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Estimates of Speciated and Total Mercury Dry Deposition

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Contents

- **Introduction**
- **Six-year AMNet dry deposition estimation**
- **A literature review on mercury dry deposition**



Introduction – dry deposition

- Gaseous or particulate pollutants are transported to any surfaces (canopy, soil, water, snow, ice), where they are adsorbed and removed from the atmosphere
- Flux - pollutant mass removed from the atmosphere by the surface per unit surface area in unit time ($\text{kg ha}^{-1} \text{yr}^{-1}$, $\mu\text{g m}^{-2} \text{yr}^{-1}$)
- Meteorological, biological, and chemical conditions, as well as pollutants physical and chemical properties all affect the dry deposition process
- Resistance analogy is a commonly used approach in modelling dry deposition; in this approach, a parameter called dry deposition velocity is defined: $\text{Flux} = V_d * C_{\text{con}}$



Introduction – wet deposition

- Gaseous or particulate pollutants are first incorporated into hydrometeors (cloud and rain droplets, ice crystals and snow particles) and then delivered to earth surface
- Wet deposition includes contributions from in- and below-cloud scavenging
- Droplet size distribution and chemical condition, and pollutants physical and chemical properties are dominant factors affecting wet deposition
- Various approaches have been used to model this process



Introduction – a few notes

- Dry deposition happens all the time, even during precipitation; wet deposition is episodic
- Dry and wet deposition are equally important on large temporal and spatial scale
- The only removal mechanisms – controlling pollutants life time in air, and their input to various ecosystems

$$\frac{\partial C}{\partial t} = \textit{emission} + \textit{transportation} + \textit{transformation} + \textit{deposition}$$

- Need to be treated in CTMs for predicting pollutants concentration distribution
- Need to be quantified at monitoring stations for ecosystem effects studies



Six-year AMNet dry deposition estimation

Zhang L., Wu Z., Cheng I., Wright L.P, Olson M.L., Gay D.A, Risch M.R., Brooks S., Castro M.S., Conley G.D., Edgerton E.S., Holsen T.M., Luke W., Tordon R., and Weiss-Penzias P.:

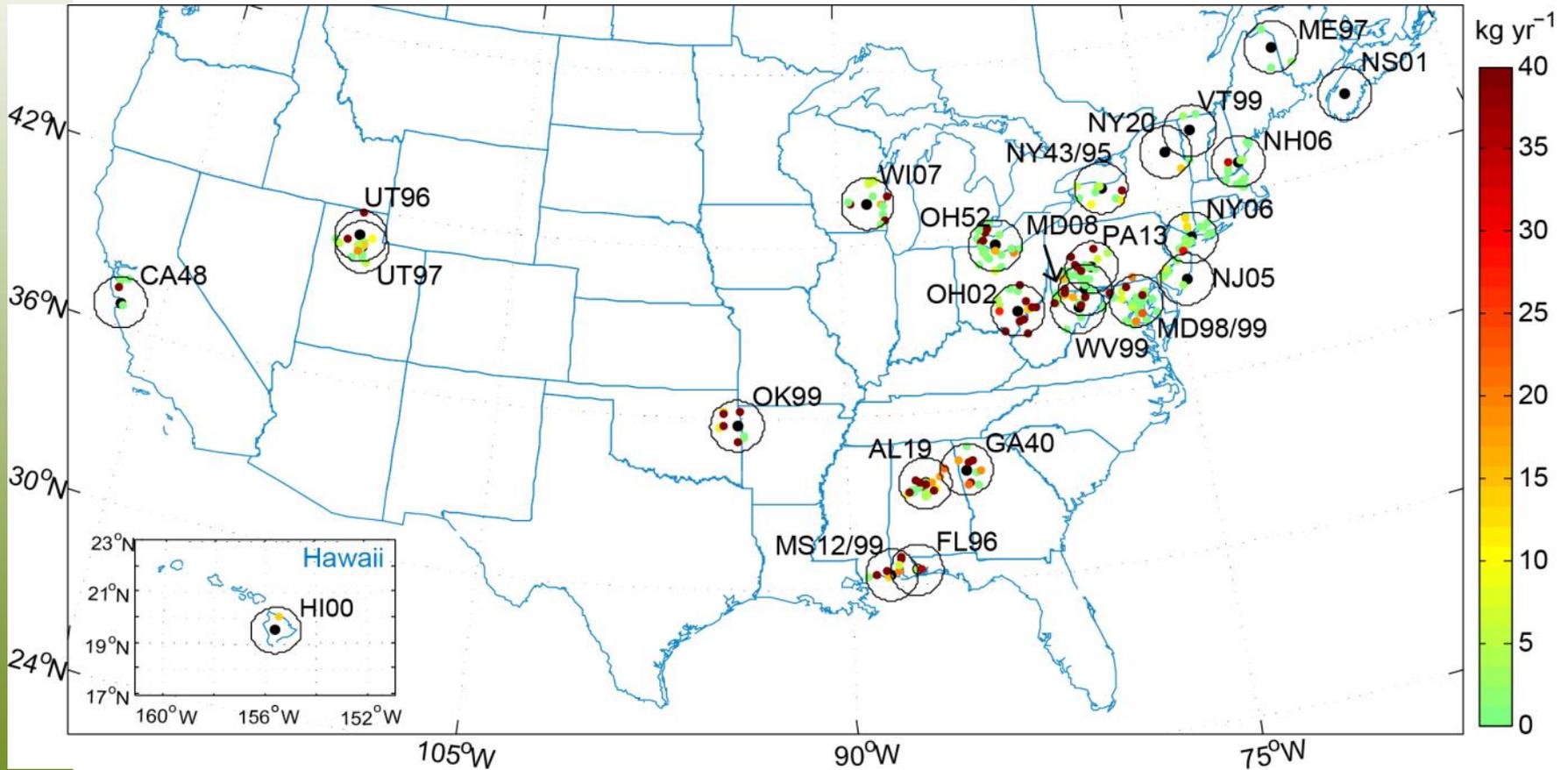
The estimated six-year mercury dry deposition across North America, **Environmental Science and Technology**, 50, 12864-12873, 2016.



