Trends and modeling of the total gaseous mercury flux and deposition in the leaf litter fall in a Northeastern red maple canopy

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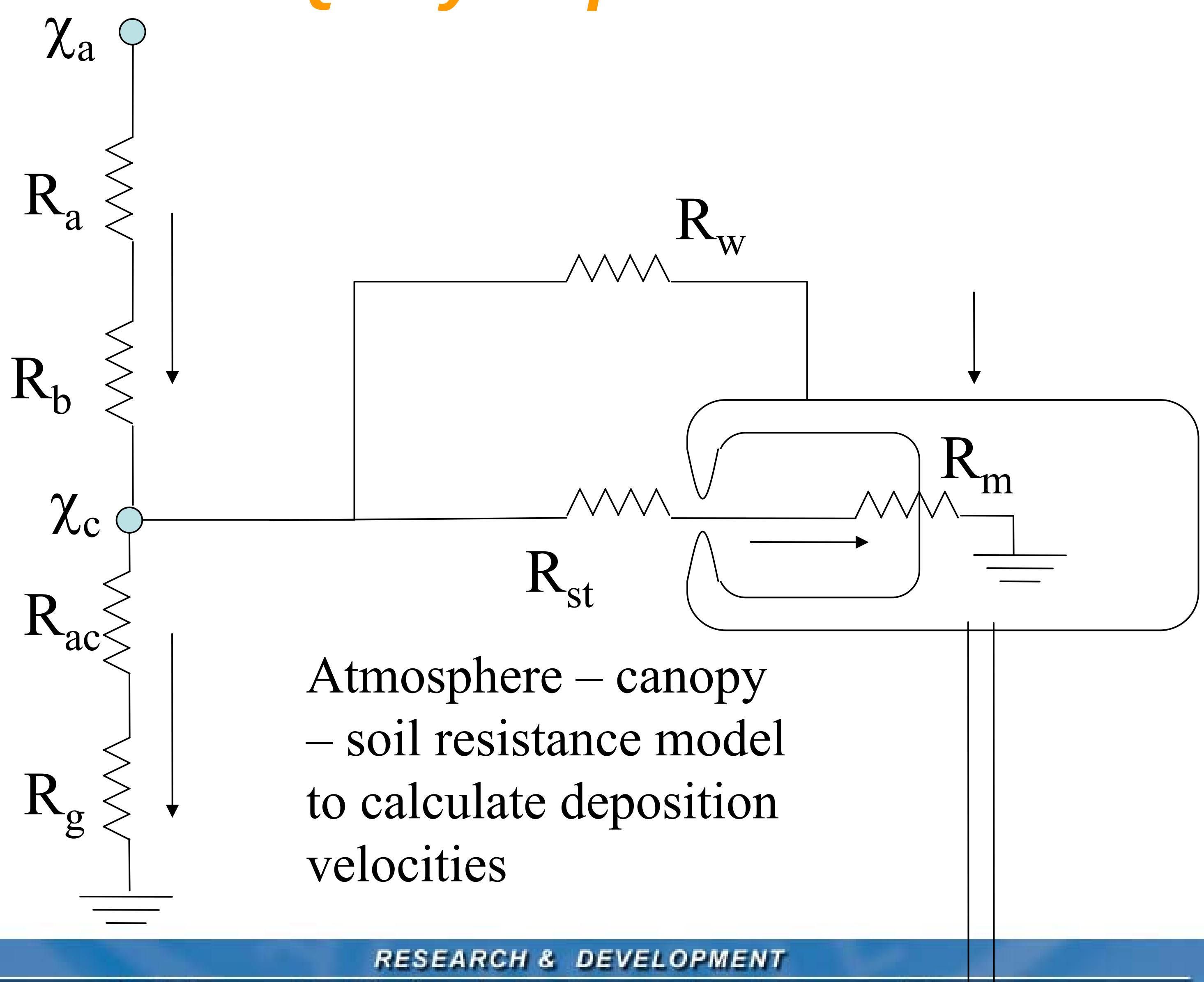
Outline

- Bi-directional framework in CMAQ
- Site and measurement description
- Results
  - TGM canopy fluxes
  - Atmospheric – canopy compensation points
  - Under canopy fluxes
  - Wet and litter fall deposition
  - Soil and vegetation concentrations
- Conclusions

Background

- Natural mercury emissions/re-emissions
  - Estimated to be a large fraction of the total mercury emissions
  - Believed to contribute to long range transport of mercury though re-emissions
  - Residence time in terrestrial media can be on the order of decades
- Once in the terrestrial system mercury is available for methylation
- Largest pools of mercury are in the terrestrial system
  - Emission/re-emission processes are a means of transport through the atmosphere

