Wetland Mercury Methylation Declines Rapidly Following Reductions in Sulfate Deposition

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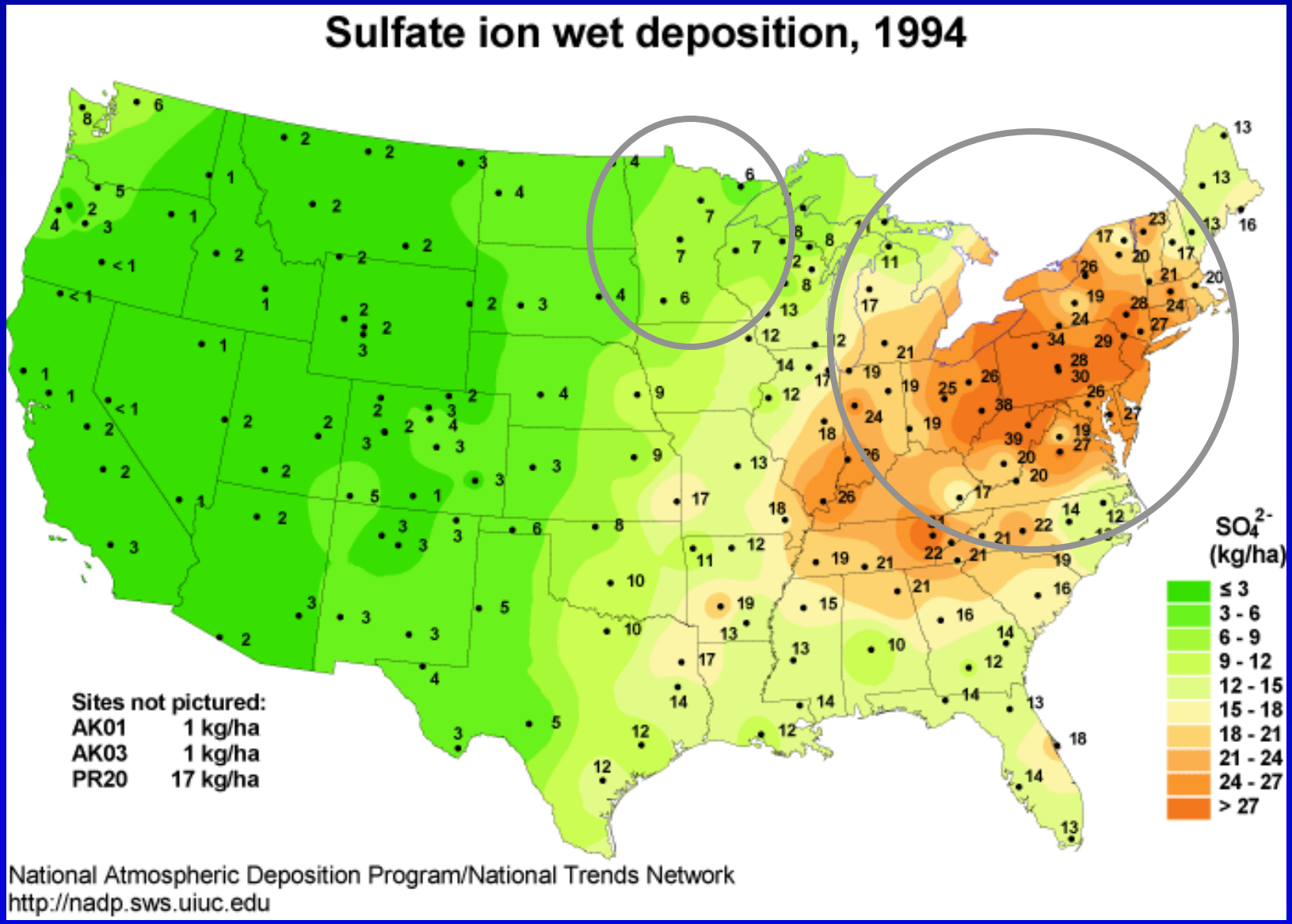
Long-term project – many hands