AMNet dry deposition project

- One of the major goals of NADP/AMNet is to provide estimates of mercury dry deposition with reasonable accuracy
- NADP/TDEP sub-committee approved Zhang and Gay (2014) proposal in 2014 NADP fall meeting
- The first six-year (2009-2014) AMNet data were finalized
- Three dry deposition algorithms for GOM, PBM and GEM, respectively, were published and ready for calculating 2-hourly speciated fluxes. Land cover data were extracted from MODIS. Meteorological data were extracted from the archived surface-layer model output of Canadian weather forecast model.
- First six-year results published in EST in December 2016
- NADP/TDEP approved the first-six year results to be posted on NADP website in April 2017.

Atmospheric Mercury Network (AMNet)



Deposition algorithms

- **A big-leaf gaseous dry deposition model for GOM (Zhang et al., 2003, 2012)**
- **A bulk particle dry deposition model for PBM (Zhang and He, 2014)**
- **A bi-directional air-surface exchange model for GEM (Wright and Zhang, 2015)**



Coarse PBM is included in deposition budget

$$F_{finePBM} = C_{AMNet} * V_{dF}$$

$$F_{total} = C_{AMNet} \left(V_{dF} + \frac{CF}{1 - CF} V_{dC} \right)$$

CF - mass fraction of coarse PBM (assumed 0.3 for all the sites)

