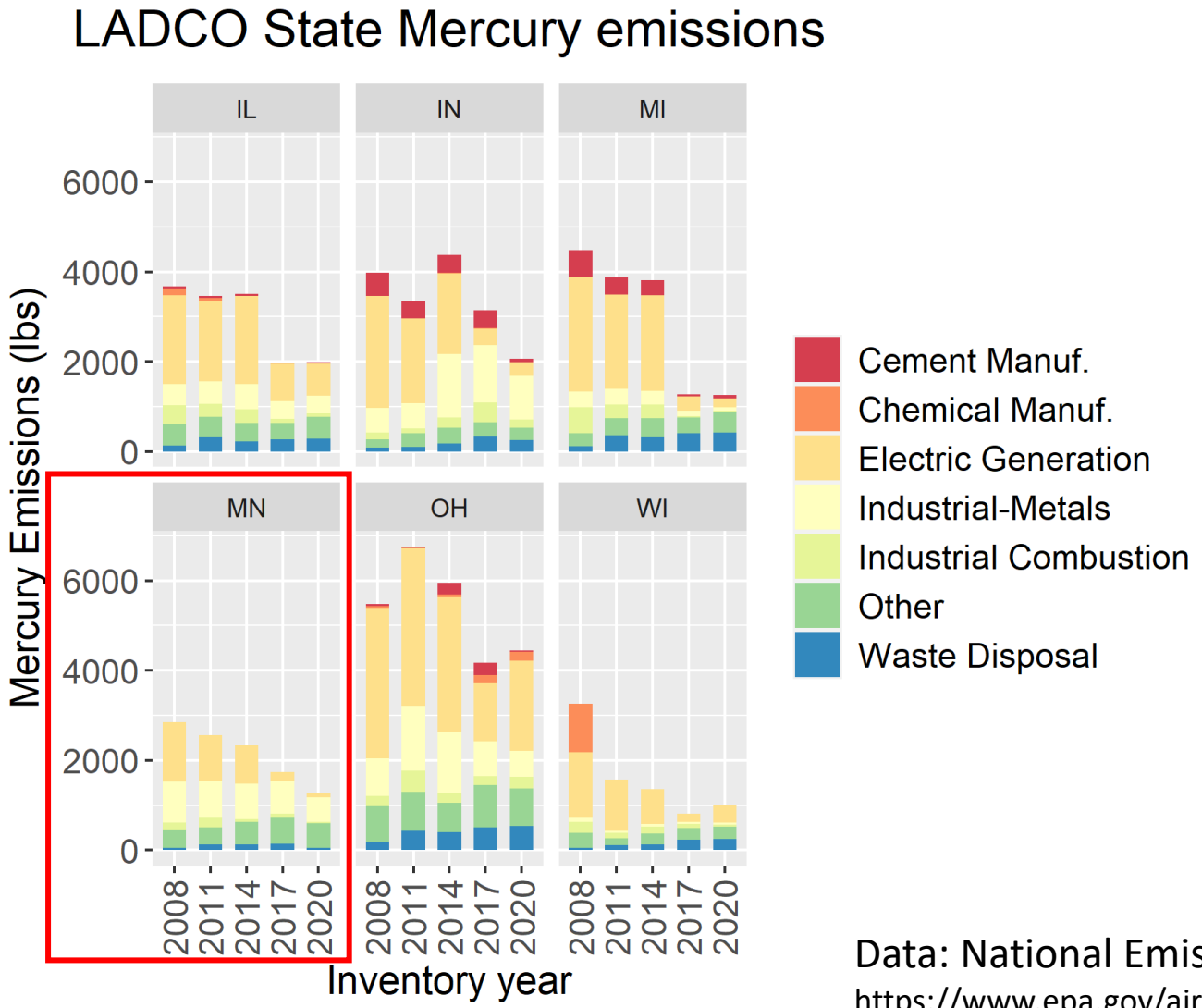
- Likely increased at least through 2013
- No consensus on direction or magnitude

Data: National Emissions Inventory (NEI) as reported in Brigham et al. (2021)

4415 West Harrison St., Suite 548
Hillside, IL 60162

LADCO

Mercury Emissions



Reductions of 19% (OH) to 72% (MI) since 2008

- Largest reductions from Electricity Generation

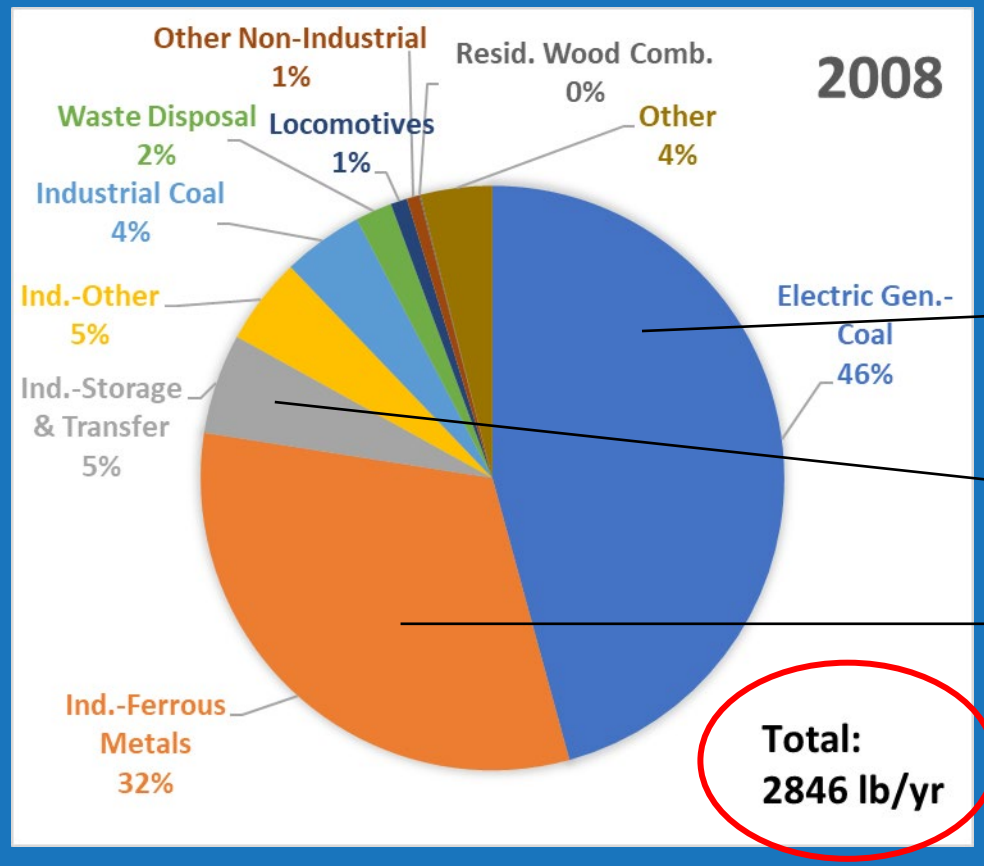
Minnesota: 55% reductions

- Most reductions from Electricity Generation

Data: National Emissions Inventory (NEI)
https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei

