Mercury Dynamics in a Coastal Plain Watershed: A Multiple Model Approach

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9th INTECOL International Wetlands Conference, Orlando, Florida, USA, 4 June 2012

United States Environmental Protection Agency

Presentation Outline

- Motivation and background
- Study questions
- Study site and approach
- Results and analysis
- Insights for watershed Hg modeling
- Summary

Motivation for Research

- Increase in watershed-scale Hg cycling research past two decades = important insights on Hg inputs, outputs, and processes in specific regions
- Watershed Hg models important tools for assessing and predicting ecological/human risks of Hg
  - Particularly true for Coastal Plain of US—a region of high methylmercury production and bioaccumulation
- Few spatially-explicit watershed models exist focusing on Hg cycling from landscape to surface waters
- Watershed models that capture wide range of landscape Hg processing are limited

Research Goals

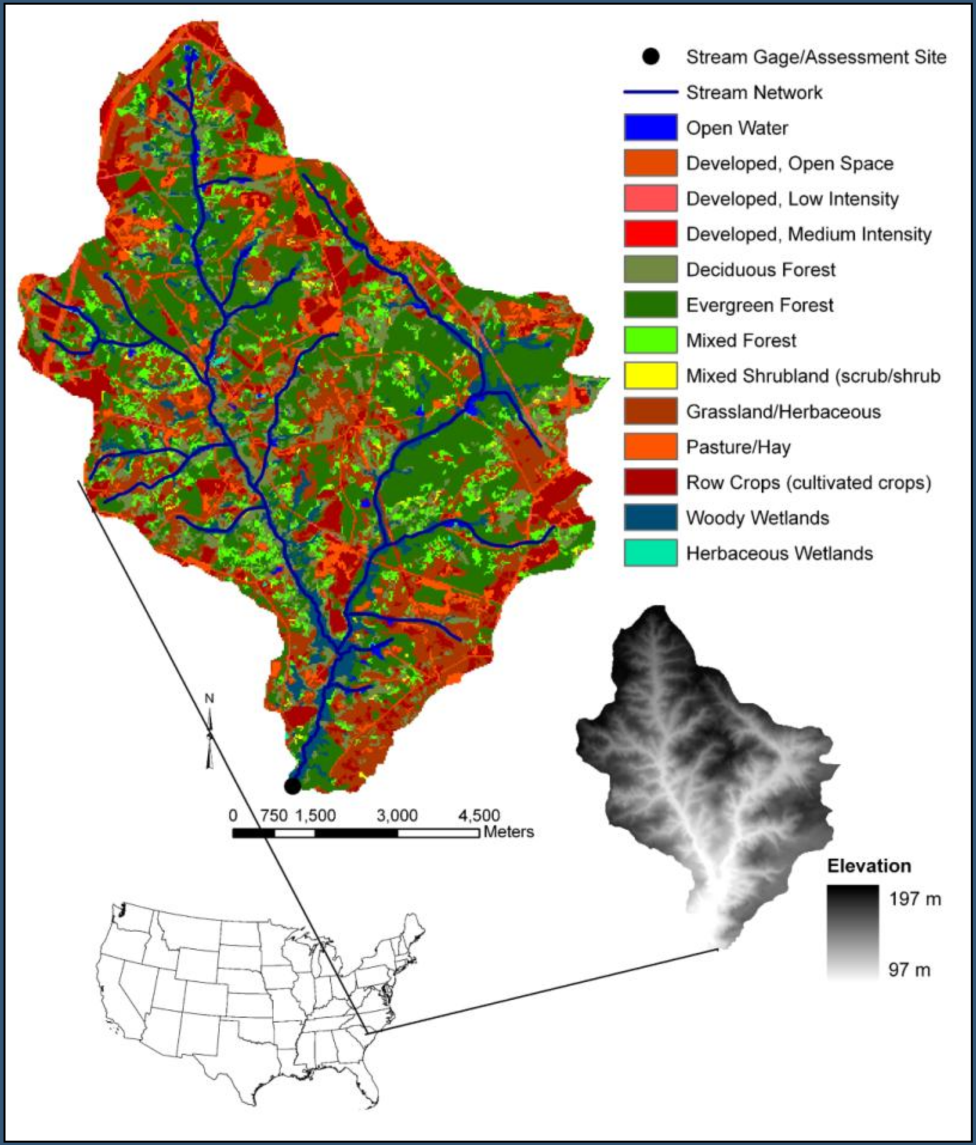
Primary: Assess Hg cycling within a small Coastal Plain watershed (McTier Creek) using multiple watershed models with distinct mathematical frameworks that emphasize different system dynamics