Flux missed by the network



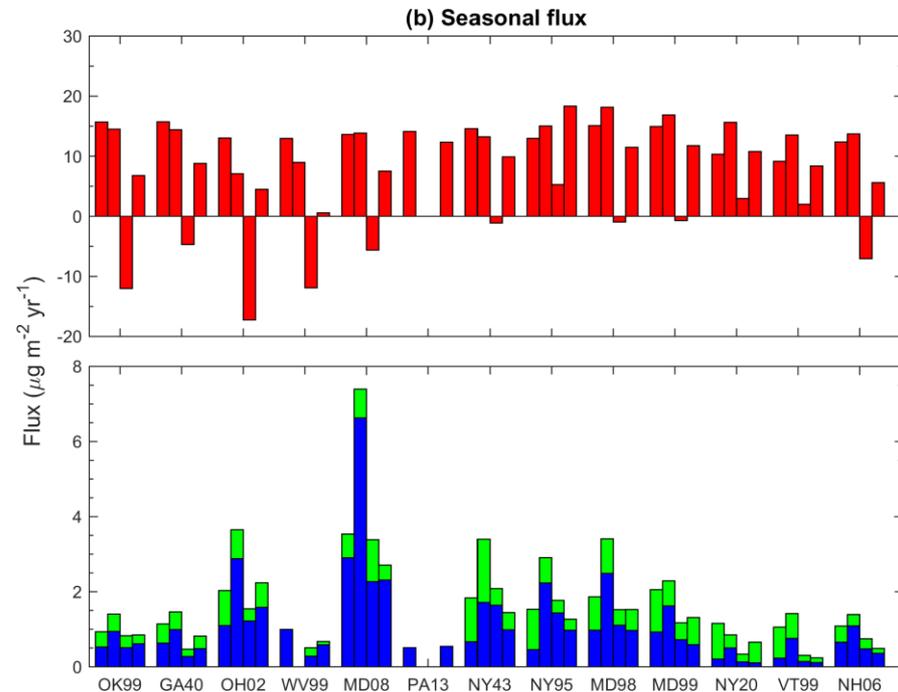
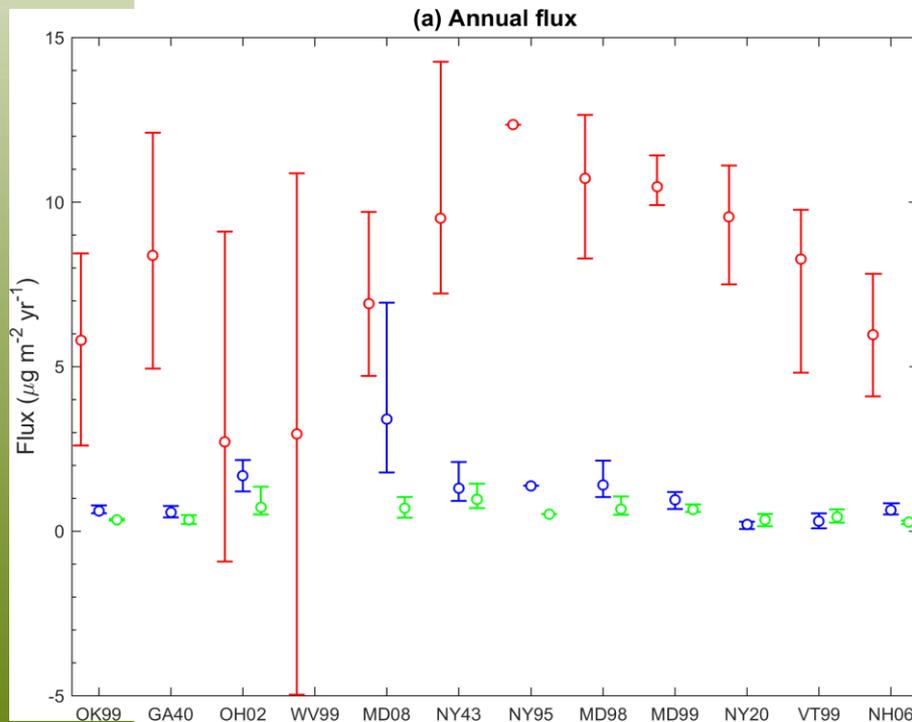
24 sites and with 3 collocated sites

AMNet site ID	Site name	Latitude, longitude	Data coverage	Site category ^d
AL19	Birmingham	33.5530, -86.8148	Jan/2009-Dec/2014	Urban
CA48	Elkhorn Slough	36.8100, -121.780	Jan/2010-Dec/2011	Suburban
FL96	Pensacola	30.5500, -87.3753	Jan/2009-Dec/2014	Rural
GA40	Yorkville	33.9283, -85.0456	Jan/2009-Dec/2014	Rural
HI00	Mauna Loa	19.5362, -155.576	Jan/2011-Dec/2014	Rural
MD08	Piney Reservoir	39.7054, -79.0126	Jan/2009-Jul/2013	Rural
MD98/99	Beltsville	39.0283, -76.8171	Jan/2009-Dec/2014	Rural
ME97	Presque Isle	46.6964, -68.0332	Dec/2013-Dec/2014	Suburban
MS12/99	Grand Bay NERR	30.4124, -88.4038	Jan/2009-Dec/2014	Rural
NH06	Thompson Farm	43.1088, -70.9485	Jan/2009-Nov/2011	Rural
NJ05	Brigantine	39.4649, -74.4488	Jun/2009-Apr/2010	Suburban
NS01	Kejimikujik National Park	44.4321, -65.2031	Jan/2009-Dec/2014	Rural
NY06	NYC	40.8679, -73.8782	Jan/2009-Dec/2014	Urban
NY20	Huntington Wildlife Forest	43.9736, -74.2232	Jan/2009-Dec/2014	Rural
NY43/95	Rochester	43.1463, -77.5483	Jan/2009-Dec/2014	Suburban
OH02	Athens	39.3080, -82.1182	Jan/2009-Dec/2014	Rural
OH52	South Bass Island	41.6582, -82.8272	Jan/2013-Dec/2014	Rural
OK99	Stilwell	35.7508, -94.6696	Jan/2009-Nov/2014	Rural
PA13	Allegheny Portage	40.4571, -78.5603	Nov/2009-Sep/2011	Rural
UT96	Antelope Island	41.0885, -112.119	Jun/2009-Jun/2011	Suburban
UT97	Salt Lake City	40.7118, -111.961	Jan/2009-Dec/2014	Urban
VT99	Underhill	44.5285, -72.8682	Jan/2009-Dec/2014	Rural
WI07	Horicon	43.4557, -88.6169	Jan/2011-Dec/2014	Rural
WV99	Canaan Valley Institute	39.1189, -79.4522	Jan/2009-Oct/2012	Rural