CMAQ Dry Deposition Model

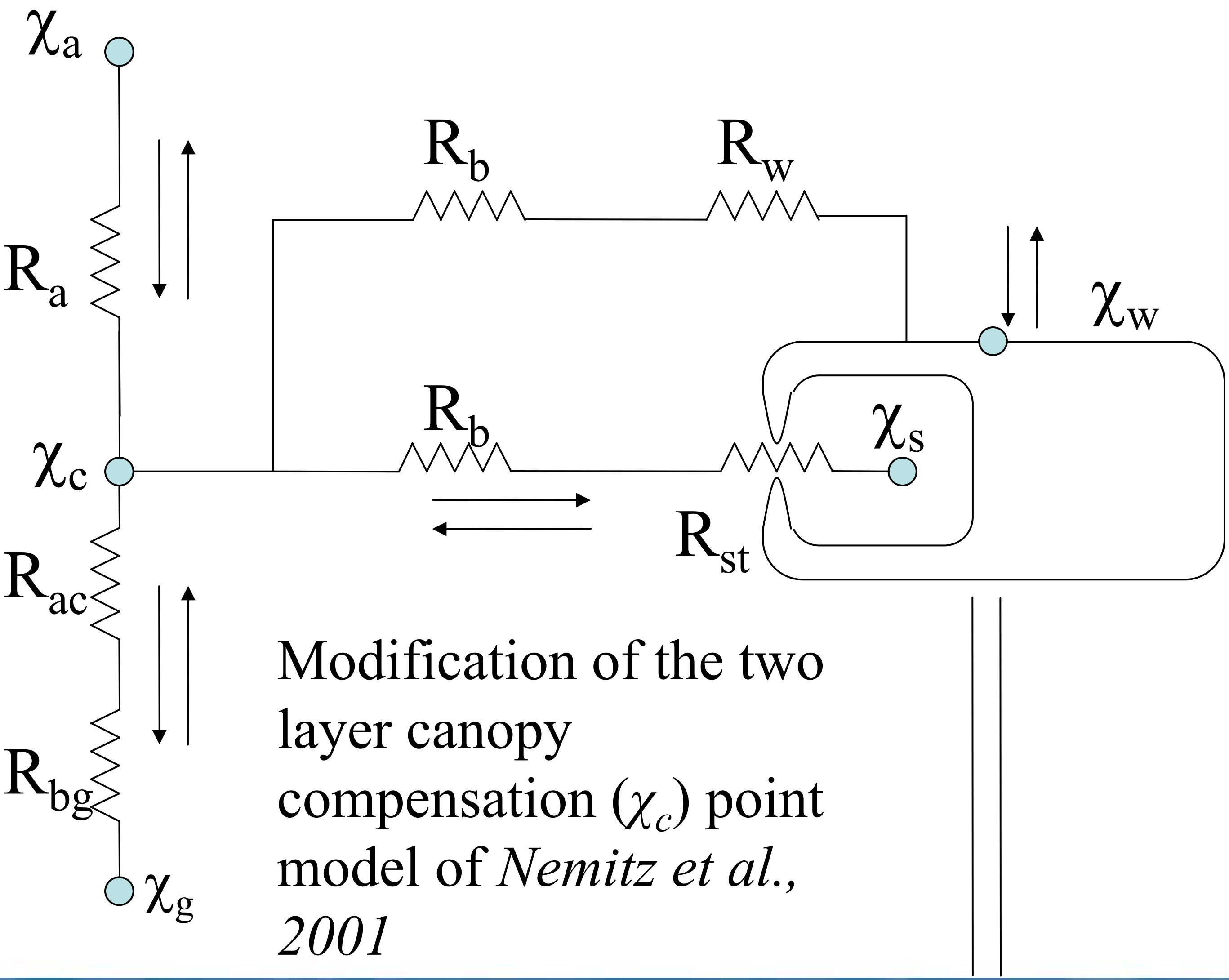


Atmosphere – canopy – soil resistance model to calculate deposition velocities

Modeling bi-directional surface exchange

- Bi-directional surface exchange capability is being developed in CMAQ
  - Adaptation of NH_3 bi-directional algorithms for mercury
- Modification of dry deposition routines
  - Adds canopy, soil and vegetation concentrations to parameterize a concentration gradient
  - Uses a resistance analogy to model exchange coefficients
- Requires knowledge of surface properties and in-canopy air movement
  - Where mercury is deposited will determine the mechanisms of its re-emission

Bi-directional Model

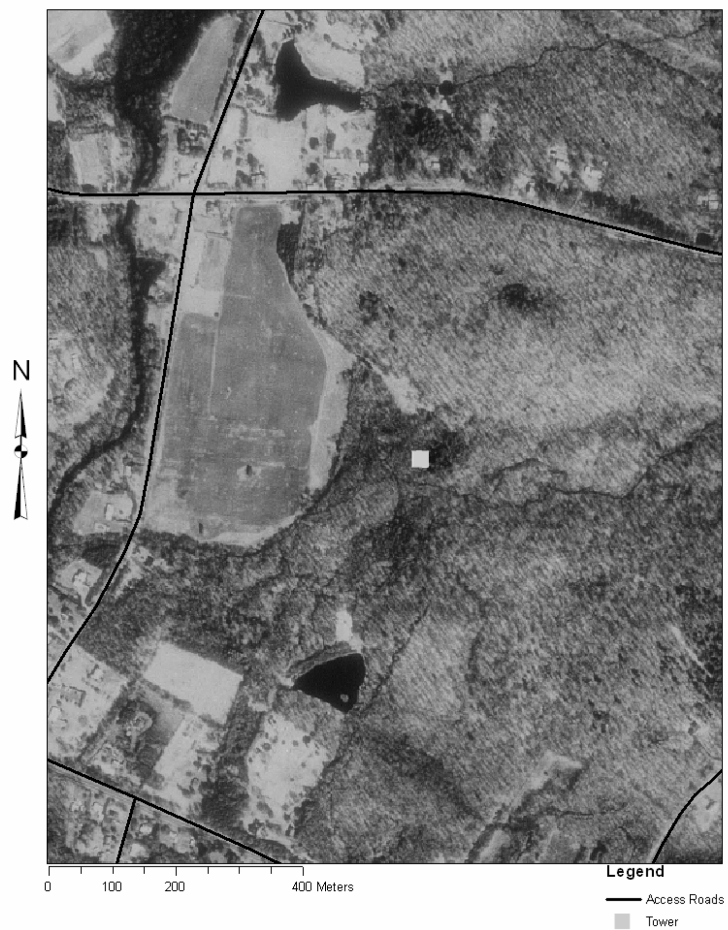


Modification of the two layer canopy compensation (χ_c) point model of *Nemitz et al., 2001*