LADCO

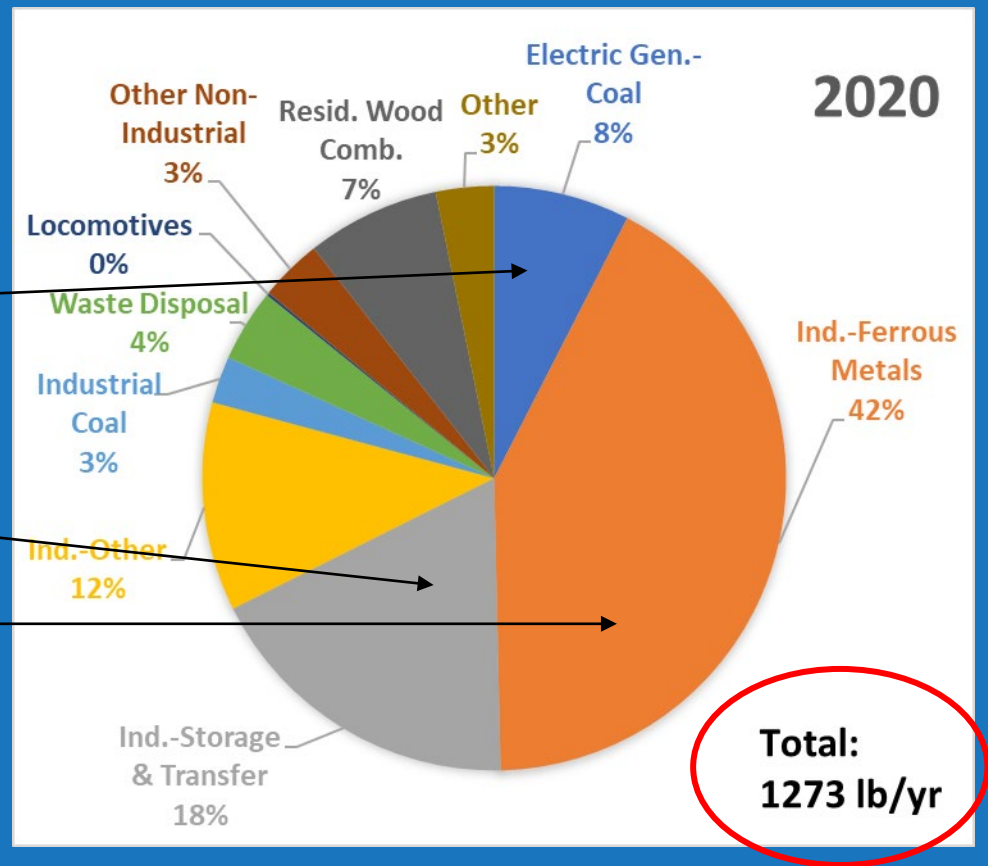
Mercury Emissions from MN Sources



- 93%

+ 45%

- 41%



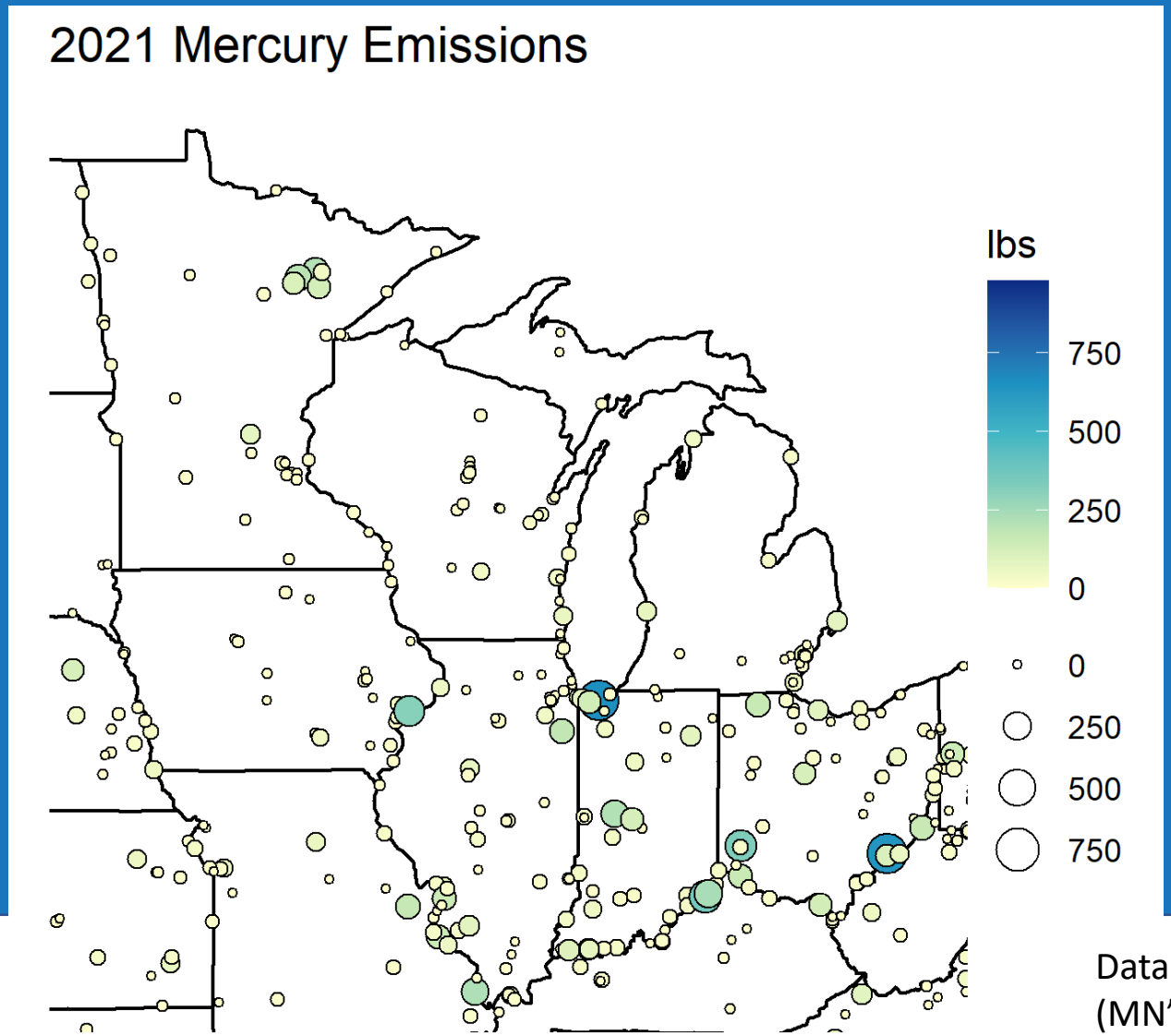
Largest source: electricity generation with coal → Largest source: ferrous metals industry

4415 West Harrison St., Suite 548
Hillside, IL 60162

Data: National Emissions Inventory (NEI)
https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei

LADCO

Mercury Emissions from Point Sources



Almost all large* sources in the region are in the metals industry:

- Steel plants
- Other metal processing facilities (Mn & Al)
- Taconite facilities
- (One coking plant)

Electricity generating units have lower emissions as a result of regulations and shutdowns