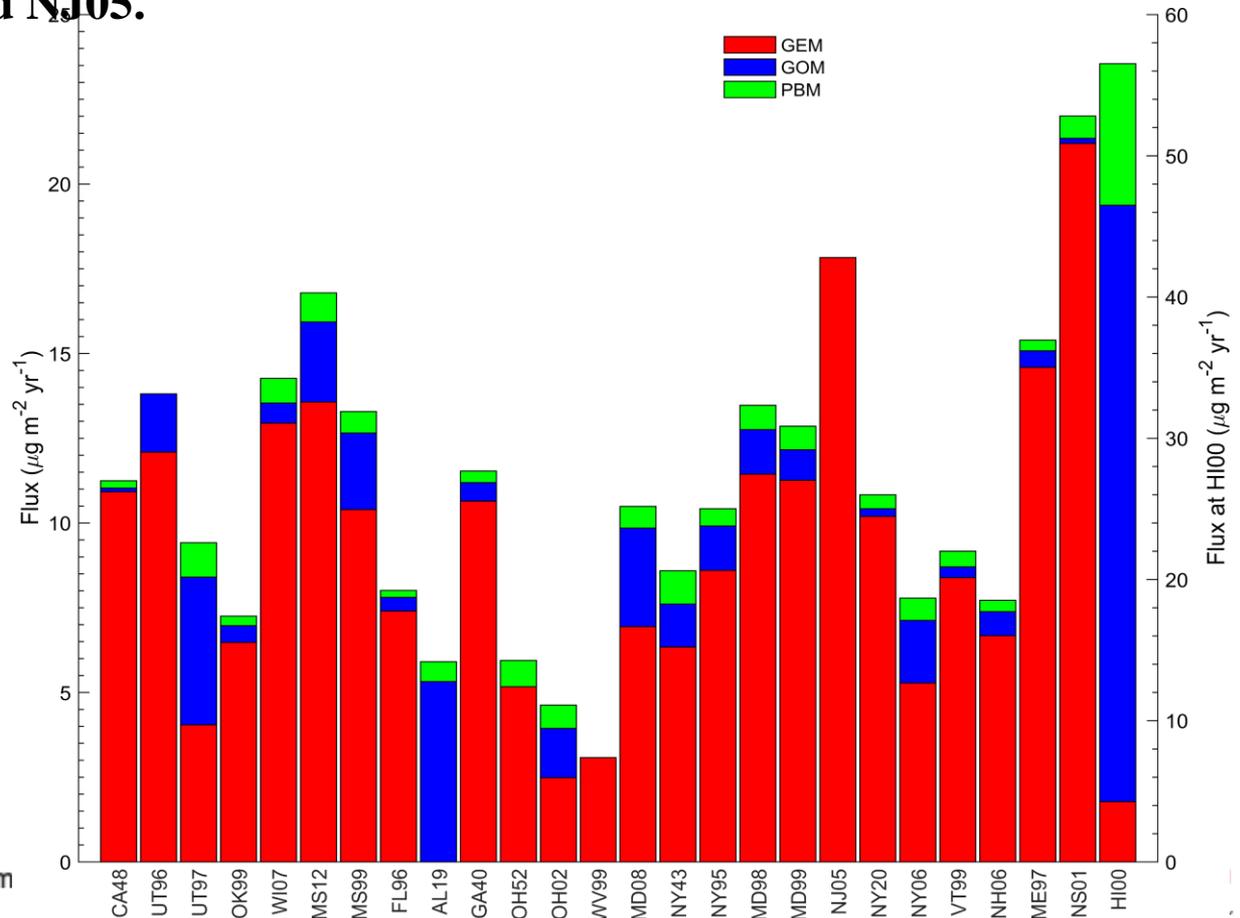
Annual and seasonal Fluxes

Estimated speciated mercury (GEM-red, GOM-blue, and PBM-green) fluxes over deciduous broadleaf forests. (a) Mean (circle) and range (vertical line) of the annual values between 2009-2014; and (b) six-year average of seasonal values (four columns at each site represent winter, spring, summer, and autumn, respectively). Only sites having the specific land cover within a 3-km radius circle are shown and sites are arranged in order from the Pacific to Atlantic coast.