Total gaseous mercury flux measurements

- Located in rural Coventry, CT
- Employed relaxed eddy accumulation technique
- Fluxes taken at 1.2 canopy heights in a 21 meter closed red maple stand
- Wetland to west and southwest
- Oak stand to the north east on a slightly elevated hill

Coventry, CT mercury flux tower



TGM flux

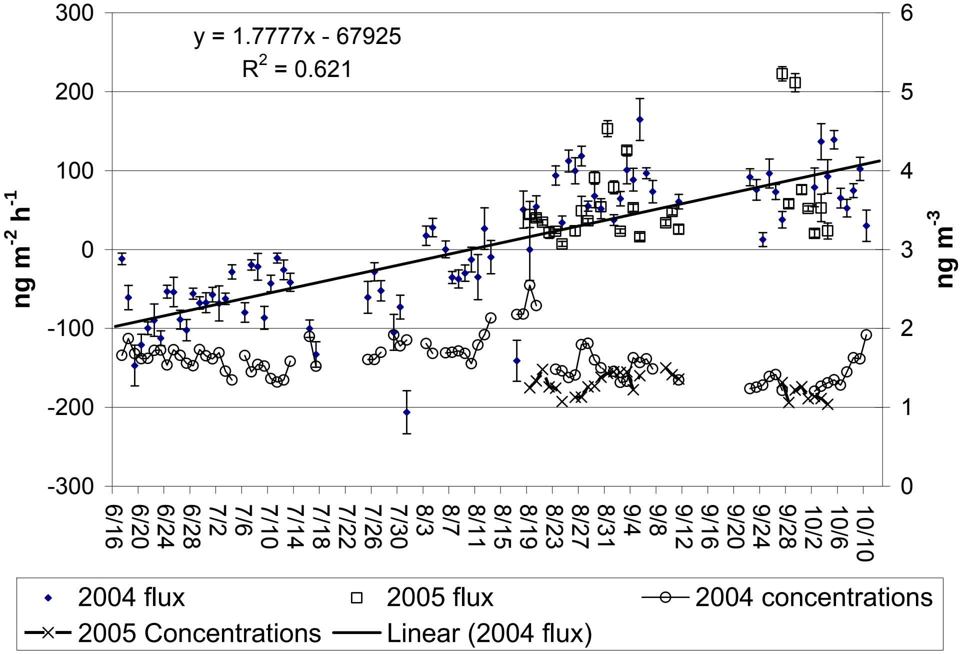
Atmosphere-canopy flux

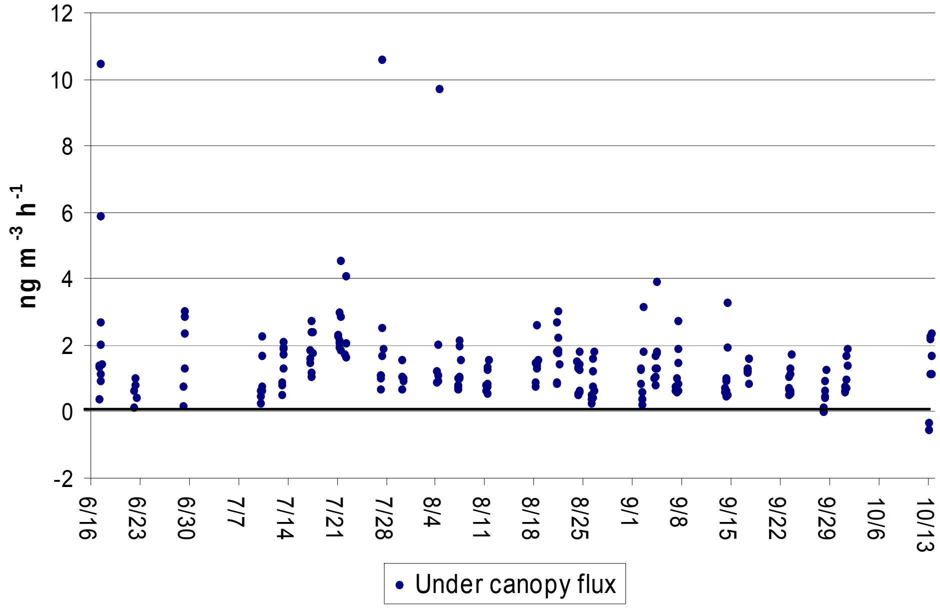
• Measured using the REA micrometeorological technique