- Simulating total Hg (Hg_T) concentrations and fluxes
- Hg_T first step towards MeHg dynamics

Secondary: Identify current needs in watershed-scale Hg modeling

McTier Creek Watershed, South Carolina, US

- Sand Hills region of Upper Coastal Plain, SC
- 79 km^2 drainage area
- Mixed land cover: 49% forest, 21% grassland and herbaceous, 16% agriculture, 8% wetland, 5% developed, 1% open water
- Shallow groundwater system
  - Low to normal flow: toward stream channel
  - High flow: same with increased area of groundwater-surface water exchange



Approach: Models (2007-2009 Simulation)

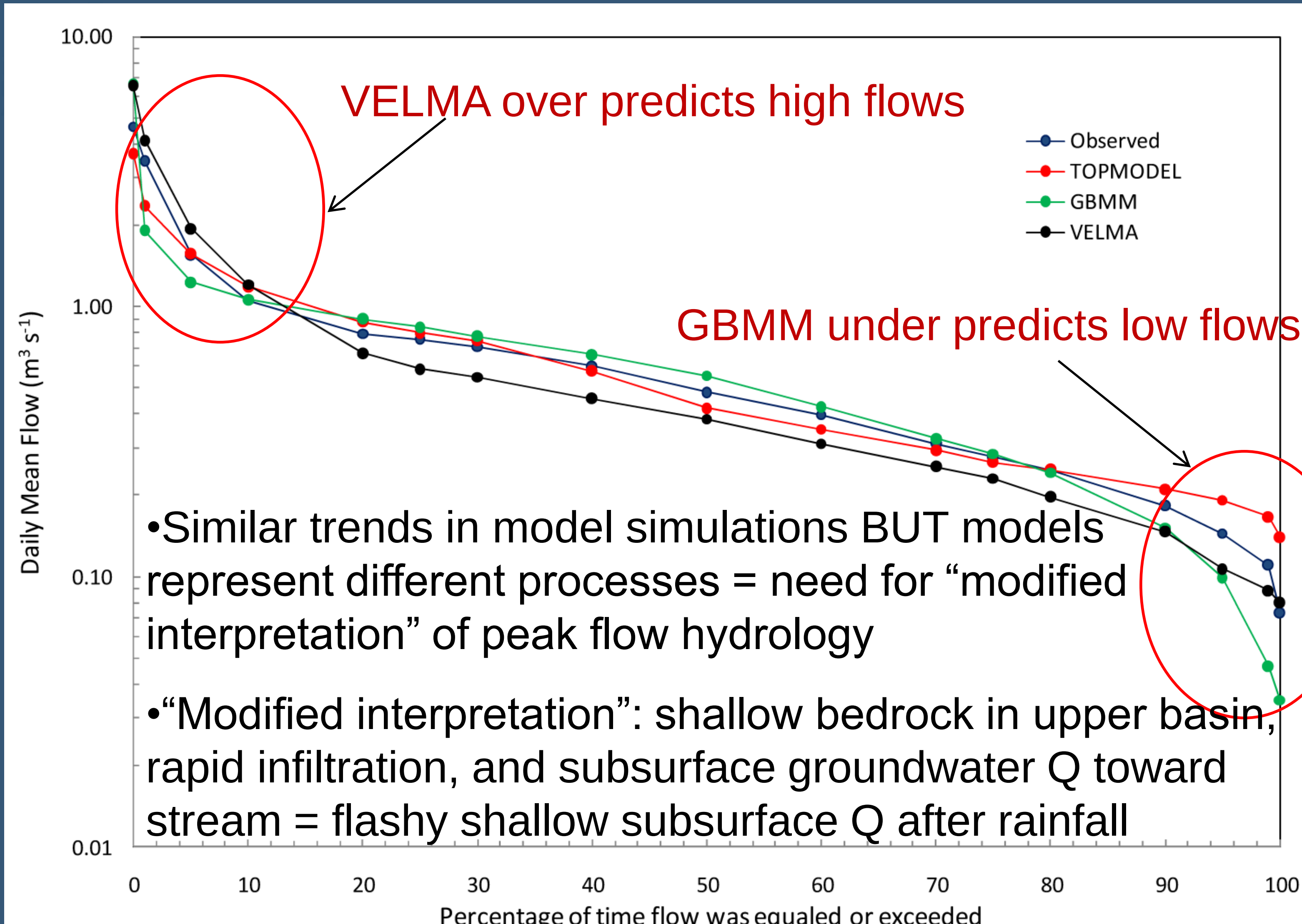
- *Grid Based Mercury Model (GBMM).* Spatially-explicit, process-based. Driven by surface runoff and sediment delivery (K_d for soil water partitioning) = mostly linked to particulate fraction of Hg_T. Provides source contribution from land cover types.
- *Visualizing Ecosystems for Land Management Assessment for Hg (VELMA-Hg).* Spatially-explicit, process-based. Hg_T fluxes associated with multi-soil layer hydrology and C, N, and Hg cycling = mostly linked to dissolved fraction of Hg_T
- *TOPLOAD.* Empirical, based on TOPMODEL hydrology. Identifies flow components contributing to Hg_T fluxes.
- *S-LOADEST (seasonal results not presented today).* Regression-based water quality flux estimator. Applied for seasonal load comparisons.