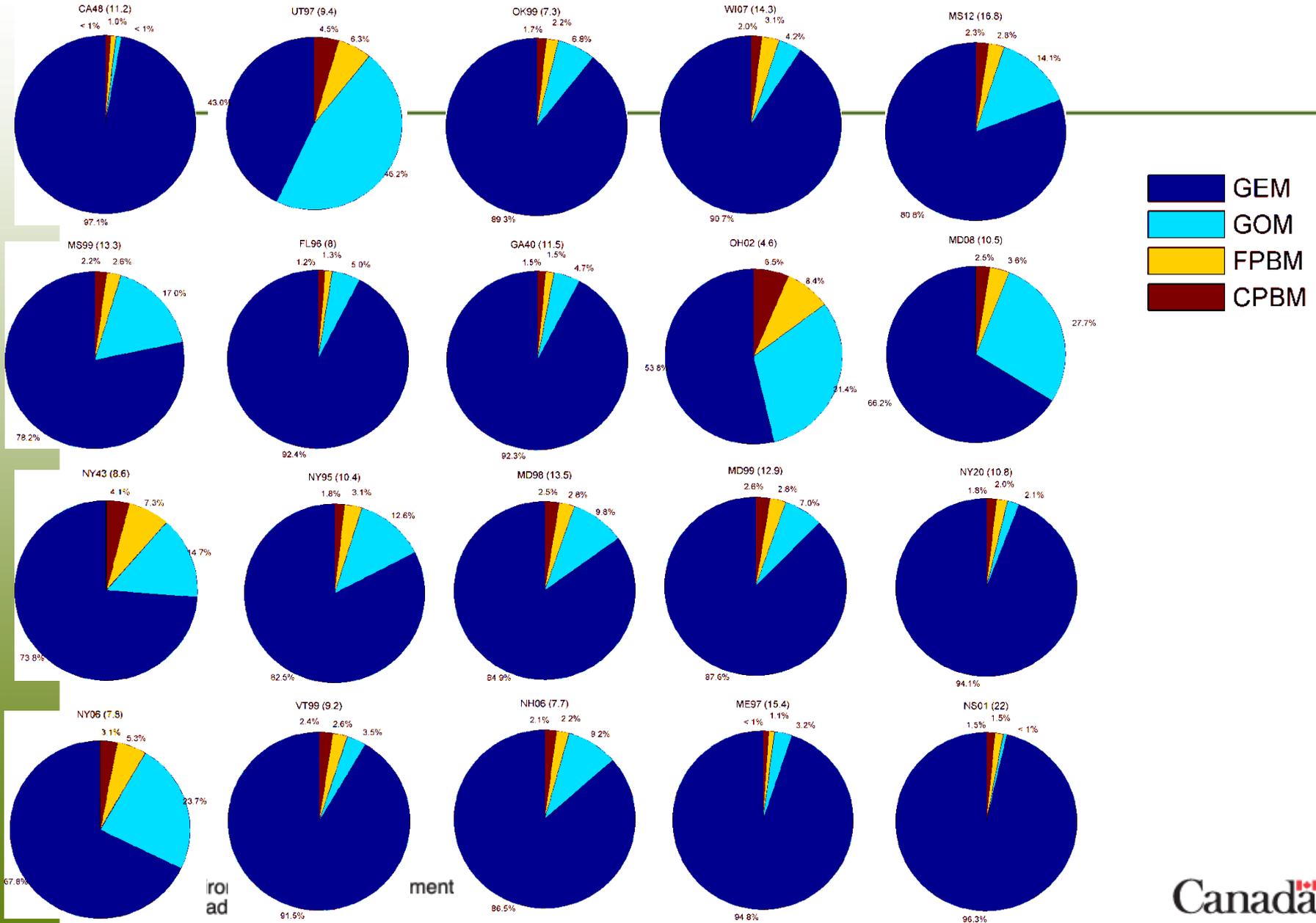


Multi-year mean fluxes

Multi-year mean land cover area-weighted dry deposition at all of the sites. Note the insufficient data coverage of PBM at UT96, GOM at OH52, and both GOM and PBM at WV99 and NJ05.