**Large sources emitted >100 lb Hg in 2021*

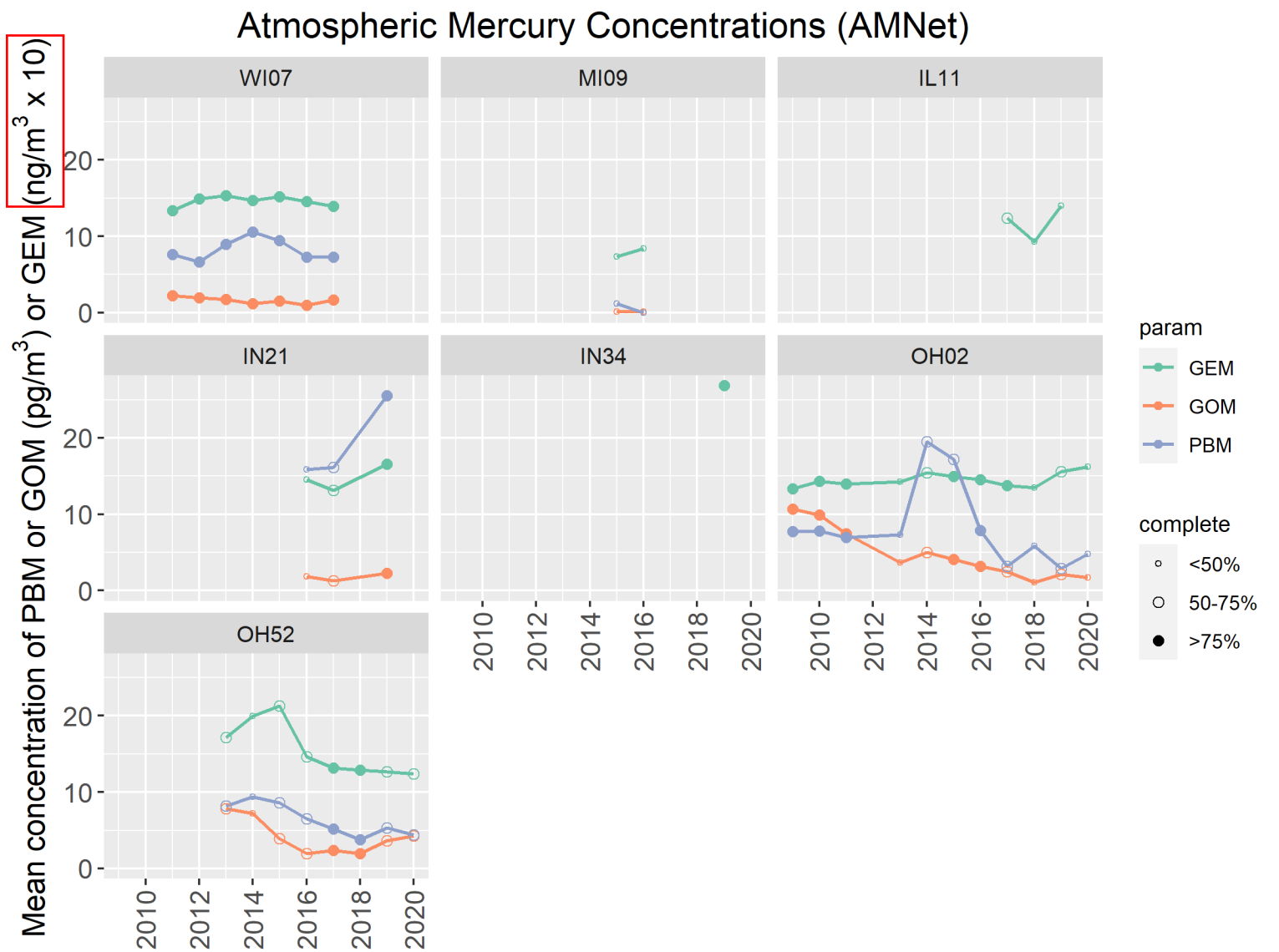
Data: EPA's Toxics Release Inventory except for MN (MN's point source air emissions inventory)

LADCO

Atmospheric Concentrations of Mercury

- GEM, GOM, and PBM
- Very sparse data in space and time
- Many years have incomplete data → Less representative

Atmospheric Concentrations of Mercury



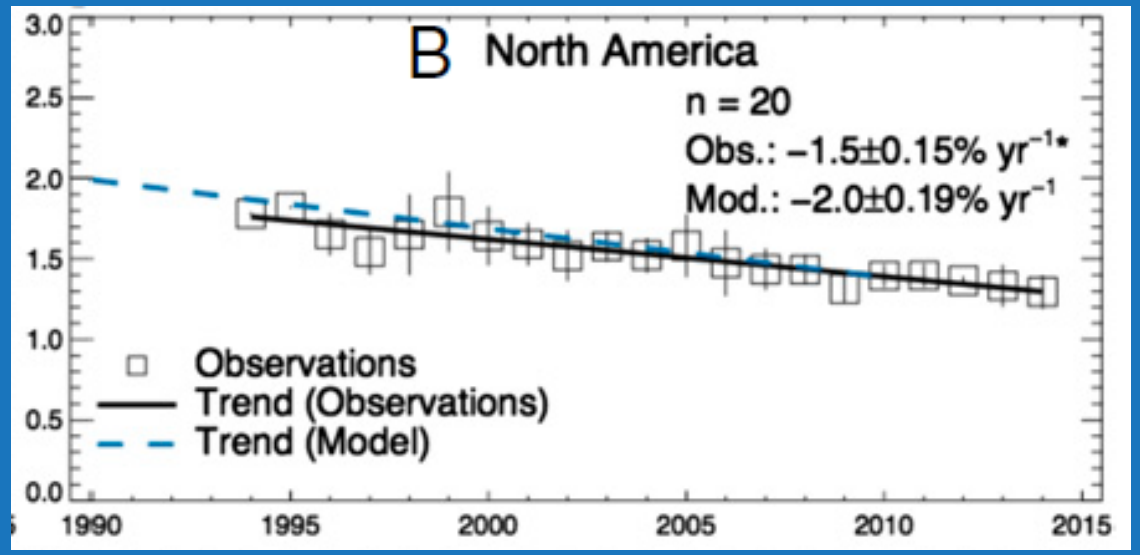
- GEM is >100 x as abundant as GOM or PBM (nanograms vs picograms)

- Focus on sites with more complete data
- GEM similar at all sites with no obvious trends
- GOM lower in Wisconsin (WI07) than in Ohio (OH02)
  - GOM seems to be decreasing at both sites
- PBM: no clear spatial or temporal trends