Under canopy air-soil flux

• Measured using the dynamic flux chamber technique

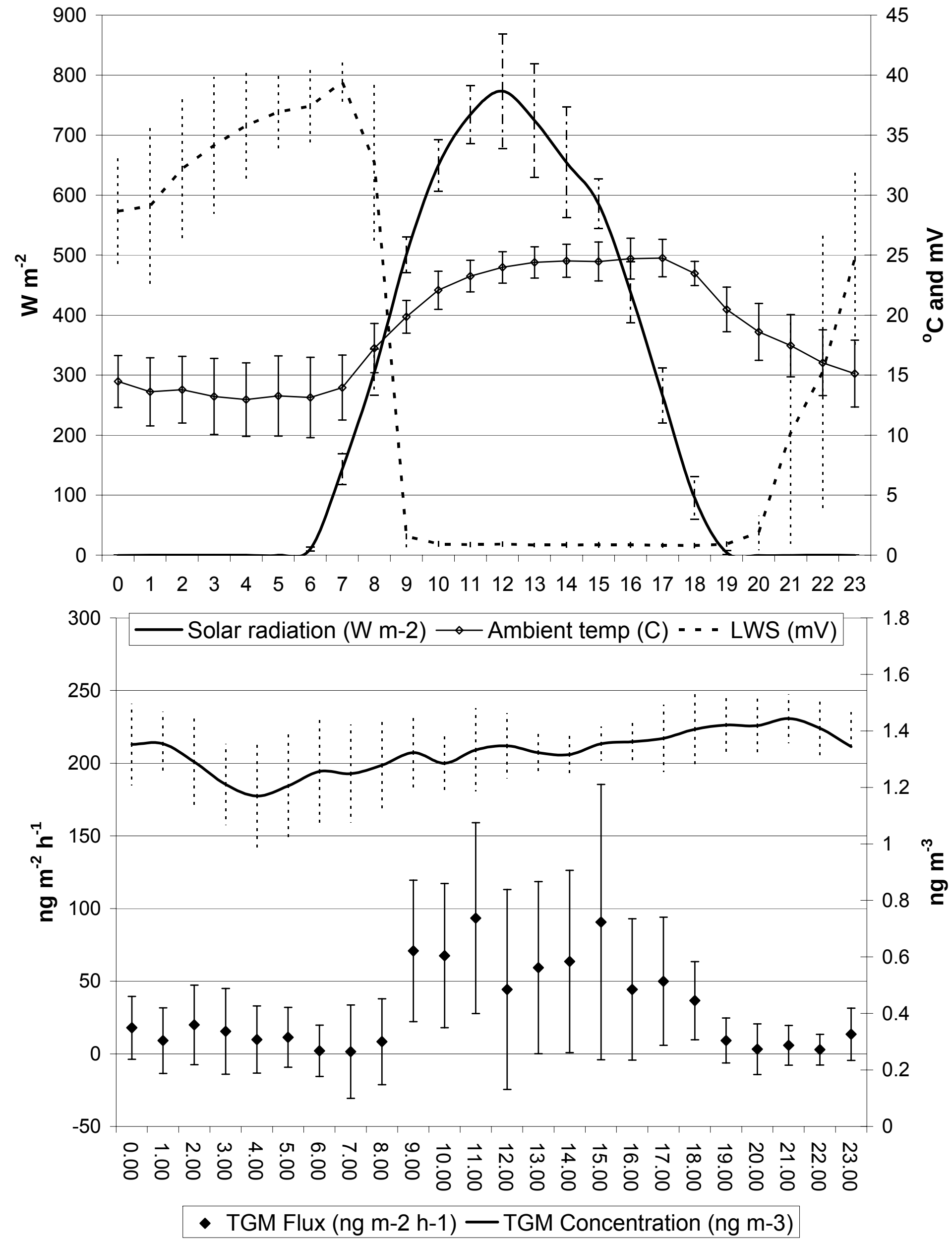
• Not continuously sampled



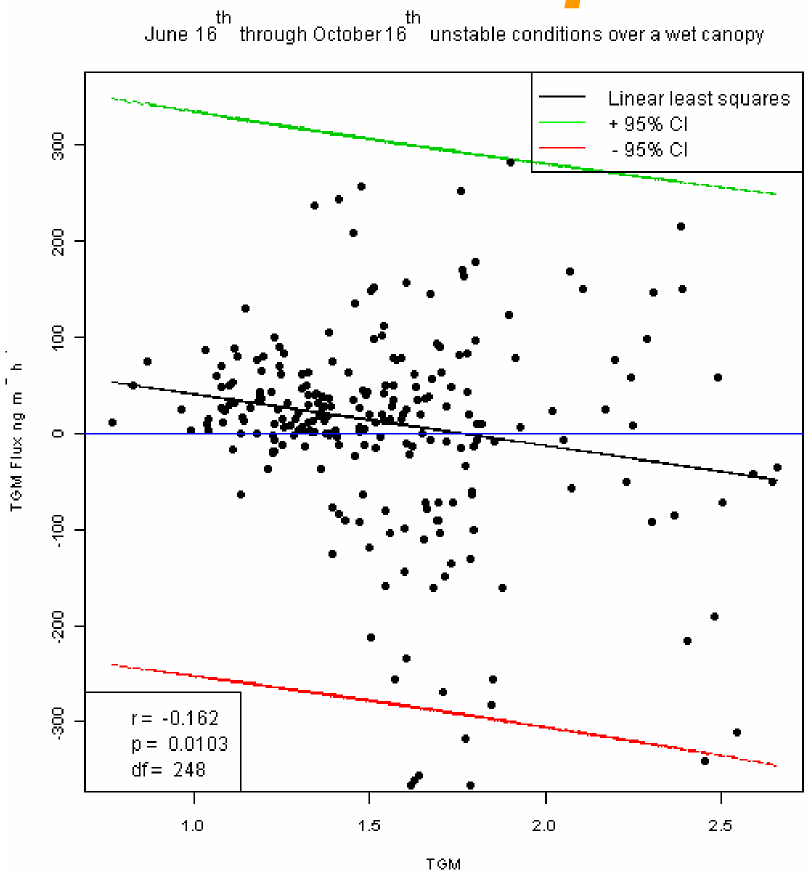


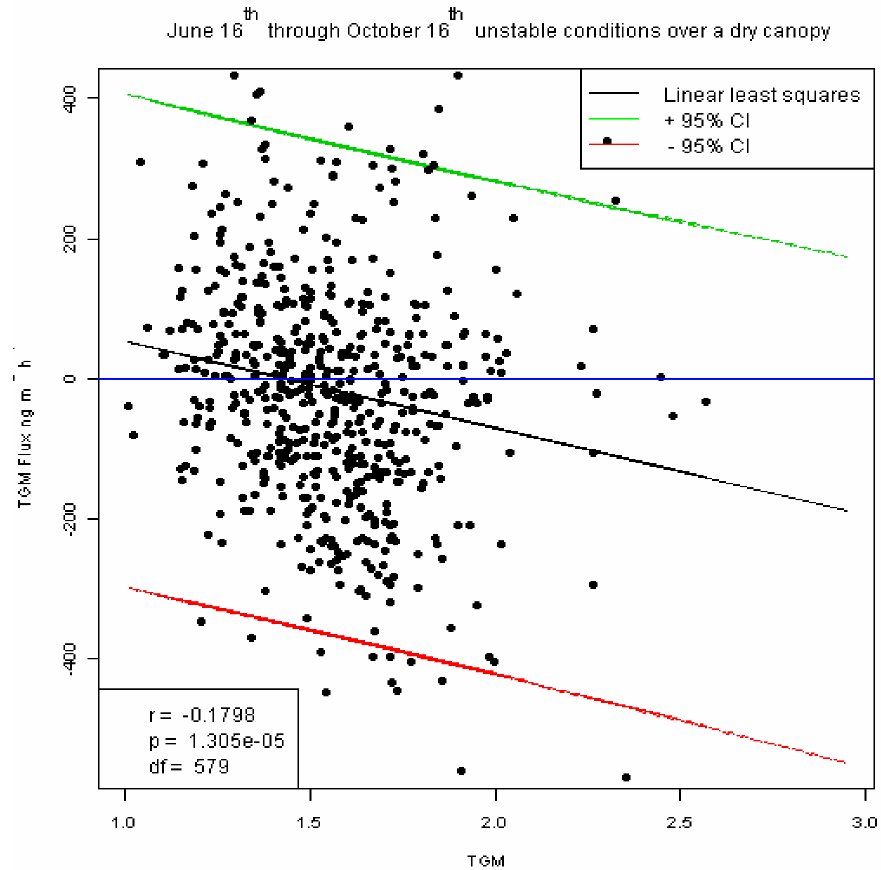
2005 average daily flux

- Aug 18th through Sept 12th 2005
- Morning peak in flux around the time that dew evaporates from canopy
- Afternoon peak in flux around peak in ambient temperature