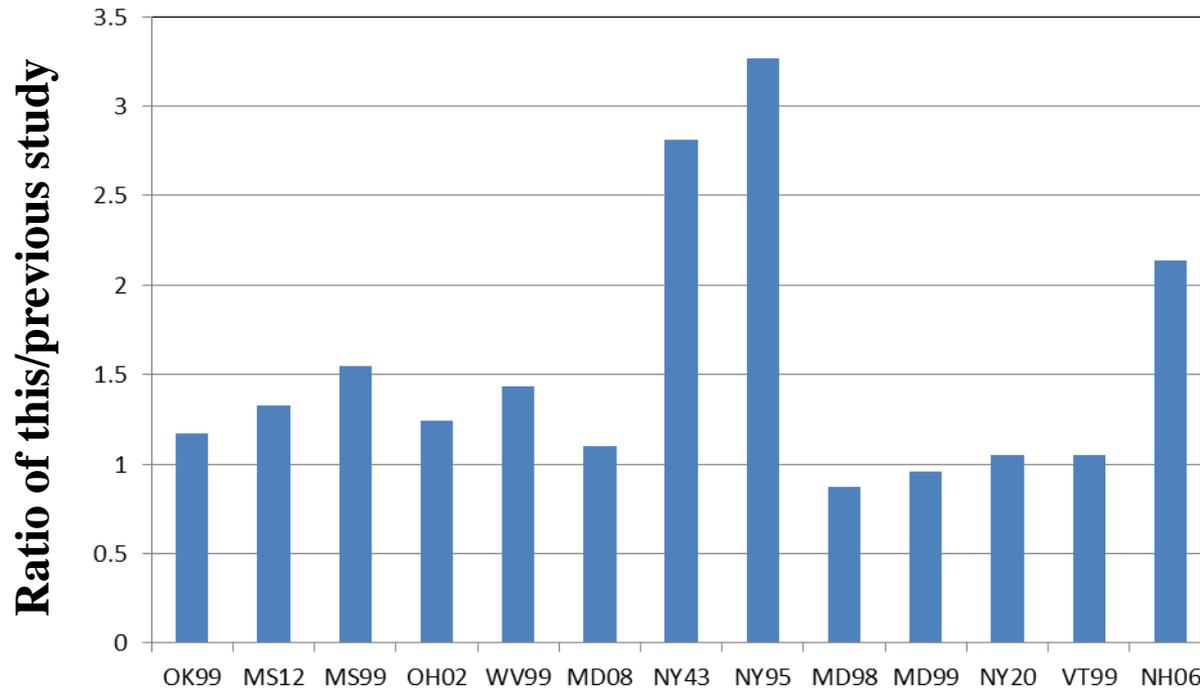


Total annual dry deposition (number shown on top of each pie) and percentage contributions from GEM, GOM and fine and coarse PBM



Uncertainty assessment - One

To assess if GEM emission was underestimated (or if net GEM deposition was overestimated) – compare with previously modeled emission (Zhang et al., 2012)



Conservative dry deposition estimation at most sites (with emission ratio > 1)



Uncertainty assessment - two

To compare with litterfall measurements at the regional scale – litterfall Hg measurements likely represent the low end of Hg dry deposition

Multi-year mean (2009-2014) Hg dry deposition ranged from 5.1 to 14.3 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to deciduous broadleaf forests, and from 5.1 to 23.8 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to all forest types.

Annual litterfall Hg (2007-2014) from predominantly deciduous forests at 15 sites in eastern U.S. ranged from 3.5 to 23.4 $\mu\text{g m}^{-2} \text{yr}^{-1}$