LADCO

Atmospheric Concentrations of Mercury

Published GEM Trends

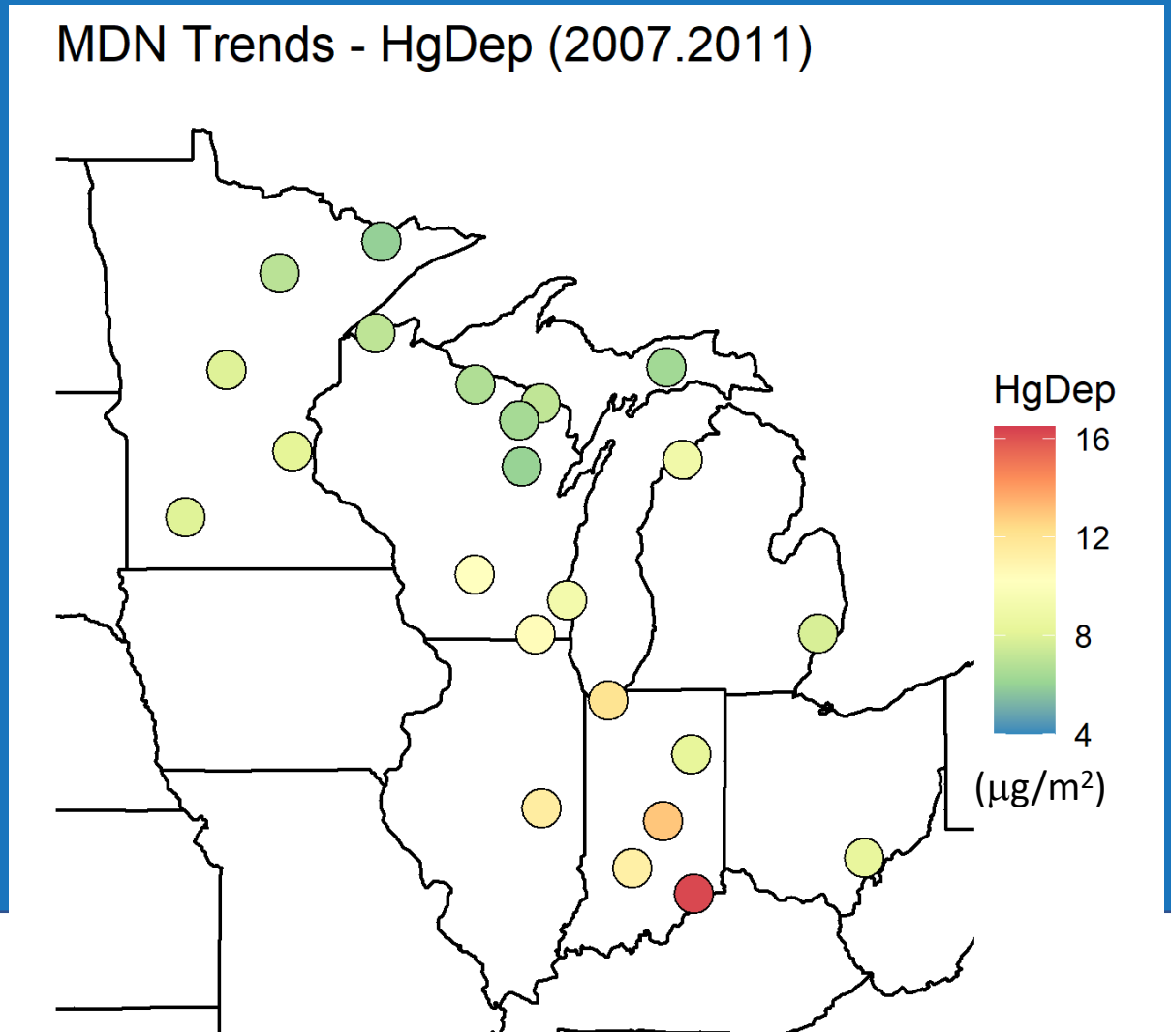


Zhang et al. (2016)

In contrast:

- North American GEM decreased 1.2 to 2.1% per year from 1990 to 2013 (Zhang et al., 2016)
  - May not see this in the Great Lakes region because decreases have slowed or because of the sparsity of sampling sites
- Atmospheric mercury concentrations have been increasing in East Asia (Obrist et al., 2018)

Wet Deposition of Mercury



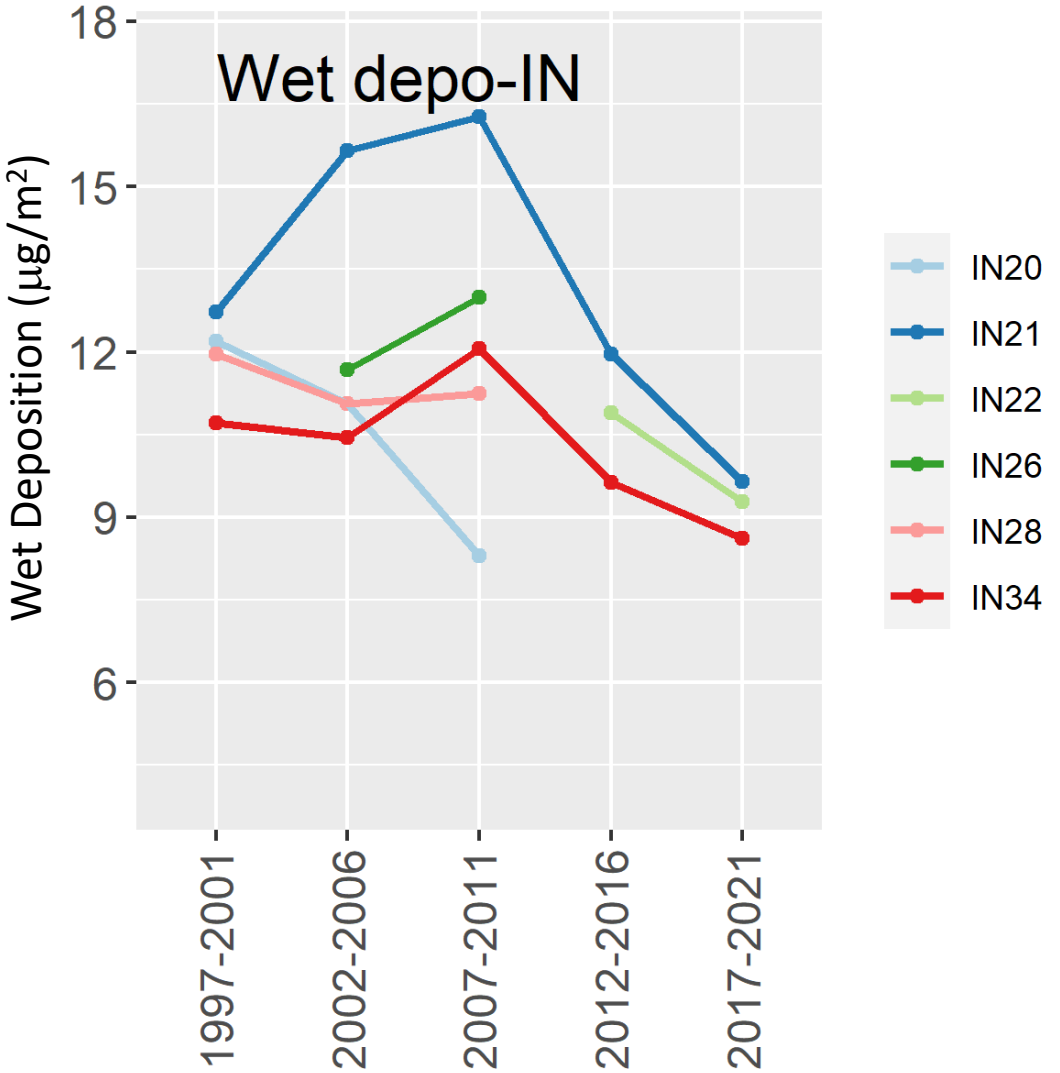
Wet deposition is greater in the southern part of the region

- Likely due to greater precipitation in southern areas (Risch and Kenski, 2018)
- Also: larger point sources of mercury in the southern states

Variation within the region suggests a role for local and regional emissions sources, as well as global emissions