- Net 2004 growing season evasive flux of 12.94 μg m^{-2}



Compensation Points

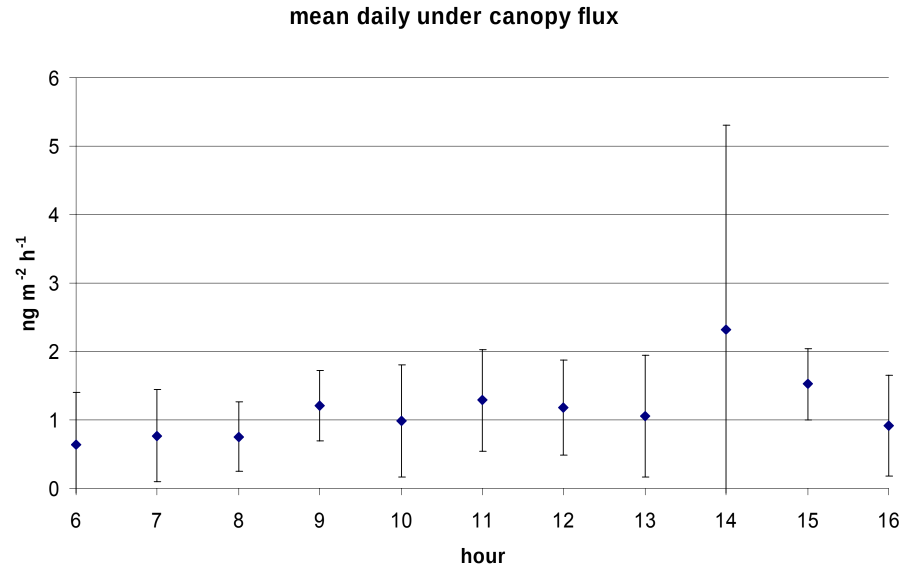


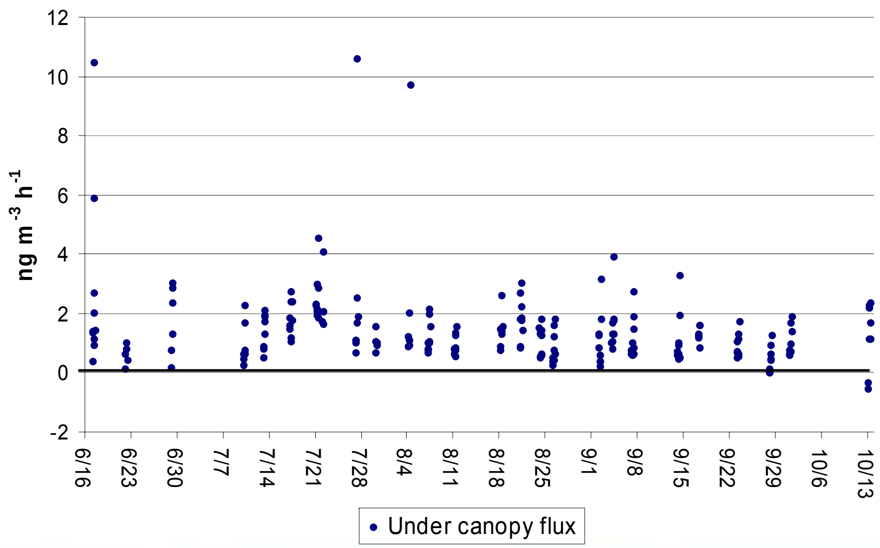


- Wet canopy compensation point of 1.76 ng m^3 (2.71 ng m^3 for stable conditions)
- Dry canopy compensation point of 1.43 ng m^3 (2.12 ng m^3 for stable conditions)
- Mean TGM concentration of 1.54 ng m3 (1.59 ng m^3 under stable conditions)
- Compensation point increase through the growing season