Uncertainty assessment - three

Adjust GOM concentrations by a factor of 3.0

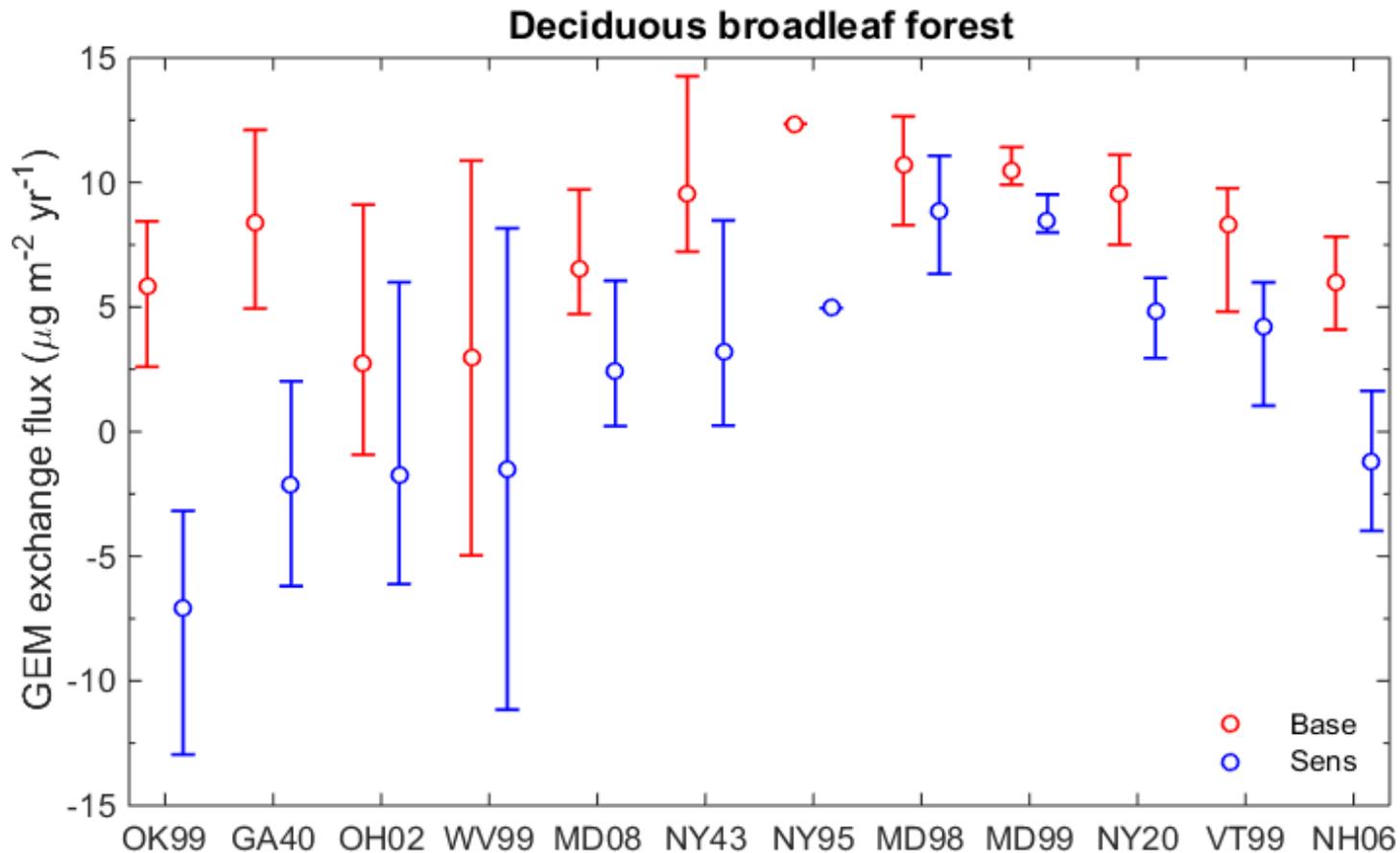
The adjusted total dry deposition to deciduous broadleaf forest ranged from **8.0 to 19.0** $\mu\text{g m}^{-2} \text{yr}^{-1}$ among all of the sites, and from 8.5 to 19.0 $\mu\text{g m}^{-2} \text{yr}^{-1}$ among the four collocated sites.

Still agree with litetrfall measurements, and does not change the big-picture (the relative contribution of GEM and GOM+PBM) - GOM plus PBM contributions to the total dry deposition increased slightly to 5-78% from the original 3-57%



Uncertainty assessment - four

A sensitivity test on GEM flux to deciduous broadleaf forest using higher emission potentials (e.g., the case in Asia with high GOM dry and wet deposition)



Conclusions

- Depending on location, multi-year mean annual Hg dry deposition was estimated to range from 5.1 to 23.8 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to forested canopies, 2.6 to 20.8 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to non-forest vegetated canopies, 2.4 to 11.2 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to urban and built up land covers, and 1.0 to 3.2 $\mu\text{g m}^{-2} \text{yr}^{-1}$ to water surfaces.
- In the rural or remote environment in North America, annual Hg dry deposition to vegetated surfaces is dominated by leaf uptake of gaseous elemental mercury
- Dry deposition exceeded wet deposition by a large margin in all of the seasons except in the summer at the majority of the sites.
- At the regional scale, the estimated annual total dry deposition is in the same range as litterfall mercury measurements.
- Uncertainty analysis suggested that GEM dry deposition over vegetated surfaces will not decrease at the same pace, and sometimes may even increase with decreasing anthropogenic emissions, suggesting that Hg emission reductions should be a long-term policy sustained by global cooperation.