Approach: Models and Data

- *Data*
  - Daily streamflow at US Geological Survey stream gage (McTier Creek at New Holland)
  - 41 samples at stream gage location (variety of flows):
    - Observed Hg_T (filtered and particulate)
    - Dissolved organic carbon (DOC)
    - Total suspended sediment (TSS)

- Set of tools (models and data) to compare conceptualizations of Hg_T dynamics to characterize Hg_T cycling
  - Data and models mutually informative
  - Parameters not forced beyond realistic values to match observations
  - Potential contributions of processes not included in the models is recognized

Results: Hydrology



- Similar trends in model simulations BUT models represent different processes = need for "modified interpretation" of peak flow hydrology
- "Modified interpretation": shallow bedrock in upper basin, rapid infiltration, and subsurface groundwater Q toward stream = flashy shallow subsurface Q after rainfall

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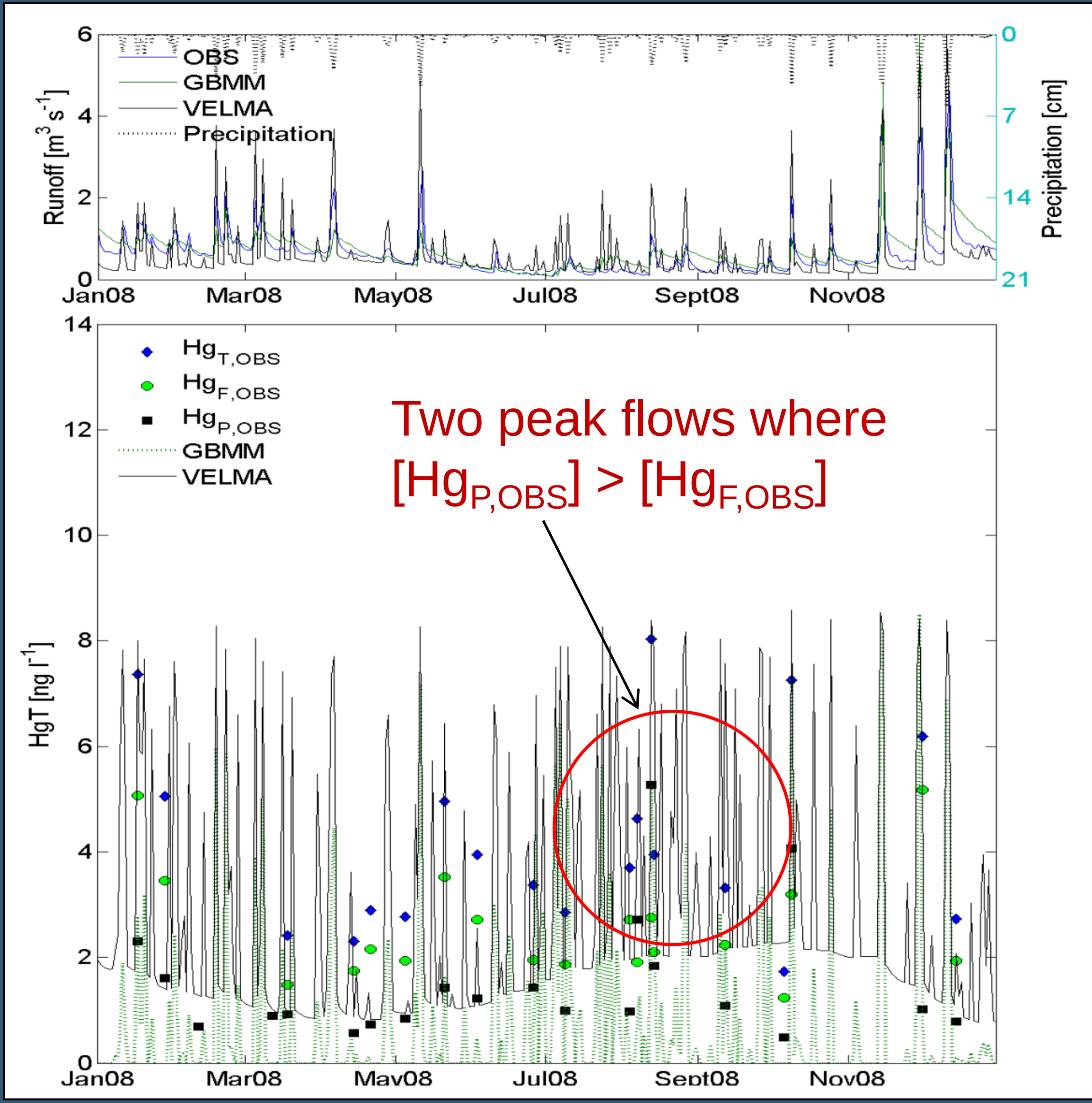
Hg_T Concentration: 2008