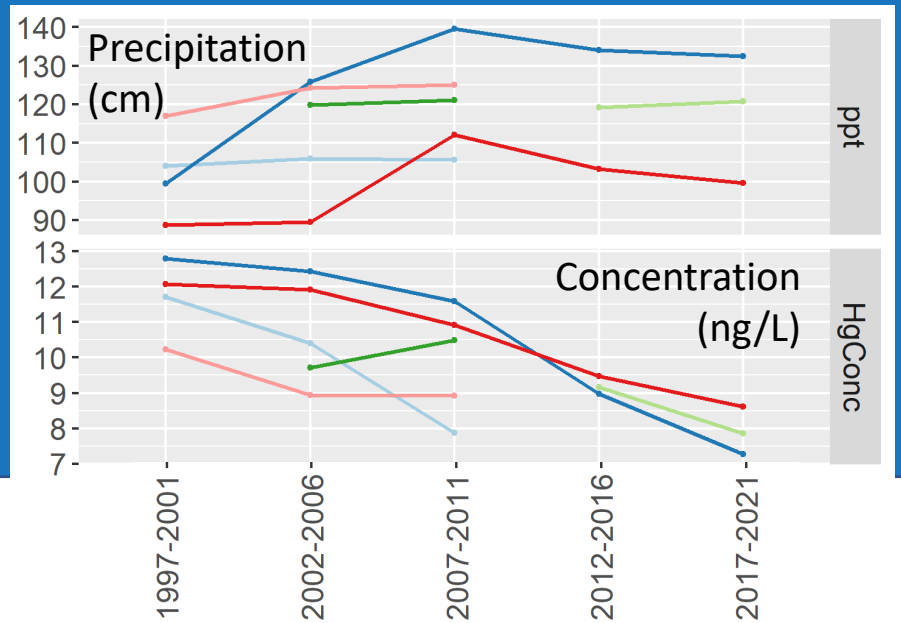
LADCO

Wet Deposition of Mercury



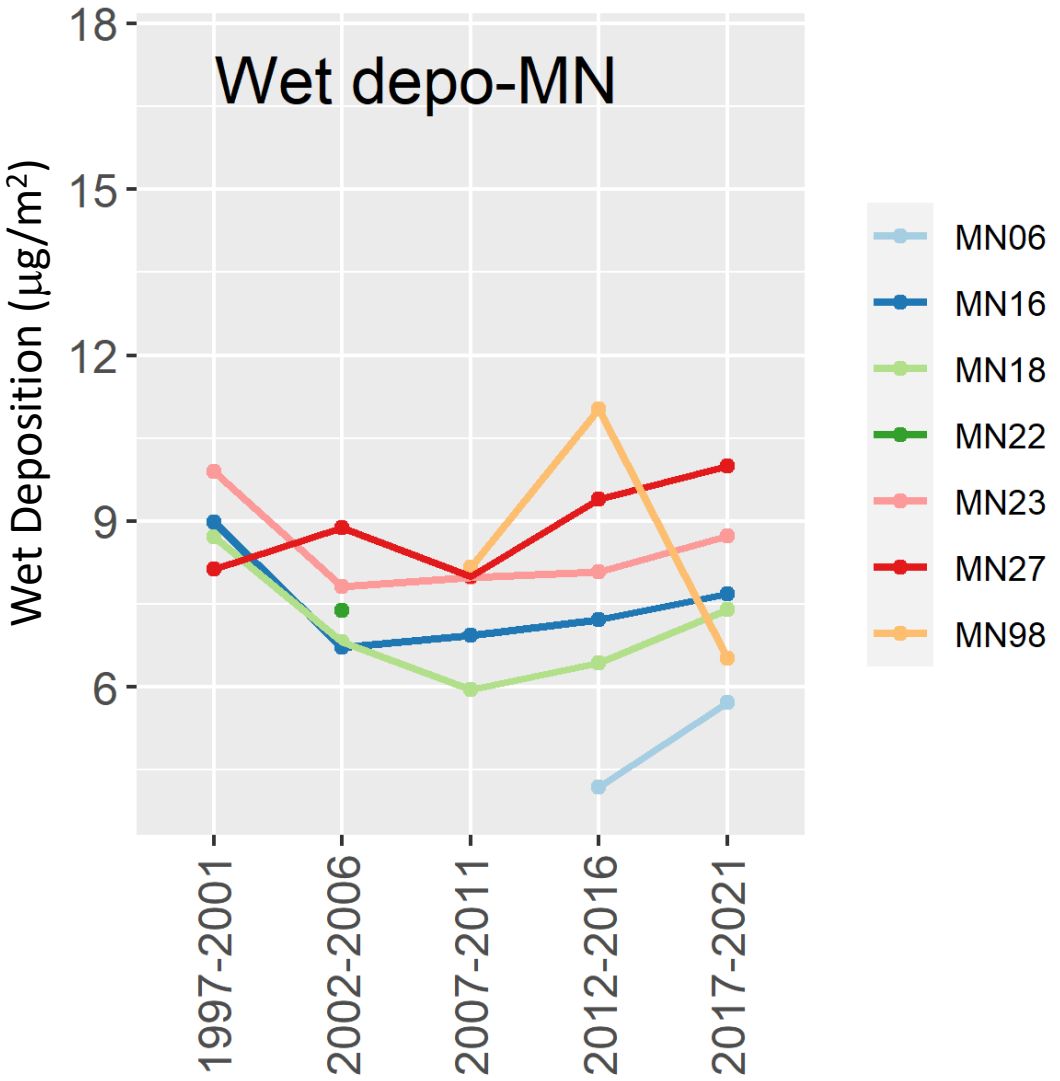
<u>Southern states</u> (IN as example):

- Wet deposition has been decreasing for at least the last 15 years
  - Largest reductions in the Ohio River Valley (IN21)
  - Steady reductions in mercury concentrations and unclear trends in precipitation
  - Mercury concentration reductions appear to be driving deposition decreases
    - Likely due to decreased local and regional emissions