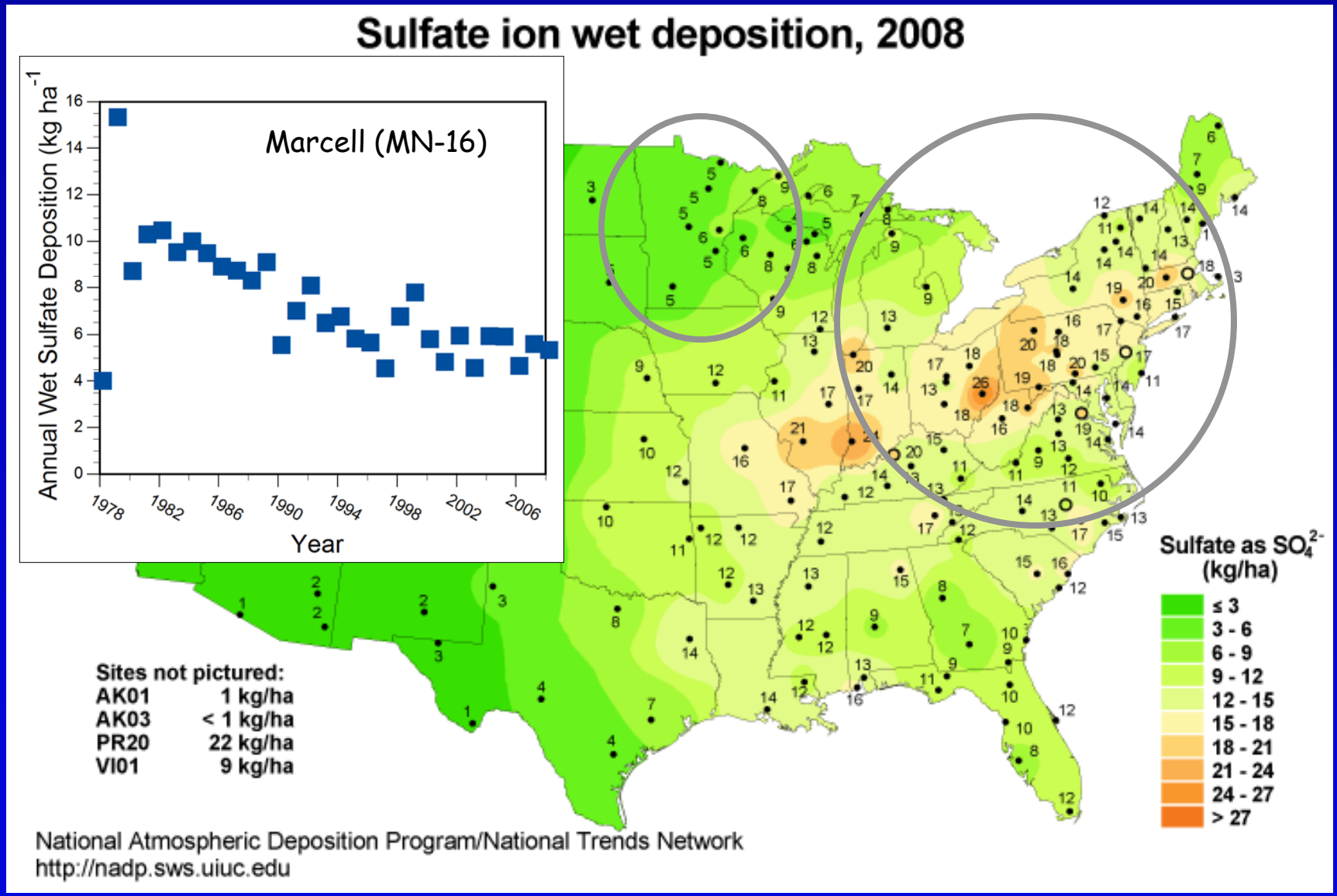
Background

- Many studies have shown a link between bacterial sulfate reduction and methylmercury (MeHg) production in wetlands. (Gilmour et al., 1992, Branfireun et al., 2001; Jeremiason et al., 2006)
- Historically atmospheric sulfate deposition was elevated across broad regions of the US. (http://nadp.sws.uiuc.edu/data)
- Recent studies suggest that declines in atmospheric sulfate deposition may lead to declines in fish mercury concentrations. (Drevnick et al., 2007)
- This research seeks to link experimental increases and reductions in atmospheric sulfate deposition with changes in MeHg production, wetland recovery processes, and consequences for mercury concentrations in biota.