- ❑ $[Hg_{F,OBS}]$ > two-thirds $Hg_{T,OBS}$
- ❑ Mean $[Hg_{T,GBMM}]$ similar to mean $[Hg_{P,OBS}]$: 1.18 ng L^{-1} & 1.27 ng L^{-1}
- ❑ Mean $[Hg_{T,VELMA}]$ similar to mean $[Hg_{T,OBS}]$: 3.54 ng L^{-1} & 3.92 ng L^{-1}



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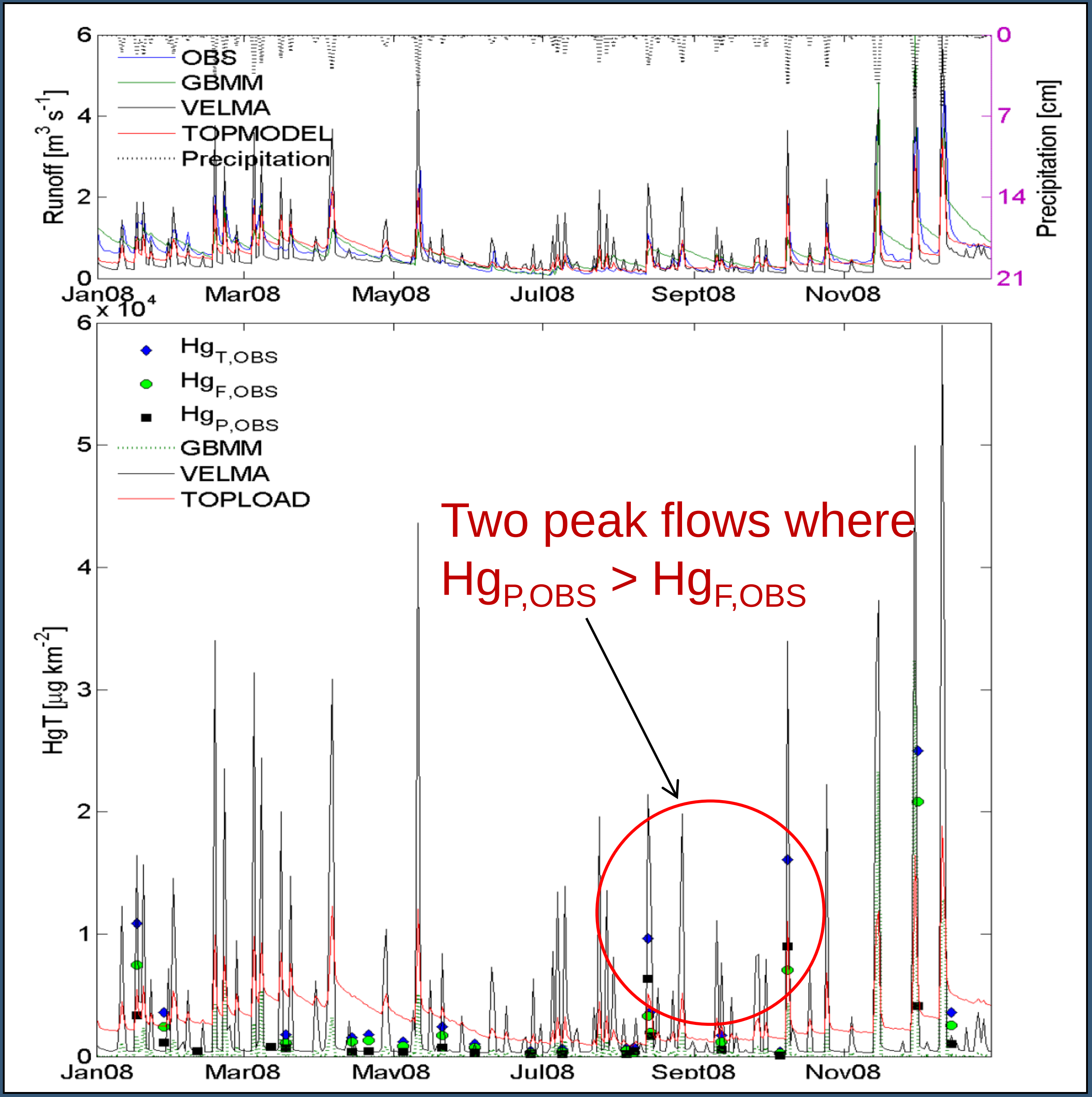
Hg_T Fluxes: 2008