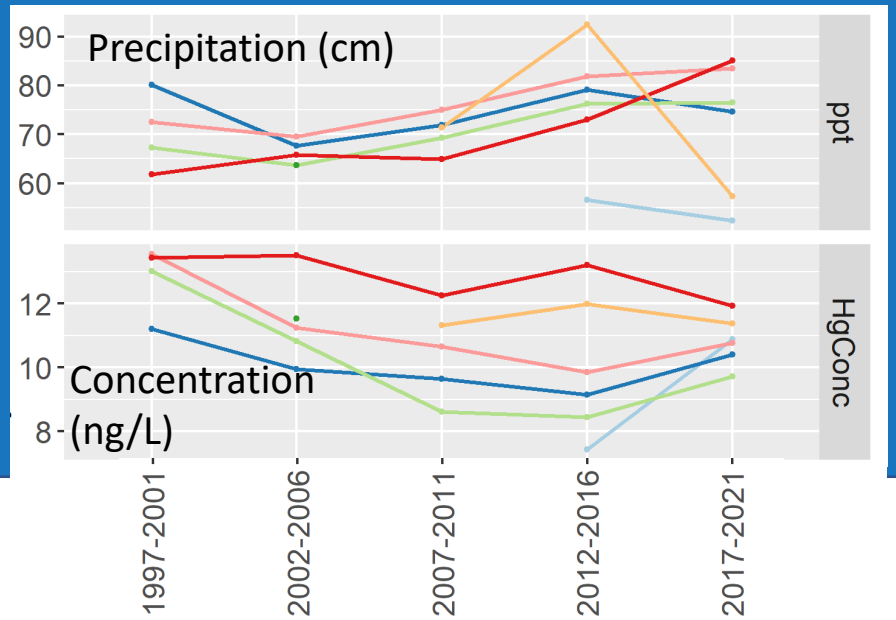
LADCO

Wet Deposition of Mercury



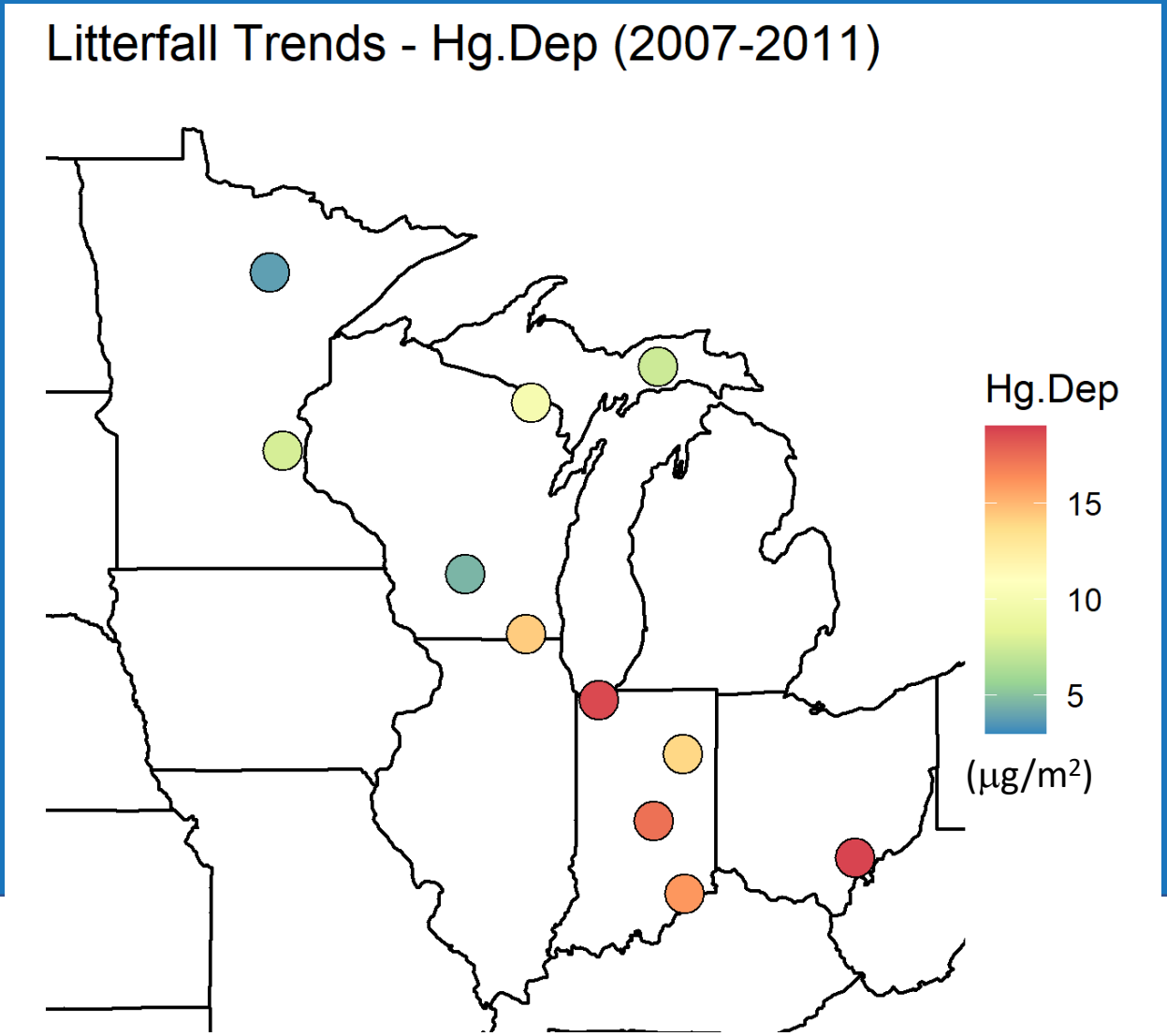
Northern states (MN as example):

- Wet deposition is flat to increasing
  - Increasing most consistently in MN
  - Mercury rainwater concentrations have mostly decreased but not as clearly as in the south
  - Precipitation has increased
    - Increased deposition likely primarily due to increased precipitation
  - Contrasted with earlier decreases at these sites