Under Canopy Soil Flux

- Measured using a Teflon dynamic flux chamber
- Under canopy flux measurements were taken on the drier elevated and transitional areas
- Soils under canopy were a consistent emissions source in 2004
- Flux was not correlated with soil moisture
    - Less than 10% variation in soil moisture from May through October





Vegetation Concentrations

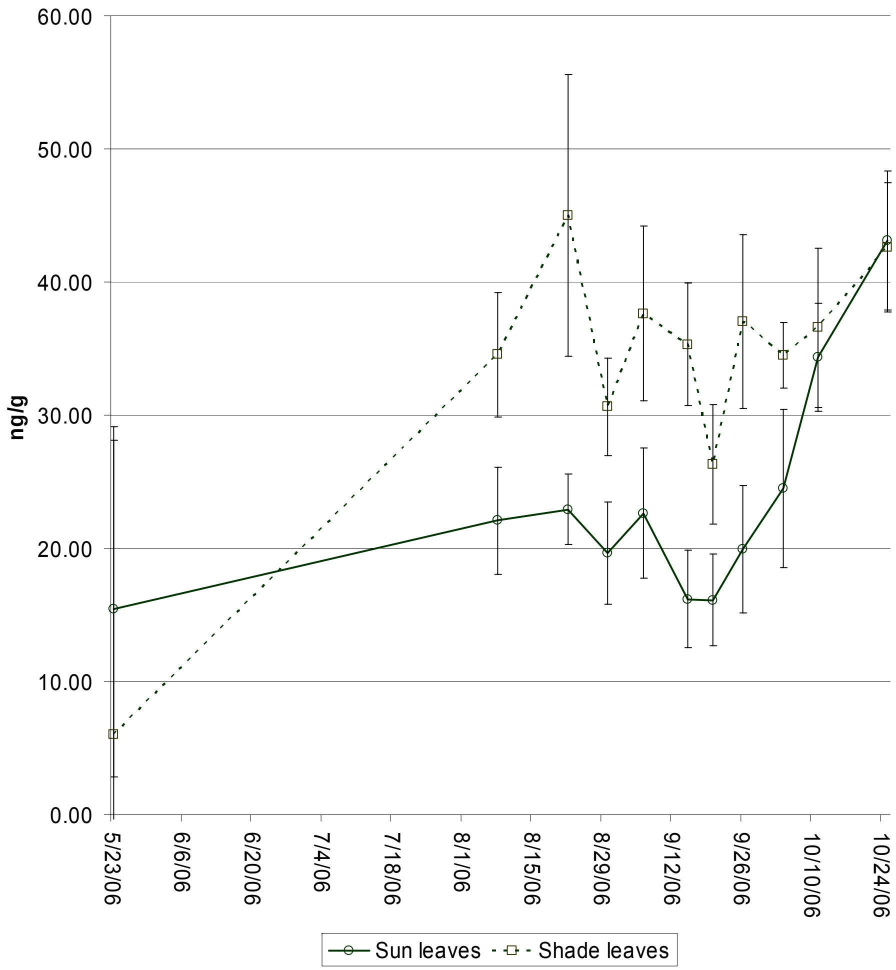
- *Acer Rubrum* leaf mercury concentrations from shade leaves (z_m~ 4 m) were consistently higher than from sun leaves (z_m ~ 20 m)