- $Hg_{F,OBS} > Hg_{P,OBS}$ for 39 of 41 sampling events
- Mean $Hg_{T,GBMM}$ fluxes (434 µg km^2 d^{-1}) low compared to other average Hg_T fluxes (flow duration = lowest low flows)
- Mean $Hg_{T,VELMA}$ fluxes higher (4438 µg km^2 d^{-1}) than all other modeled estimates (flow duration = highest high flows)



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$Hg_{T,GBMM}$

- $Hg_{T,GBMM}$ fluxes strongly linked to streamflow (OBS & GBMM, > 1.5 m^3 s^{-1})
  - Consistent with "modified conceptualization" of hydrology = flashy groundwater response under high flow becomes important for Hg transport

- $Hg_{T,GBMM}$ strongly linked to TSS_{OBS} and TSS_{GBMM}

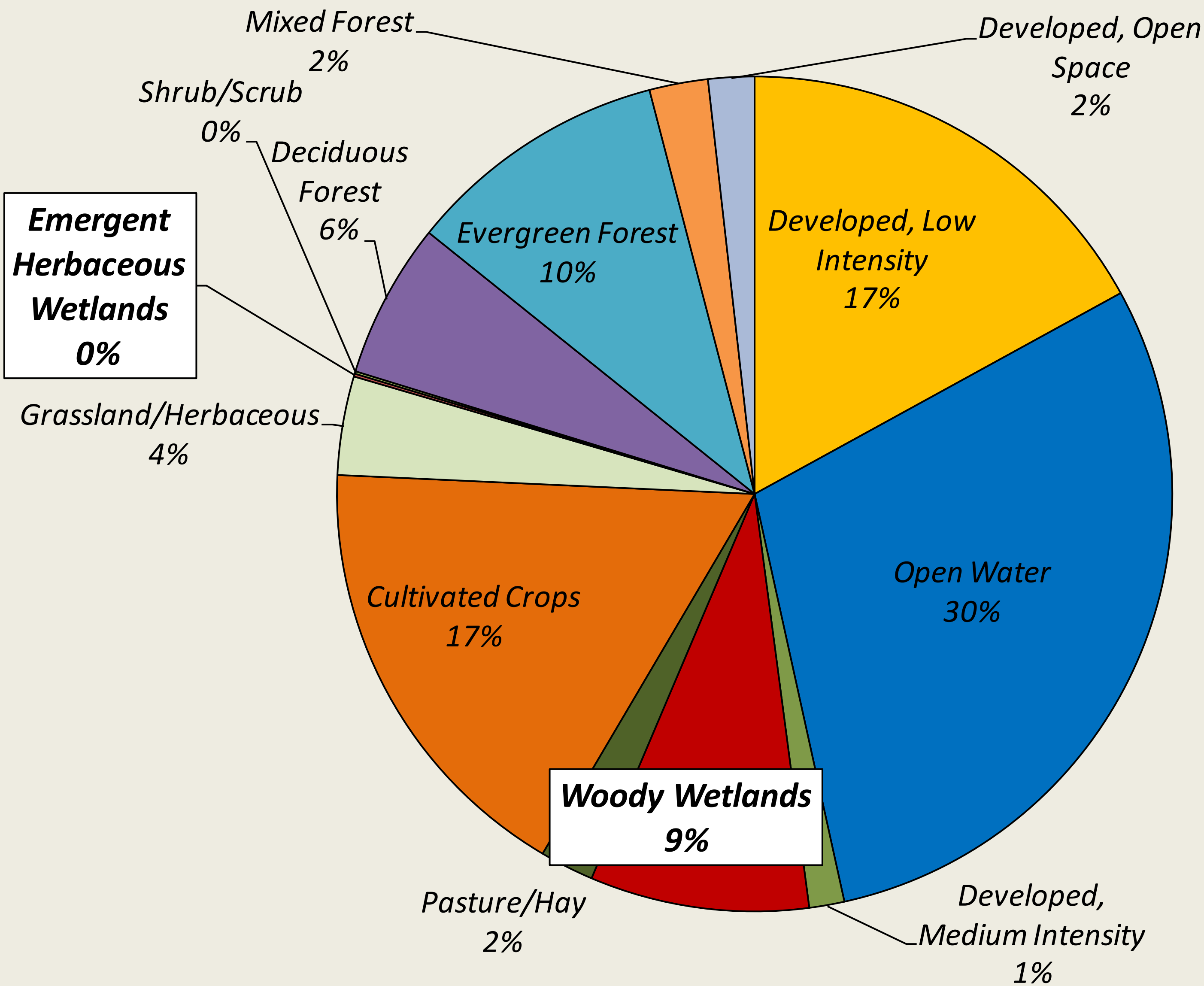
- $Hg_{T,GBMM}$ threefold lower than average $Hg_{P,OBS}$

  - Surface erosion important for Hg_T transport under high flow but need additional interpretations of in-stream Hg_T dynamics