LADCO

Dry (Litterfall) Deposition of Mercury

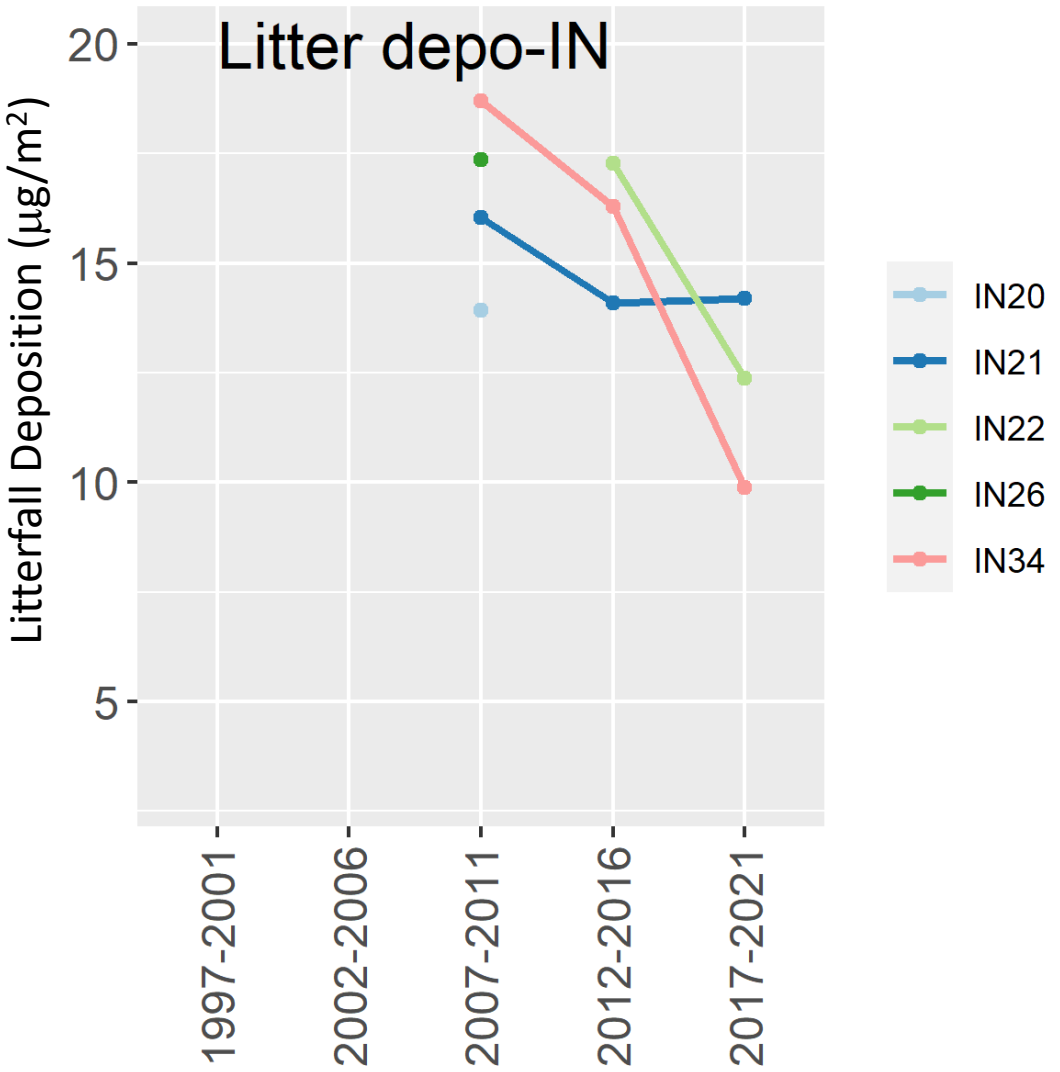


Dry deposition is greater in the southern part of the region

- Similar patterns to wet deposition

LADCO

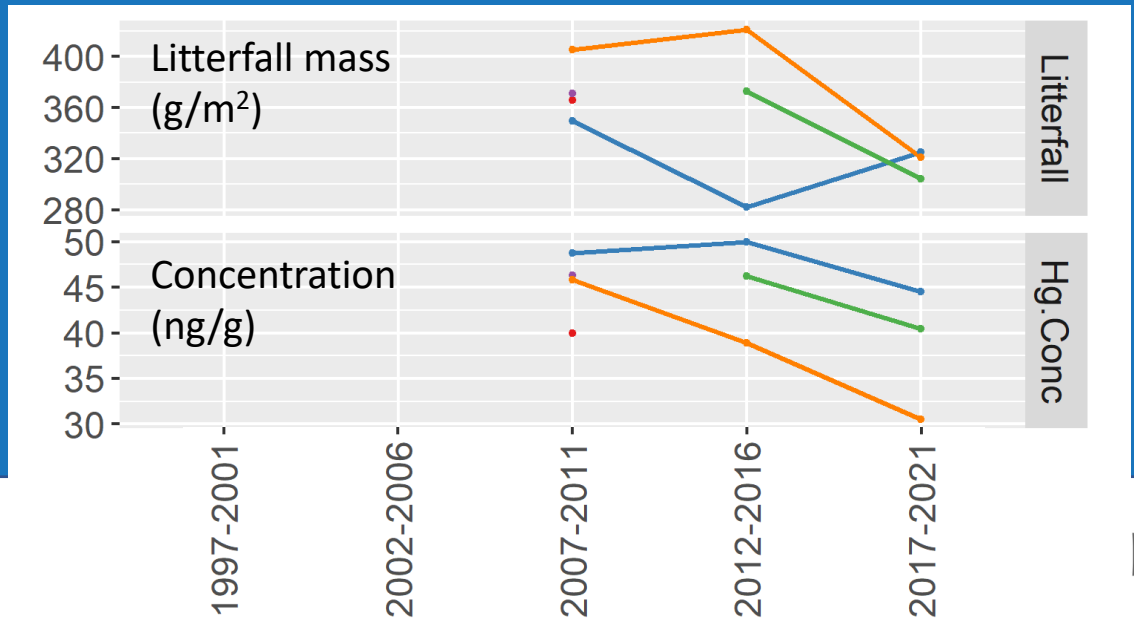
Dry (Litterfall) Deposition of Mercury



Shorter and less complete records than for wet deposition

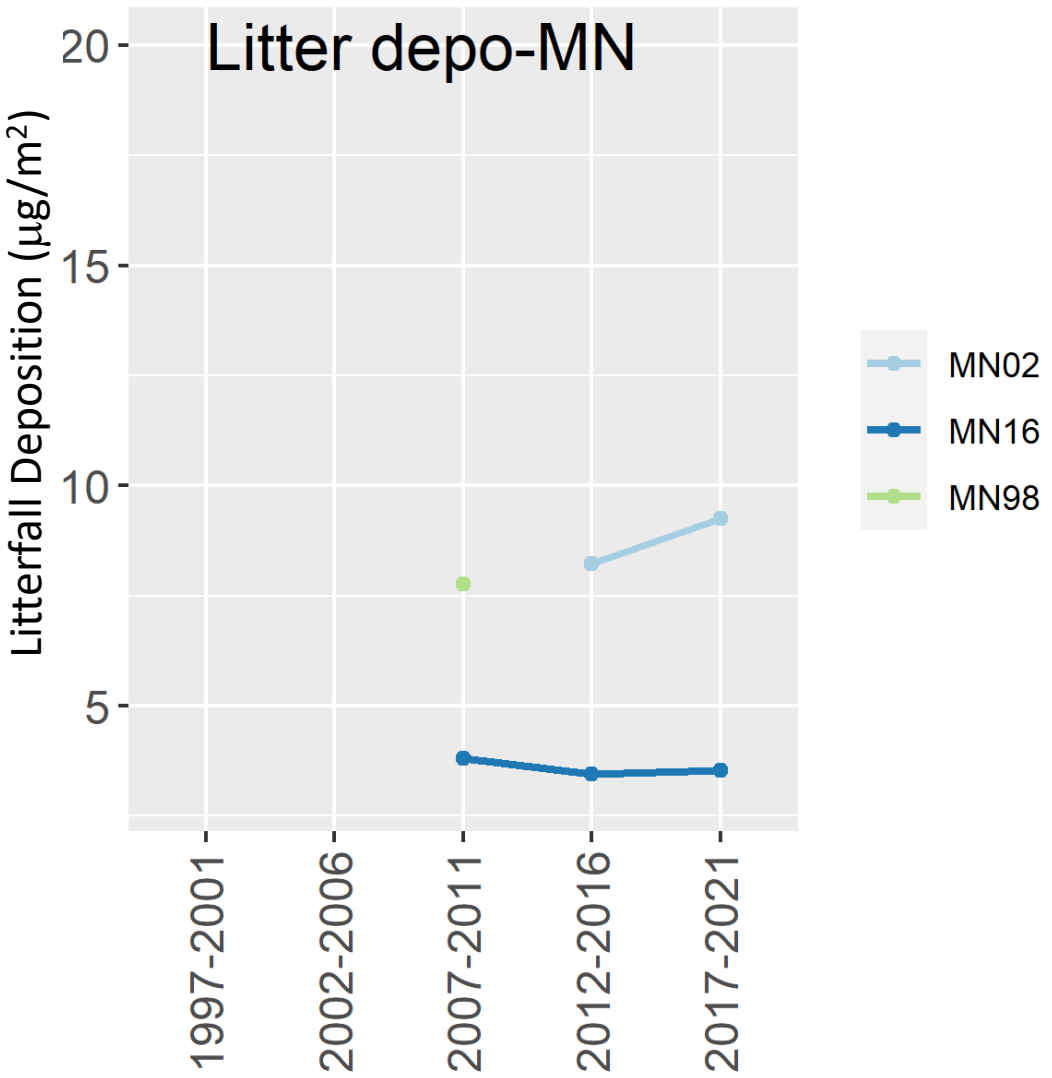
<u>Southern states</u> (IN as example):

- Clear decreases in litterfall deposition
  - Mercury concentrations decreased
  - Litterfall mass also decreased at some sites
  - Likely driven by decreased local/regional emissions