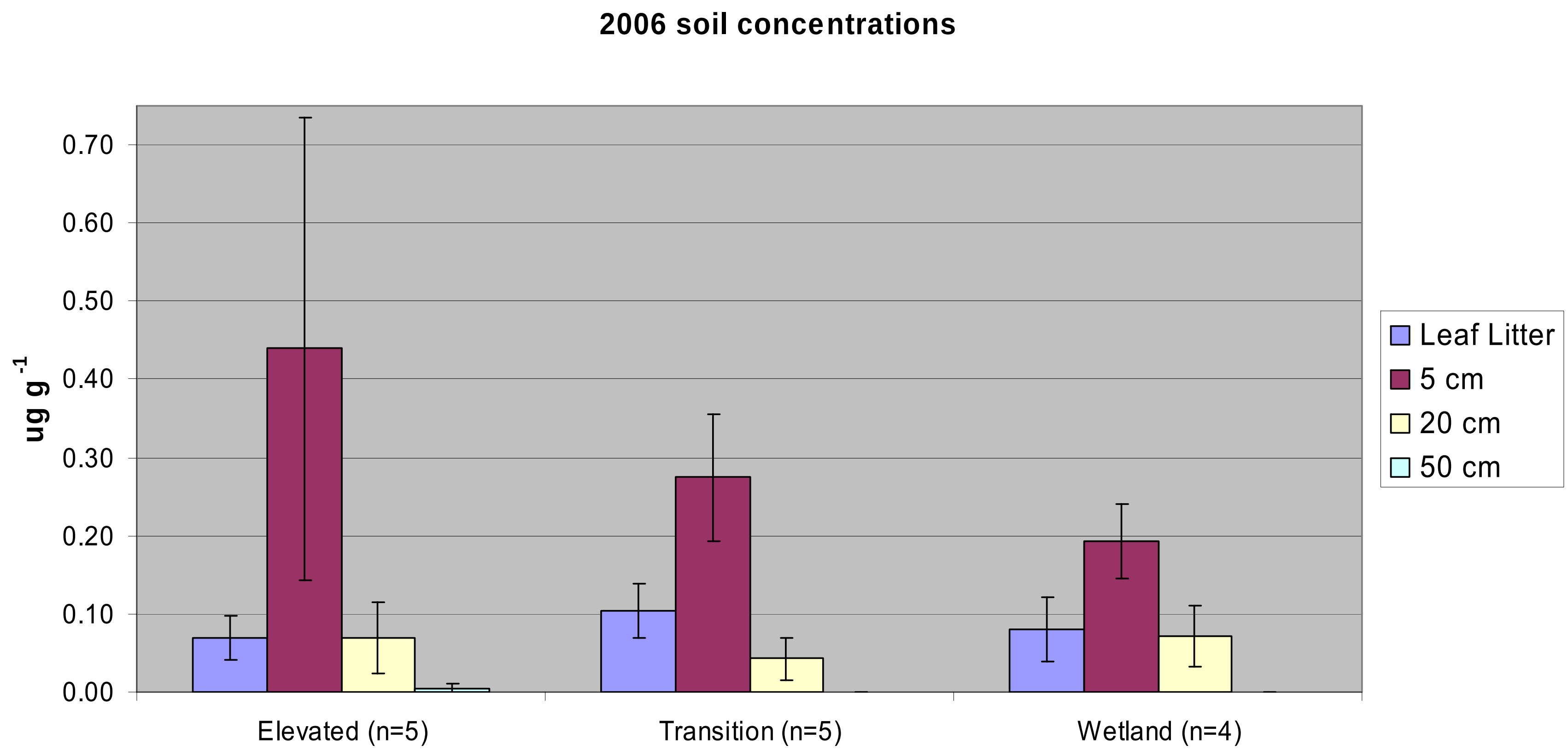
- Sun leaves are exposed to more solar radiation, higher temperatures, and higher wind speeds

- Soil was consistent emission source

- In-canopy concentration gradient is unknown

- Annual fall leaf litter deposition of 12.10 µg m^{-2}

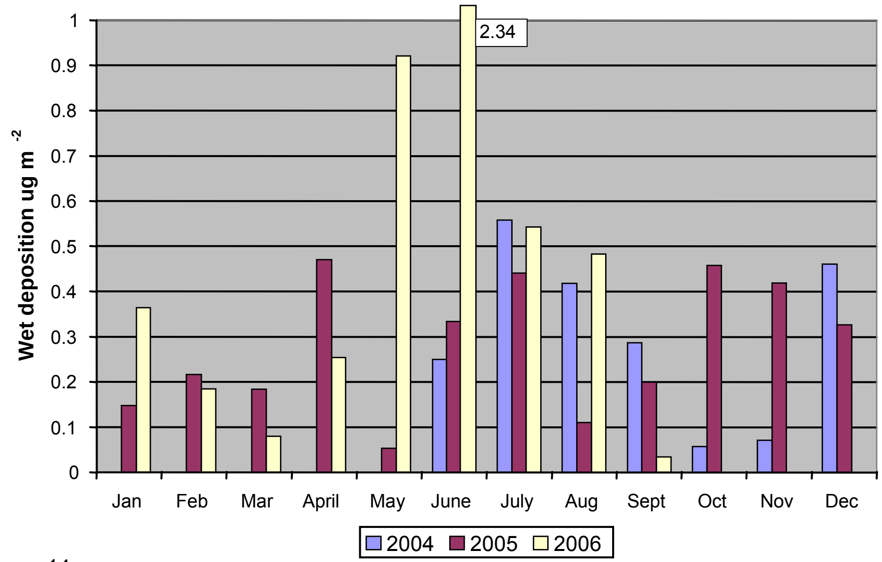


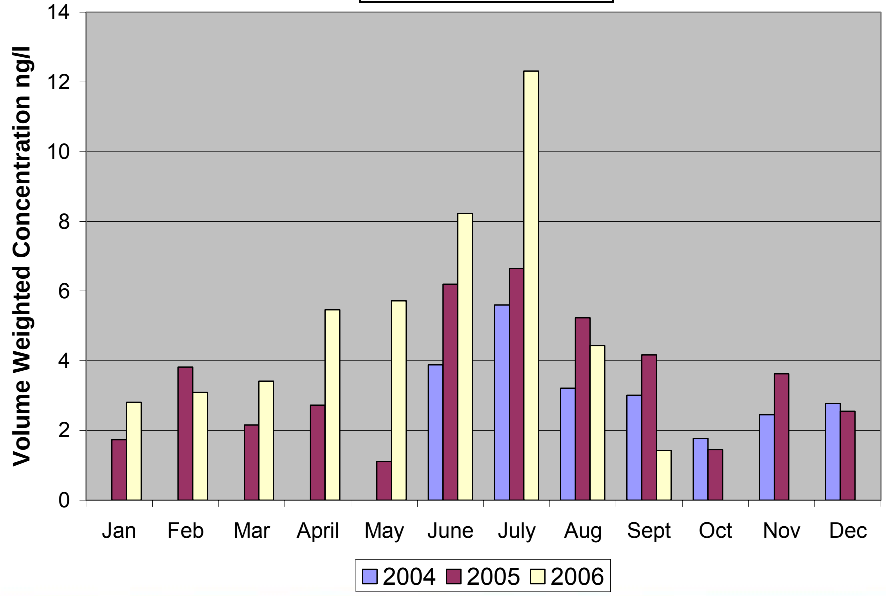


- Mercury concentrations highest in organic layer
- Soils in the wetland area had the lowest mercury concentration but the highest amount of organic matter
- TGM concentrations from the surface to 5 cm depth were best correlated with soil mercury flux (Sigler and Lee, 2006)