Sulfate Wet Deposition-1994



Sulfate Wet Deposition - 2008

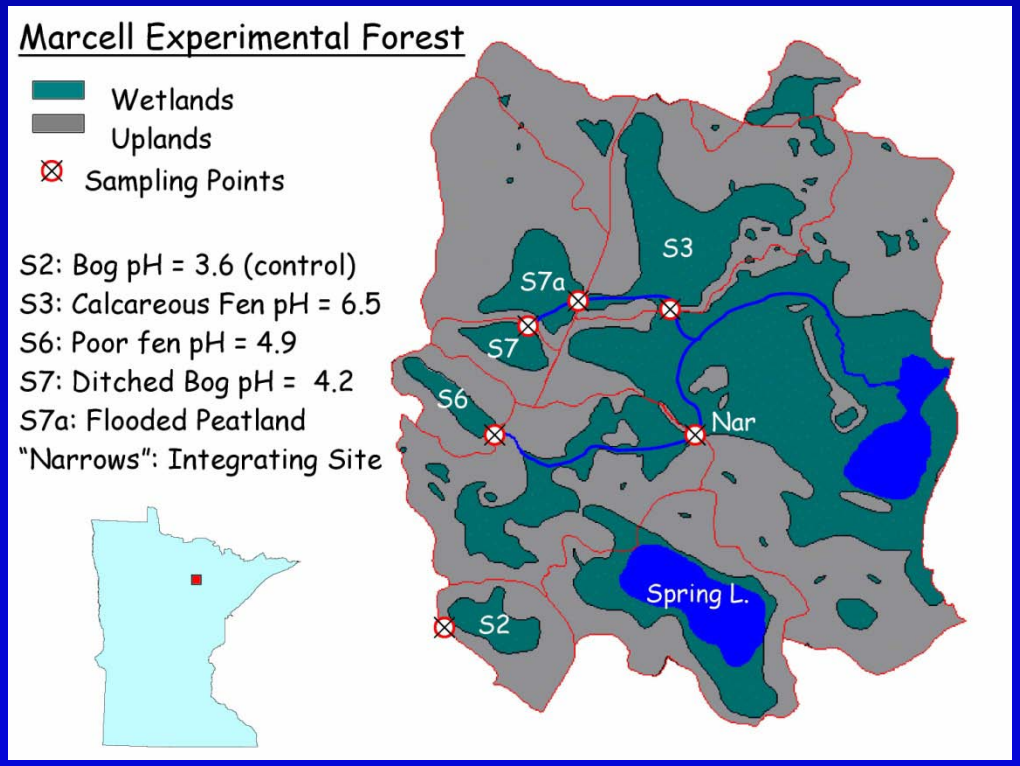


Research Questions

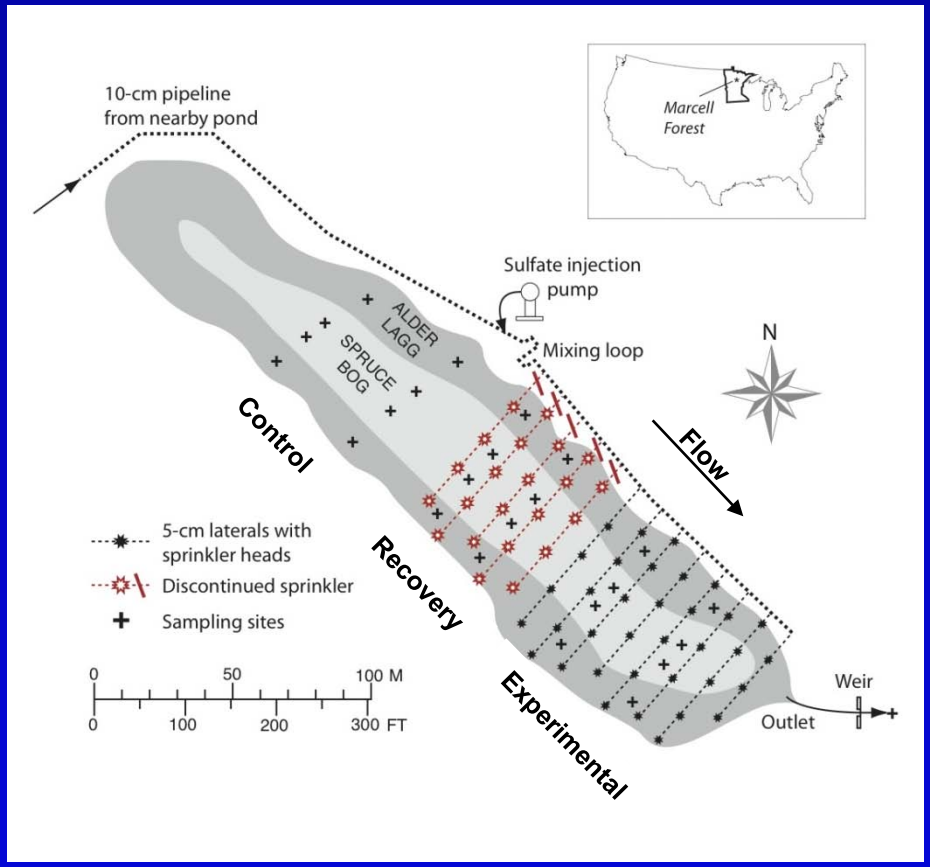
- How does MeHg production change in the porewaters of a boreal wetland when sulfate deposition is increased and when it declines?
- Do solid phase MeHg concentrations respond to declines in sulfate deposition?
- How do variations in precipitation and water level affect the process?
- What are the consequences of increasing/declining sulfate deposition for biota in a boreal wetland?

Study Site

Marcell Experimental Forest
USFS Northern Research Station
(Chippewa National Forest)



Experimental Design

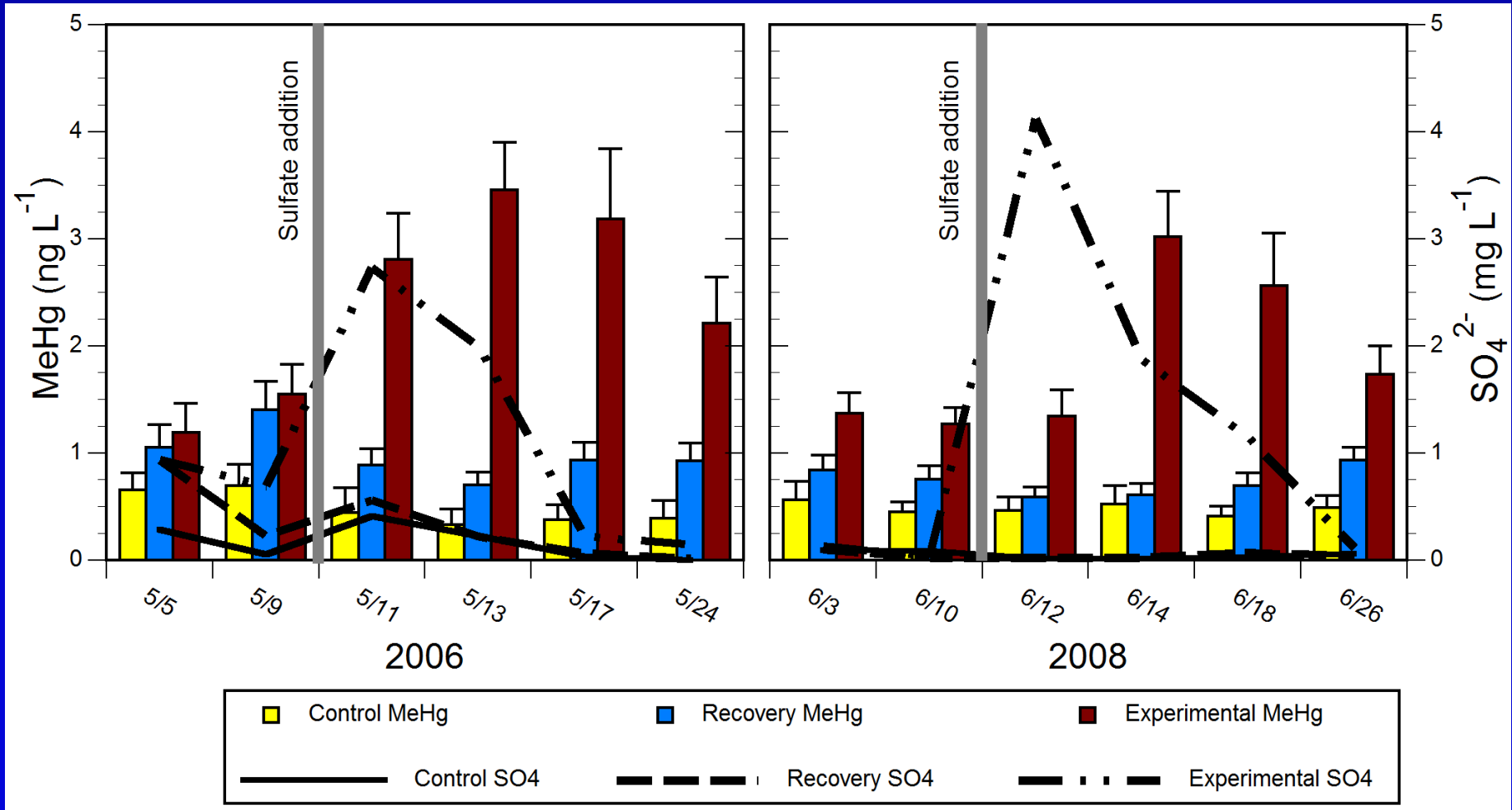


- 3 sulfate additions per year (fall 2001-fall 2008).
- Increased annual sulfate deposition rate by 4X ambient.
- Control and experimental treatments.
- Recovery treatment created in 2006.
- Porewater samples collected before and after each sulfate addition.
- Peat cores and invertebrates collected spring 2009.