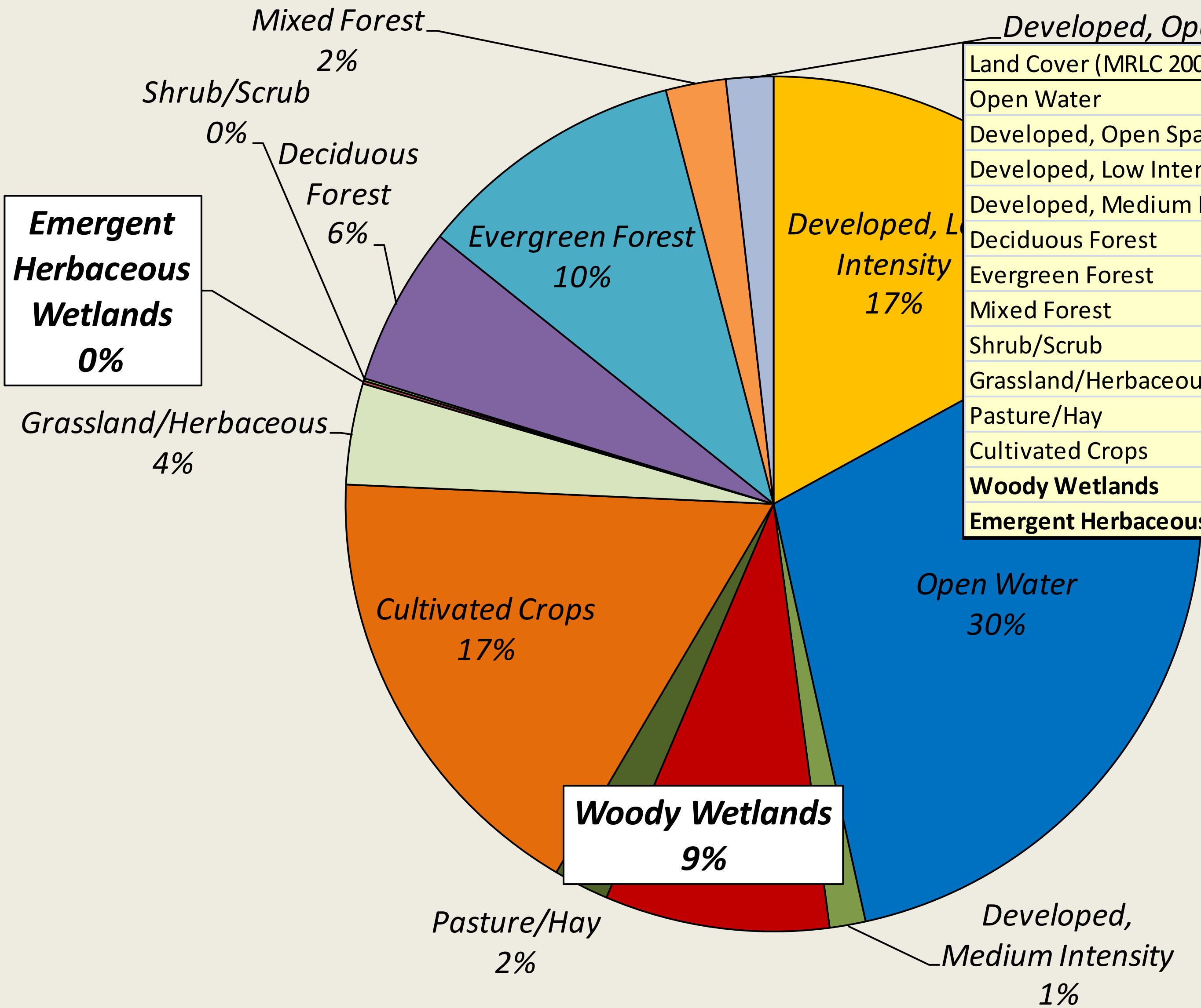
Pearson Correlation Coefficients

Concentrations	$Hg_{T,OBS}$	$Hg_{F,OBS}$	$Hg_{P,OBS}$	TSS_{OBS}	TSS_{GBMM}	$Flow_{OBS}$	$Flow_{GBMM}$
$Hg_{T,GBMM}$			0.65***	0.60***	0.87***	0.30*	0.36***
Fluxes	$Hg_{T,OBS}$	$Hg_{F,OBS}$	$Hg_{P,OBS}$	TSS_{OBS}	TSS_{GBMM}	$Flow_{OBS}$	$Flow_{GBMM}$
$Hg_{T,GBMM}$	0.62***	0.41**	0.89***	0.78***	0.84***	0.55***	0.75***
** is p<0.05, **p<0.01, ***p<0.0001*							
all relationships with observed data, n = 41; relationships among modeled data only, n=841							

GBMM: Land Cover Contributions to Hg_T Load (2007-2009) in McTier Creek Watershed



GBMM: Land Cover Contributions to Hg_T Load (2007-2009) in McTier Creek Watershed



Land Cover (MRLC 2001)	% of Watershed
Open Water	1.09%
Developed, Open Space	4.08%
Developed, Low Intensity	0.47%
Developed, Medium Intensity	0.05%
Deciduous Forest	12.88%
Evergreen Forest	30.76%
Mixed Forest	6.26%
Shrub/Scrub	0.50%
Grassland/Herbaceous	21.38%
Pasture/Hay	5.17%
Cultivated Crops	9.37%
Woody Wetlands	**7.80%**
Emergent Herbaceous Wetlands	**0.18%**

$Hg_{T,VELMA}$

- Dissolved Hg dynamics: Two different Hg/DOC interactions

1. Increase in $Hg_{T,VELMA}$, decrease in DOC_{VELMA}
   - Model structure (VSA flow dominance following high rainfall)
   - Direct runoff of high $[Hg_T]$, low [DOC] rain from VSAs
   - Low interaction of runoff with DOC in surface soils

2. Increase in $Hg_{T,VELMA}$, increase in DOC_{OBS} and $Hg_{T,OBS}$
   - DOC-bound Hg removal from floodplain soils following rainfall event