Wet Deposition

- Event based samples
- Highest concentrations and deposition in June and July
- Annual wet deposition from 2004 through mid 2006 was 6.57 µg m^{-2}
- Monthly deposition totals is often driven by several large events





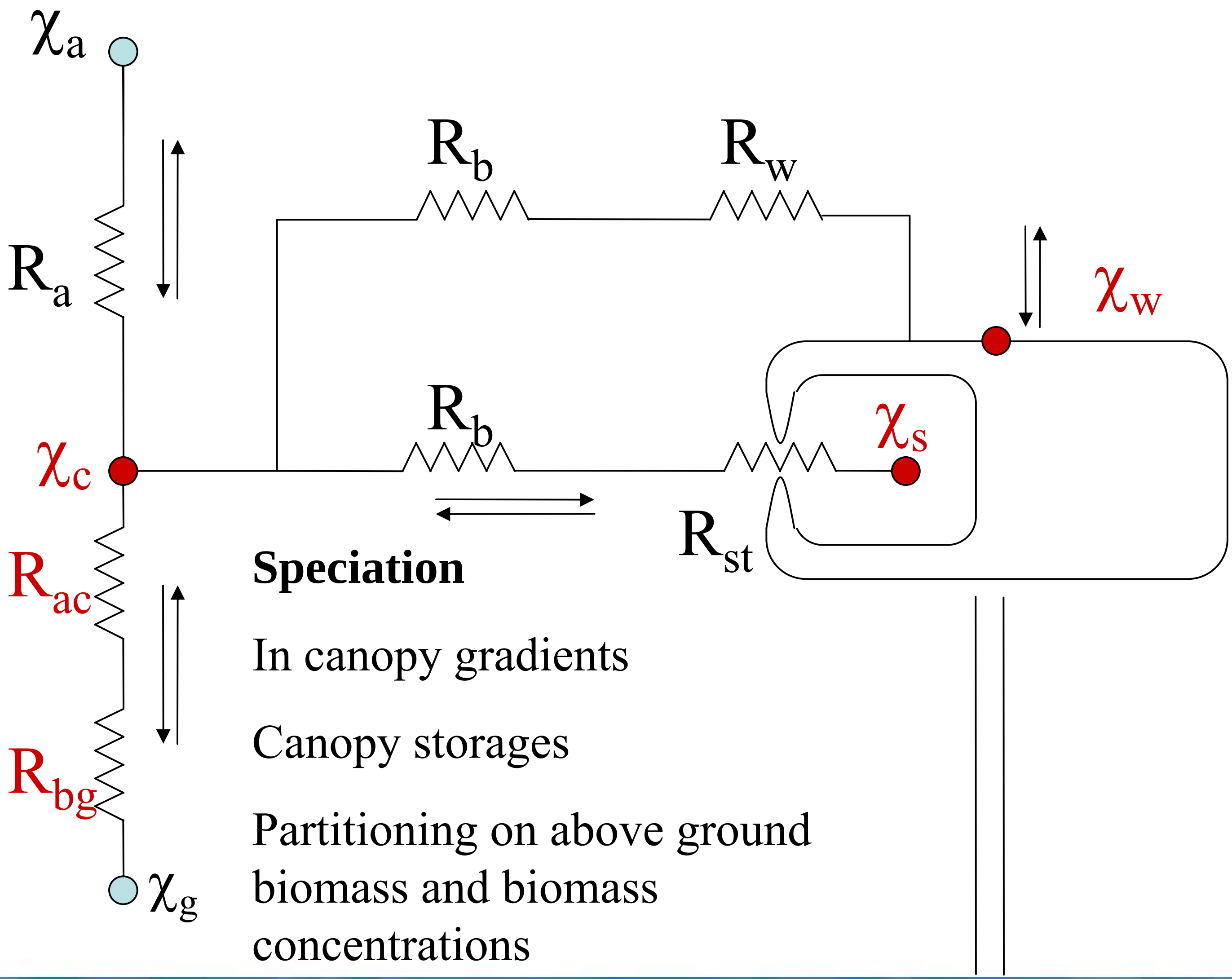
Conclusions

- Seasonality in the bi-directional TGM flux was documented over a red maple canopy
- Under canopy soils were a constant emissions source and the largest pool of mercury
- Under story leaves had a higher mercury concentration than more exposed leaves
- The atmospheric-canopy compensation point was lowest during dry unstable conditions
- Mercury concentrations in the soils were lower in the wetlands
- Soil – canopy – atmosphere mercury exchange needs further investigation

Modeling Needs

- Speciated flux measurements
  - Over a variety of land cover types
  - Are fluxes over other forest canopies this large?
- Atmosphere-canopy-soil source/sink relationship
  - In canopy concentrations, vegetative and soil concentrations
  - Where mercury deposits in the canopy/soil system will determine how it is re-emitted
  - Leaf level parameters
    - Mesophyll and cuticular concentrations of various species
    - Leaf washing experiments
- Identifying the mechanisms of emissions and deposition under wet canopy conditions
  - Does the presence of water mobilize mercury bound to leaves and soils?
  - Speciated measurements critical

Future modeling needs



Speciation

In canopy gradients

Canopy storages

Partitioning on above ground biomass and biomass concentrations

Disclaimer

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