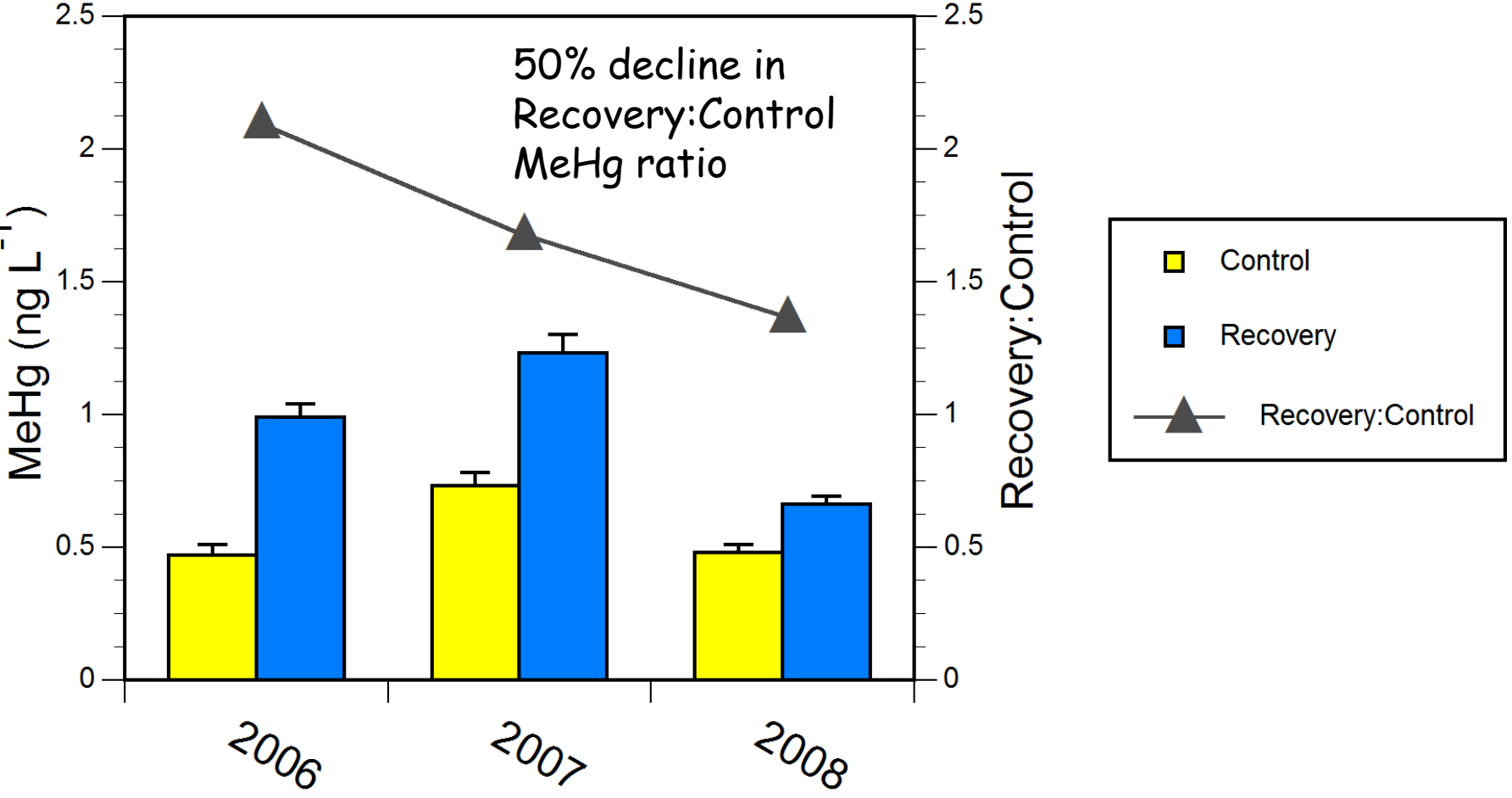
Eight field seasons of simulated rainfall