A literature review on mercury dry deposition

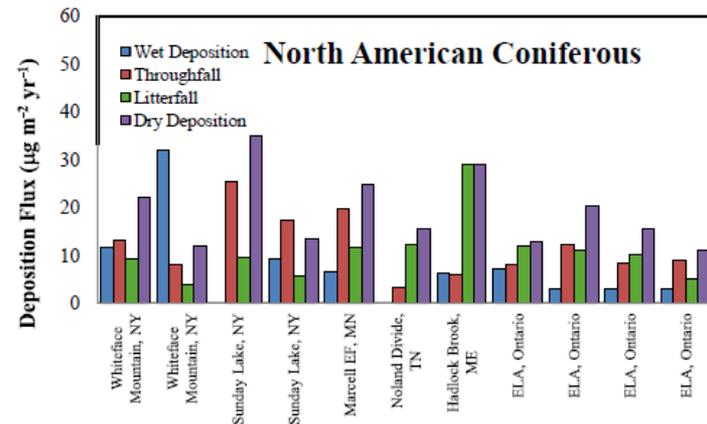
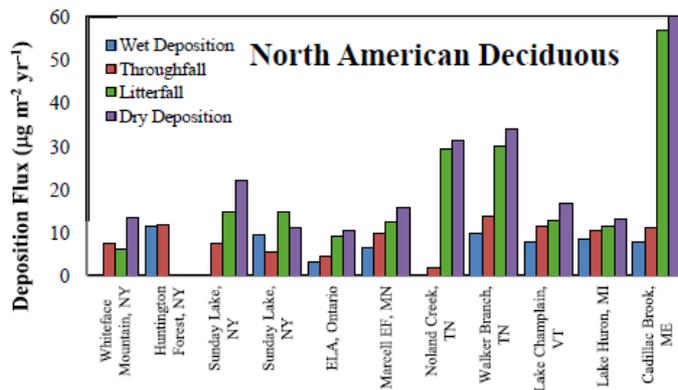
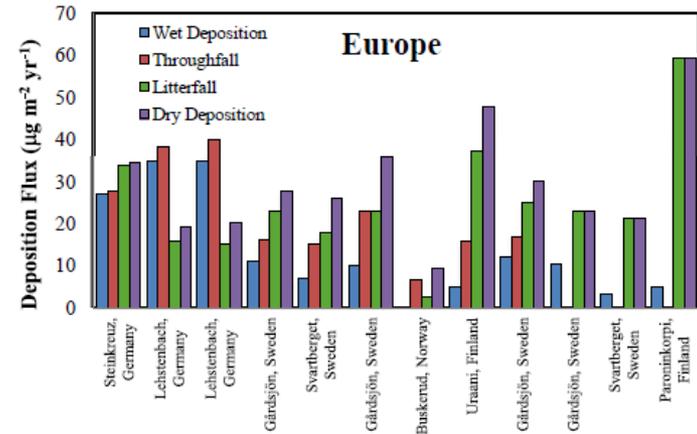
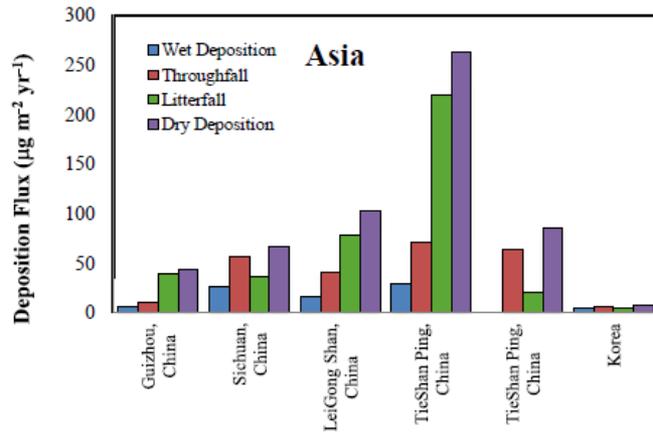
Wright L.P., Zhang L., and Marsik F.J.:
Overview of mercury dry deposition, litterfall, and throughfall studies, *Atmospheric Chemistry Physics*, 16, 13399-13416, 2016.



Contents included in the review paper

- 2. Dry deposition algorithms and estimation**
 - 2.1 In chemical transport models**
 - 2.2 At monitoring networks/sites**
- 3. Deposition (flux), litterfall, and throughfall measurement methodologies**
 - 3.1. Micrometeorological approaches**
 - 3.2. Dynamic gas flux chambers**
 - 3.3. Surrogate surface approaches**
 - 3.4. Litterfall and throughfall-based approaches**
- 4. Oxidized mercury flux measurements**
- 5. Litterfall measurements**
- 6. Throughfall measurements**
- 7. A brief comparison of dry, litterfall, throughfall and wet deposition**

Comparison of mercury deposition in dry, wet, litterfall and throughfall





Thank you!



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