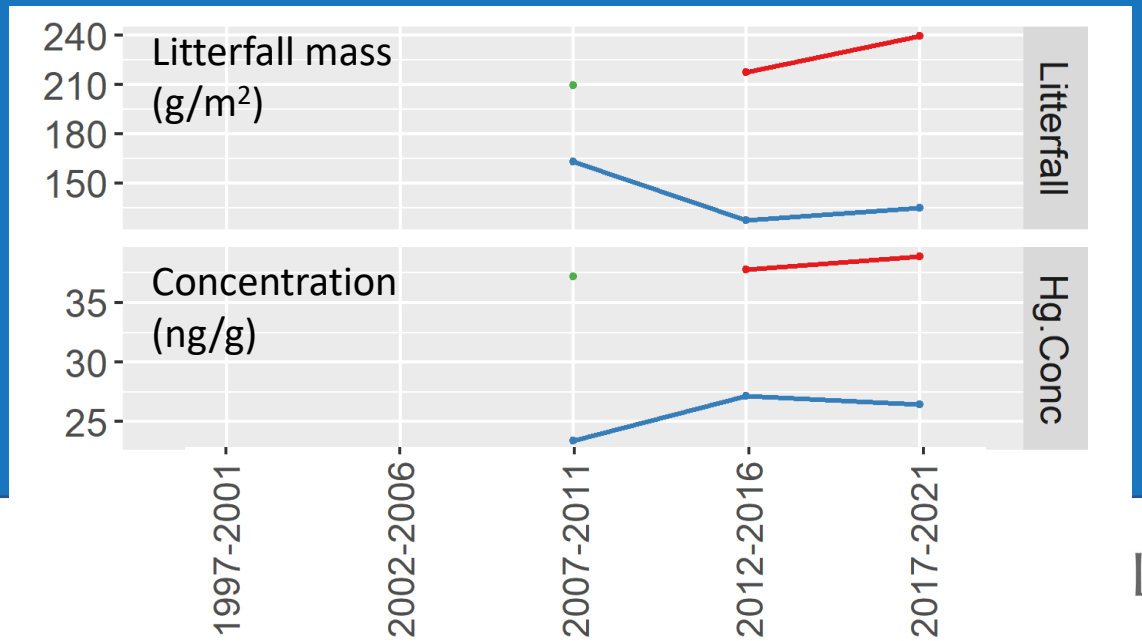
LADCO

Dry (Litterfall) Deposition of Mercury



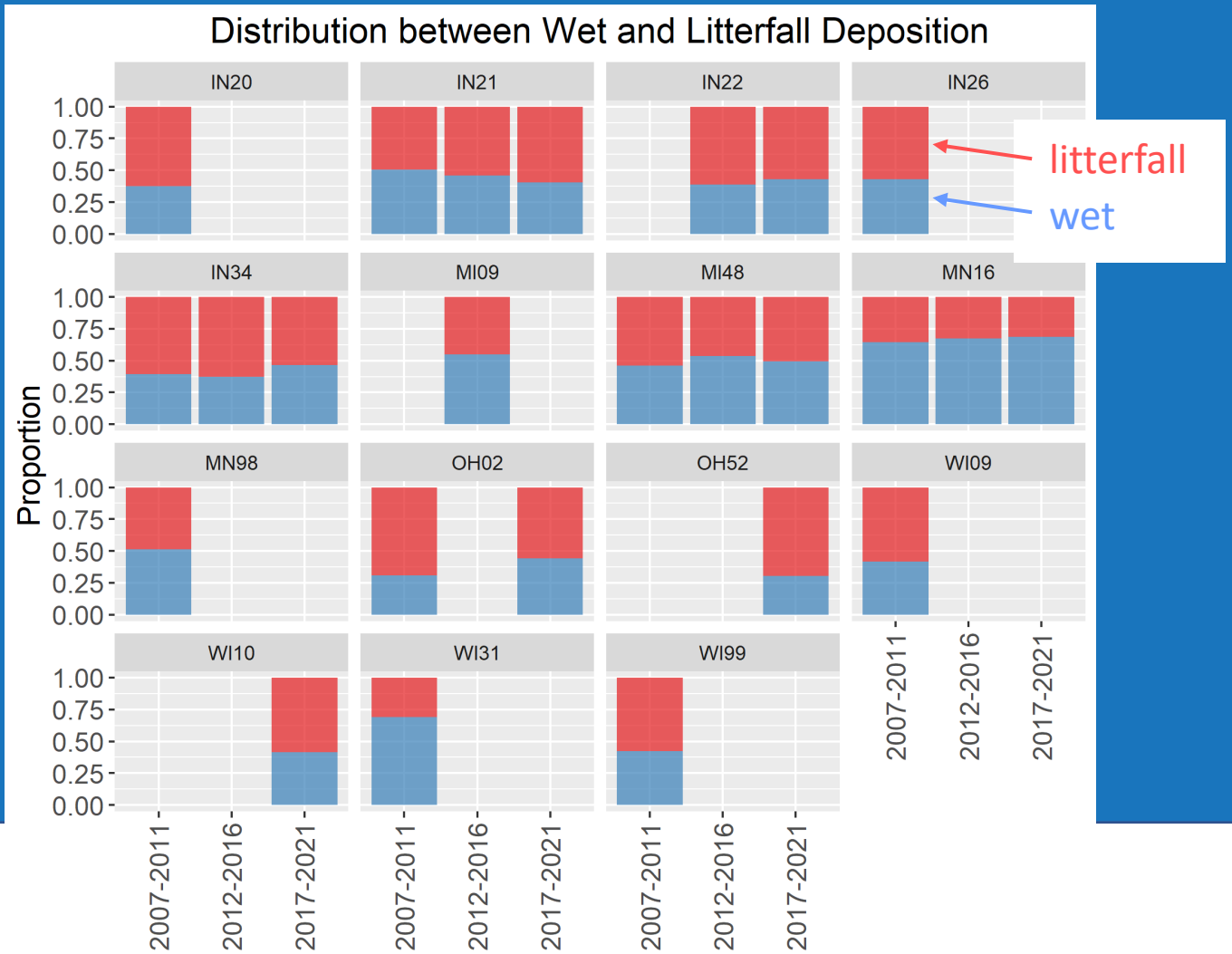
Northern states (MN as example):

- Litterfall deposition is flat relatively steady over the last 15 years
- Litterfall mass and mercury concentrations are also steady



LADCO

Comparison of Wet & Litterfall Deposition



Generally similar contributions from both litterfall and wet deposition

- Both types of deposition are important
- Litterfall seems more important at southern sites
- Wet deposition is more important at some northern sites (MN16 & WI31) but not at others

No clear trends over time

LADCO

Sources of Mercury in the Region

- Based on this analysis and literature studies
- Contributions from local and regional sources are important
  - In addition to continental and global sources
  - Evidence: decreases in Hg concentrations and deposition while global emissions are steady or increasing
  - Southern Great Lakes region:
    - Reductions occurred when major local/regional emissions sources (e.g. EGUs) were installing controls or shutting down
    - Heavy influence from local emissions
  - Northern Great Lakes region:
    - Mixed influence from local, regional, and global sources
    - Previous decreases linked to local emissions reductions (Engstrom et al., 2007)
    - Also influenced by increased precipitation → increased wet deposition

Conclusions

- Both litterfall and wet deposition of mercury are highest in southern areas
  - Near the most/largest sources
- Wet deposition is strongly decreasing in the south but weakly increasing in the north
  - Led to decreases in regional differences over time
- Litterfall deposition is decreasing in the south but trends are unclear in the north


Thank you!
Questions?
dickens@ladco.org

References

- Obrist, D. J.L. Kirk, L. Zhang, E.M. Sunderland, M. Jiskra, N.E. Selin (2018) A review of global environmental mercury processes in response to human and natural perturbations: Changes of emissions, climate, and land use. *Ambio*. https://doi.org/10.1007/s13280-017-1004-9.
- Brigham, M.E., D.D. VanderMeulen, C.A. Eagles-Smith, D.P. Krabbenhoft, R.P. Maki and J.F. DeWild (2021) Long-term trends in regional wet mercury deposition and lacustrine mercury concentrations in four lakes in Voyageurs National Park. *Appl. Sci.* 11, 1879. https://doi.org/10.3390/app11041879.
- Risch, M.R. and D.M. Kenski (2018) Spatial Patterns and Temporal Changes in Atmospheric-Mercury Deposition for the Midwestern USA, 2001–2016. *Atmosphere* 9: 29. https://doi.org/10.3390/atmos9010029.
- Zhang, Y., D.J. Jacob, H.M. Horowitz, L. Chen, H.M. Amos, D.P. Krabbenhoft, F. Slemr, V.L. St. Louis, and E.M. Sunderland (2016b) Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions. *Proc. Nat. Acad. Sci*. 113(3): 526-531. https://doi.org/10.1073/pnas.1516312113.