Eight field seasons of sampling

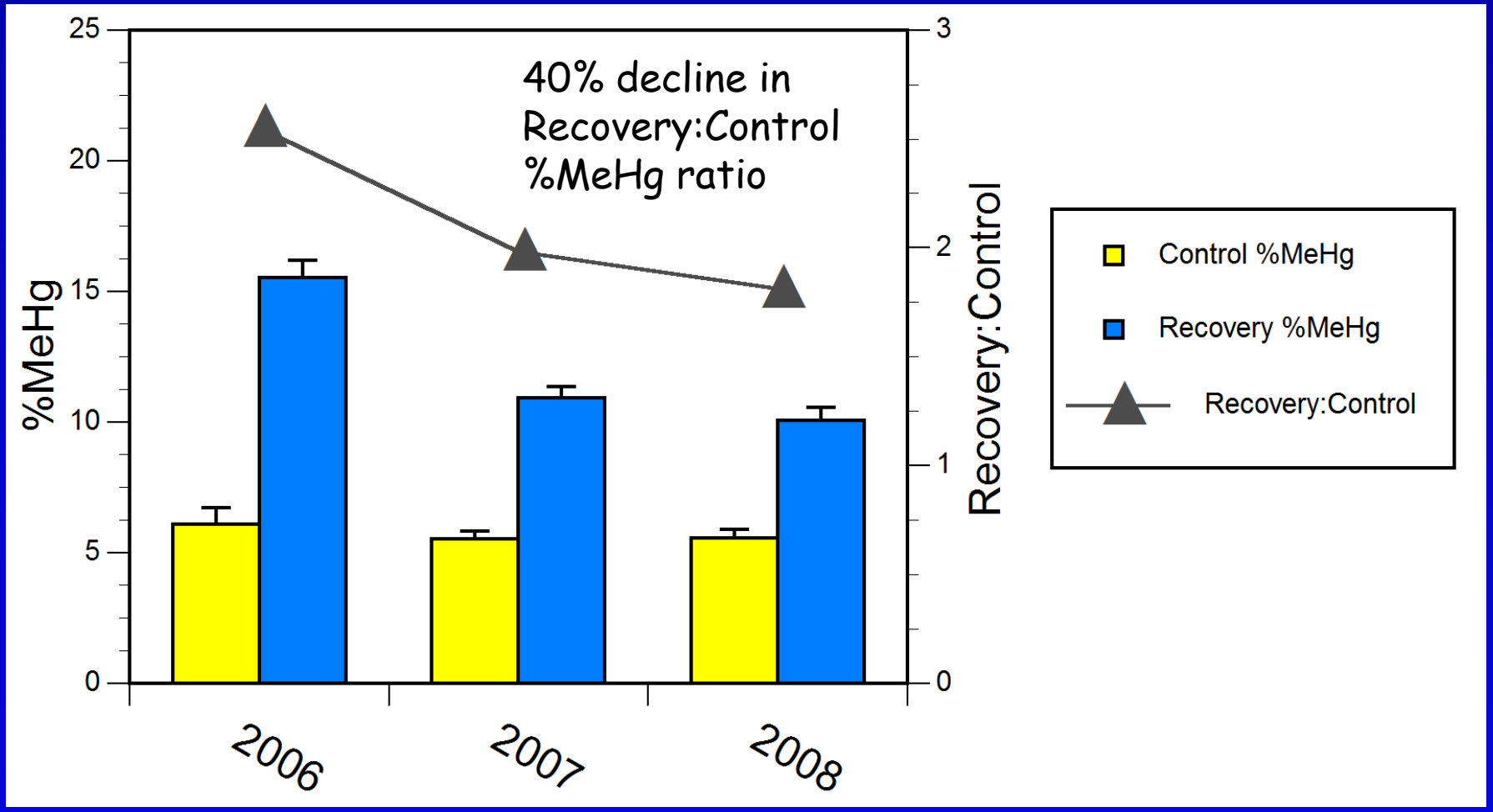
MeHg Response to Sulfate Addition Spring 2006 vs. Spring 2008



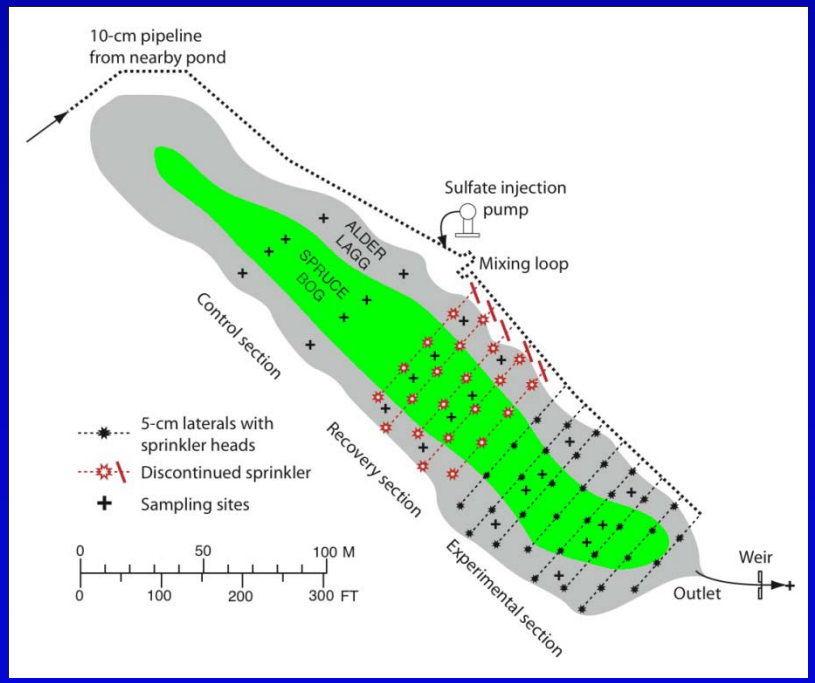
2006-2008 Annual MeHg in Porewater

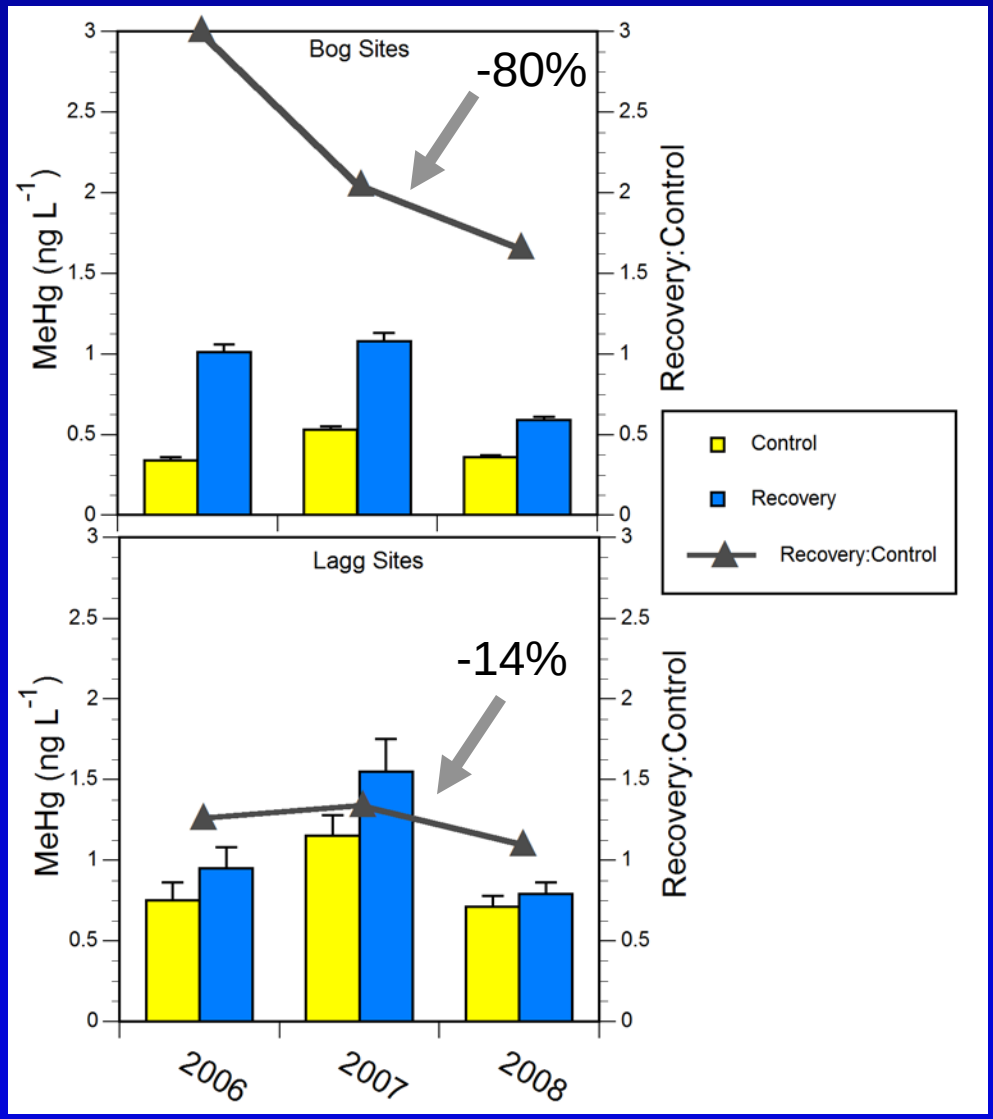


2006-2008 Annual %MeHg in Porewater

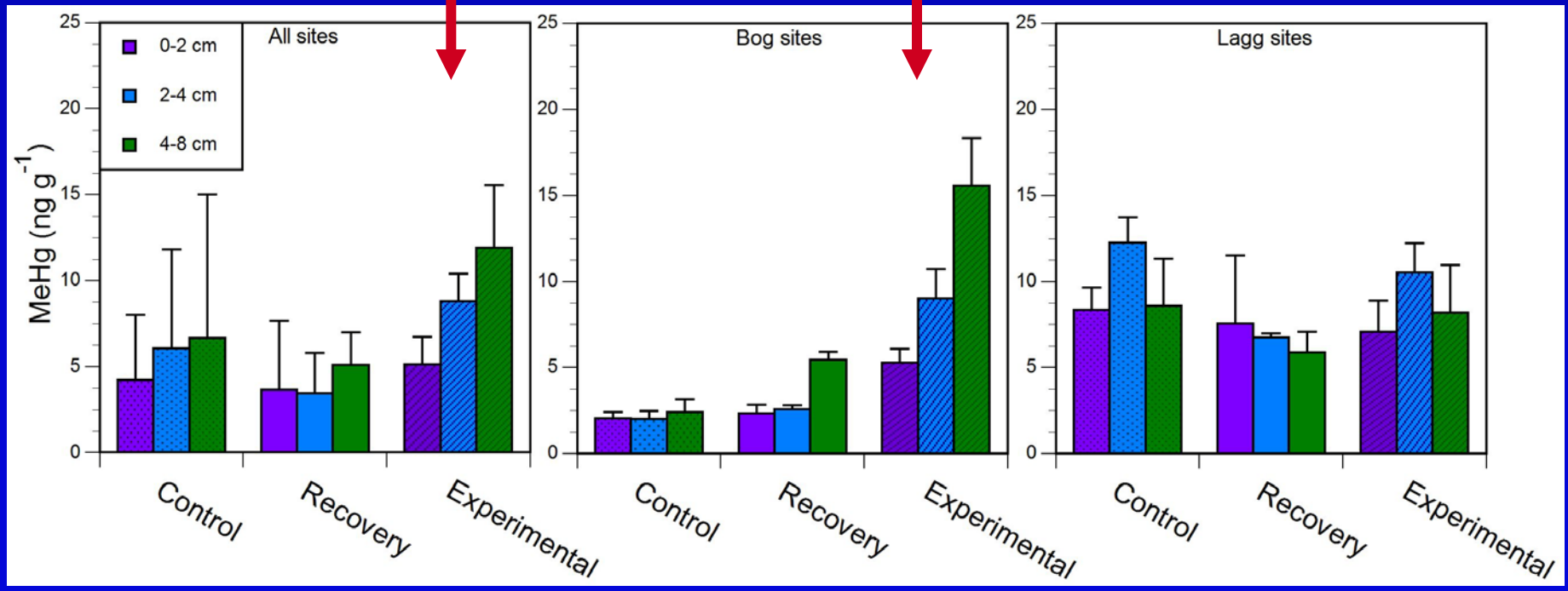


Bog vs Lagg: Influence of Local Hydrology

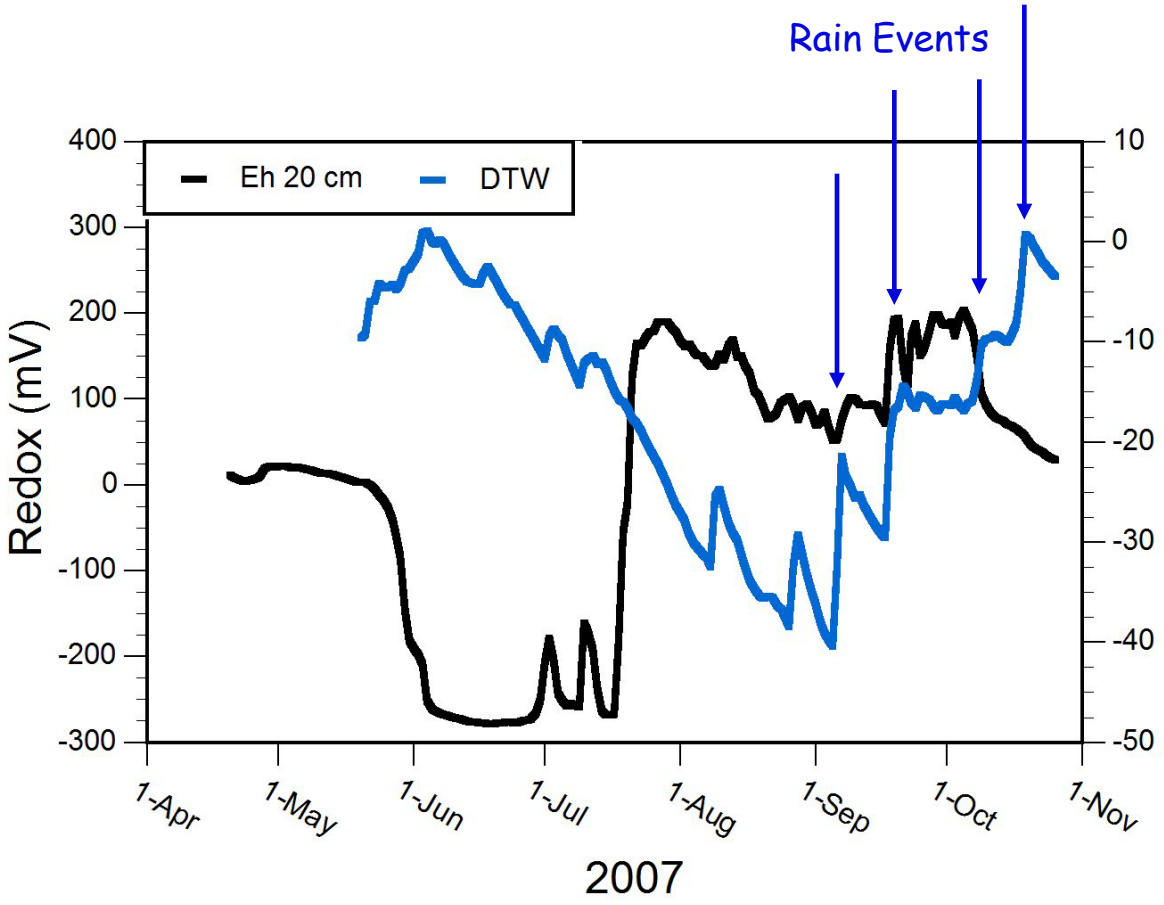




Solid Phase MeHg 2009

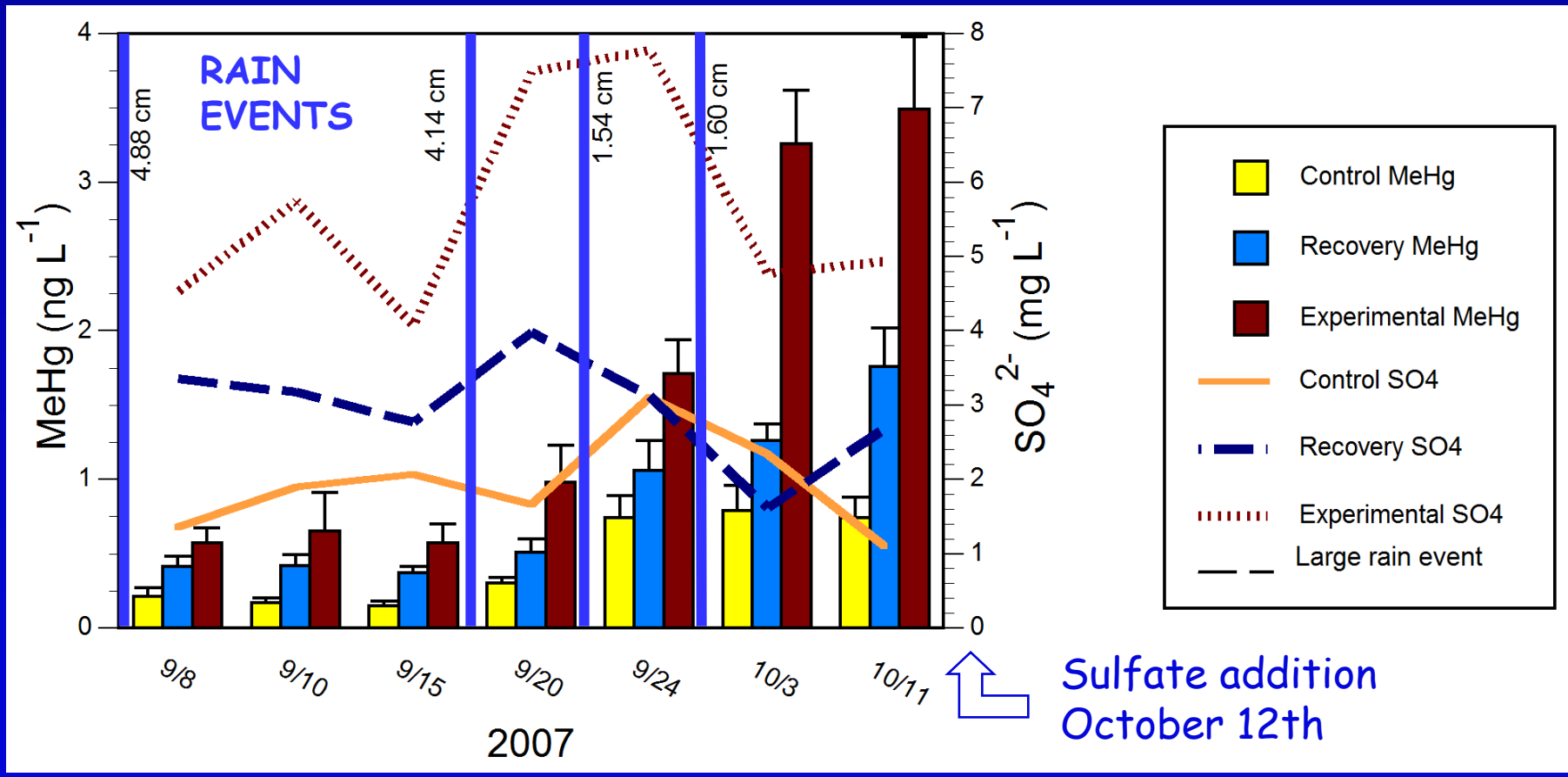


Hydrologic fluctuations



2007 summer drought

2007 fall water table rise

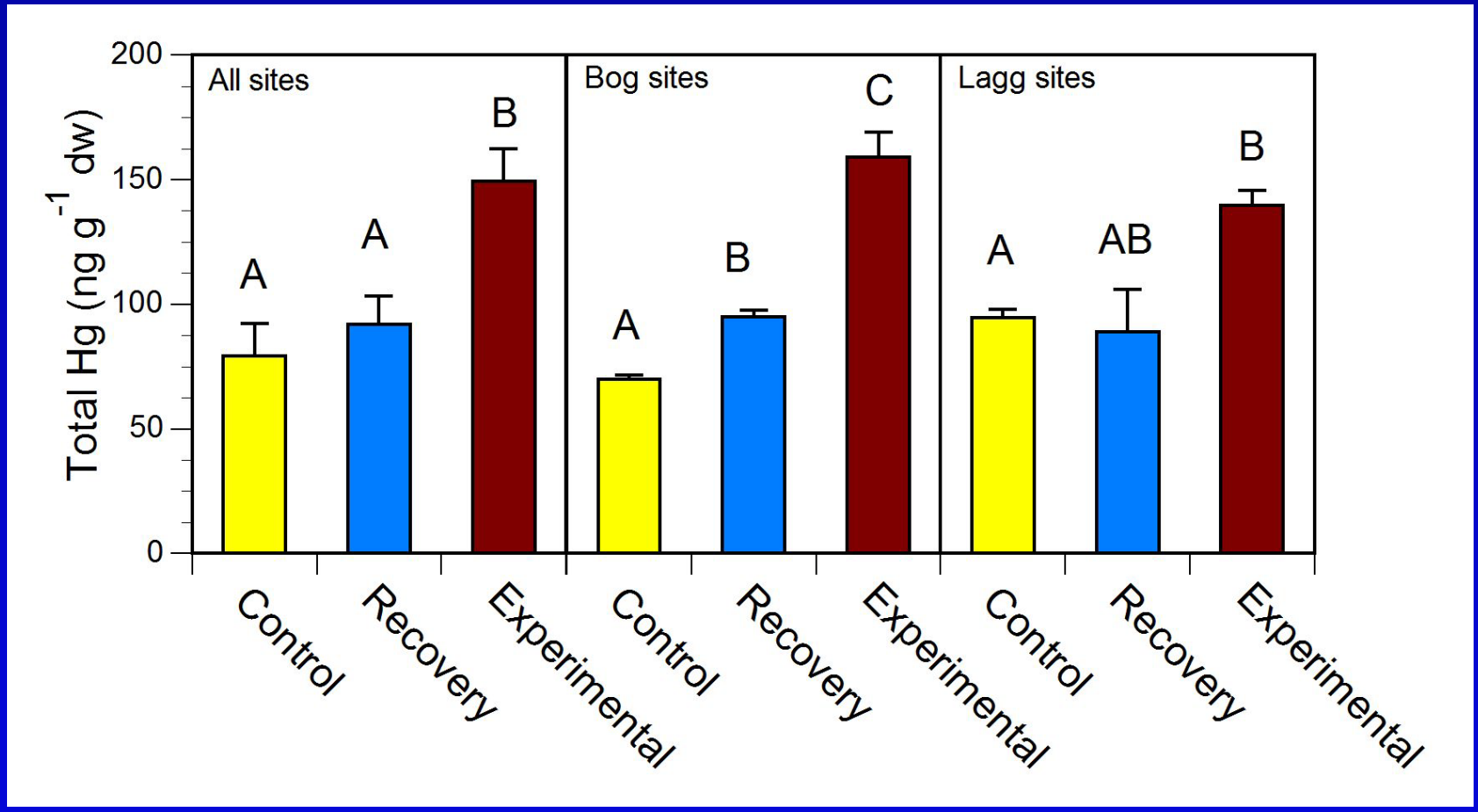


Similar effects observed in water-level mesocosm experiments

Recovery Mechanism

- Water-level fluctuations stimulate methylation by re-oxidizing labile forms of sulfur
- Sulfur pool in peat is almost entirely organic (>98%)
- Amended sulfur becomes increasingly unavailable to recycling ("sulfur aging")
- Ongoing validation by XAS (synchrotron radiation X-ray absorption spectroscopy) of organic-S in peat cores

Biotic Consequences
Mosquito total-Hg – 2009



Conclusions

- MeHg concentrations decline "rapidly" in wetland porewaters and peat following declines in sulfate deposition.
- Previously added sulfate becomes sequestered in increasingly recalcitrant organic pools with time.
- Water-level fluctuations cause re-oxidation of more labile organic-S pool, stimulating secondary methylation pulse
- Preliminary invertebrate data suggest that THg concentrations in certain biota have declined as a result of decreased sulfate deposition.
- Controls on atmospheric sulfur emissions and sulfate deposition could lead to relatively rapid declines in wetland MeHg pools with consequences for mercury accumulation in biota.