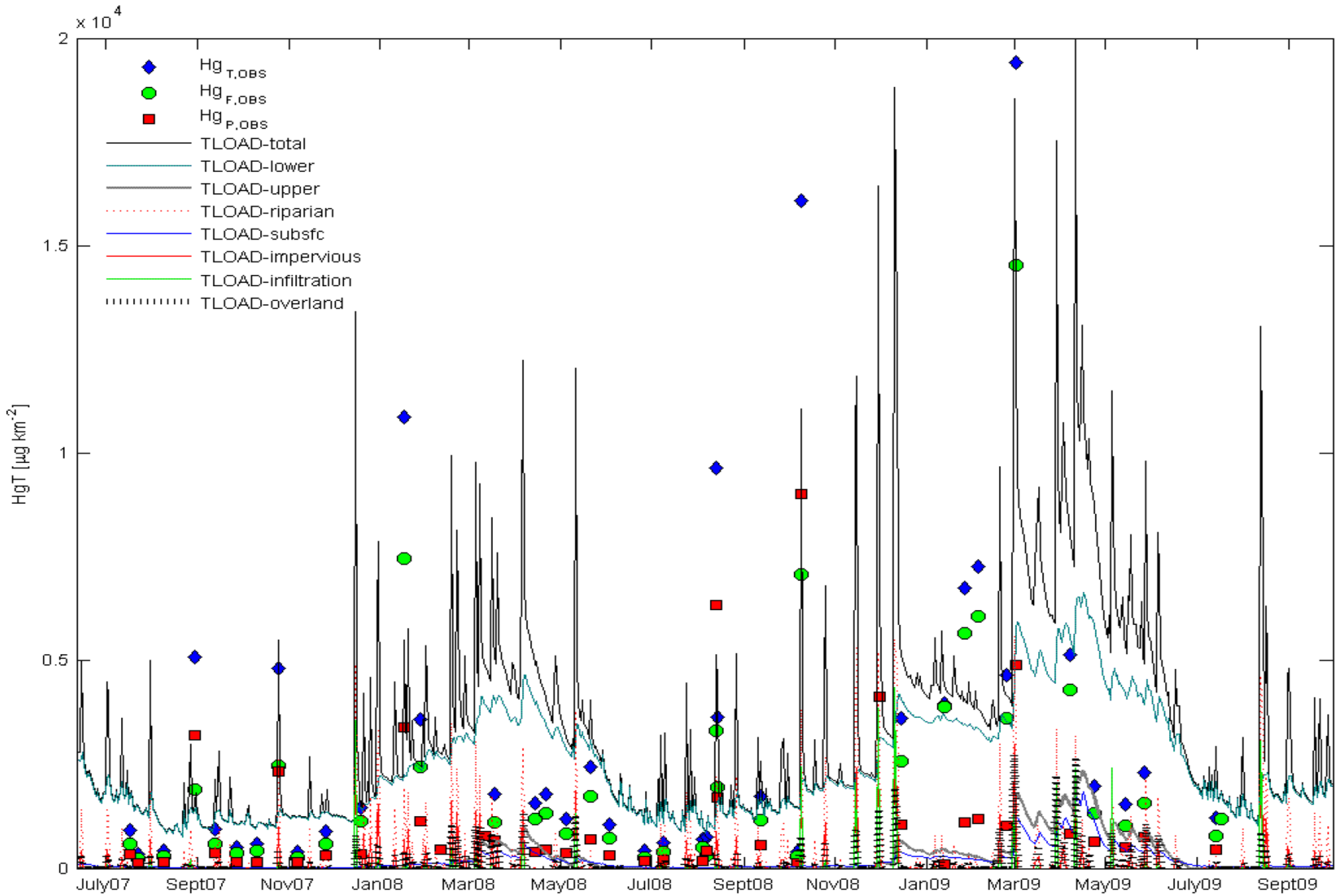
- *DOC-bound Hg more important during baseflow conditions*

Pearson Correlation Coefficients

Concentrations	$Hg_{T,OBS}$	$Hg_{F,OBS}$	$Hg_{P,OBS}$	DOC_{OBS}	DOC_{VELMA}	$Flow_{OBS}$	$Flow_{VELMA}$
$Hg_{T,vELMA}$			0.58***	0.62***	-0.55**	0.31*	0.64***
Fluxes	$Hg_{T,OBS}$	$Hg_{F,OBS}$	$Hg_{P,OBS}$	DOC_{OBS}	DOC_{VELMA}	$Flow_{OBS}$	$Flow_{VELMA}$
$Hg_{T,vELMA}$	0.59***	0.38*	0.86***	0.49**		0.51**	0.97***
** is p<0.05, **p<0.01, ***p<0.0001*							
all relationships with observed data, n = 41; relationships among modeled data only, n=841							

TOPLOAD Baseflow Hg_T fluxes driven by saturated subsurface flows (TLOAD-lower).

Peak Hg_T fluxes = increased contributions from riparian areas and shallow subsurface flows.



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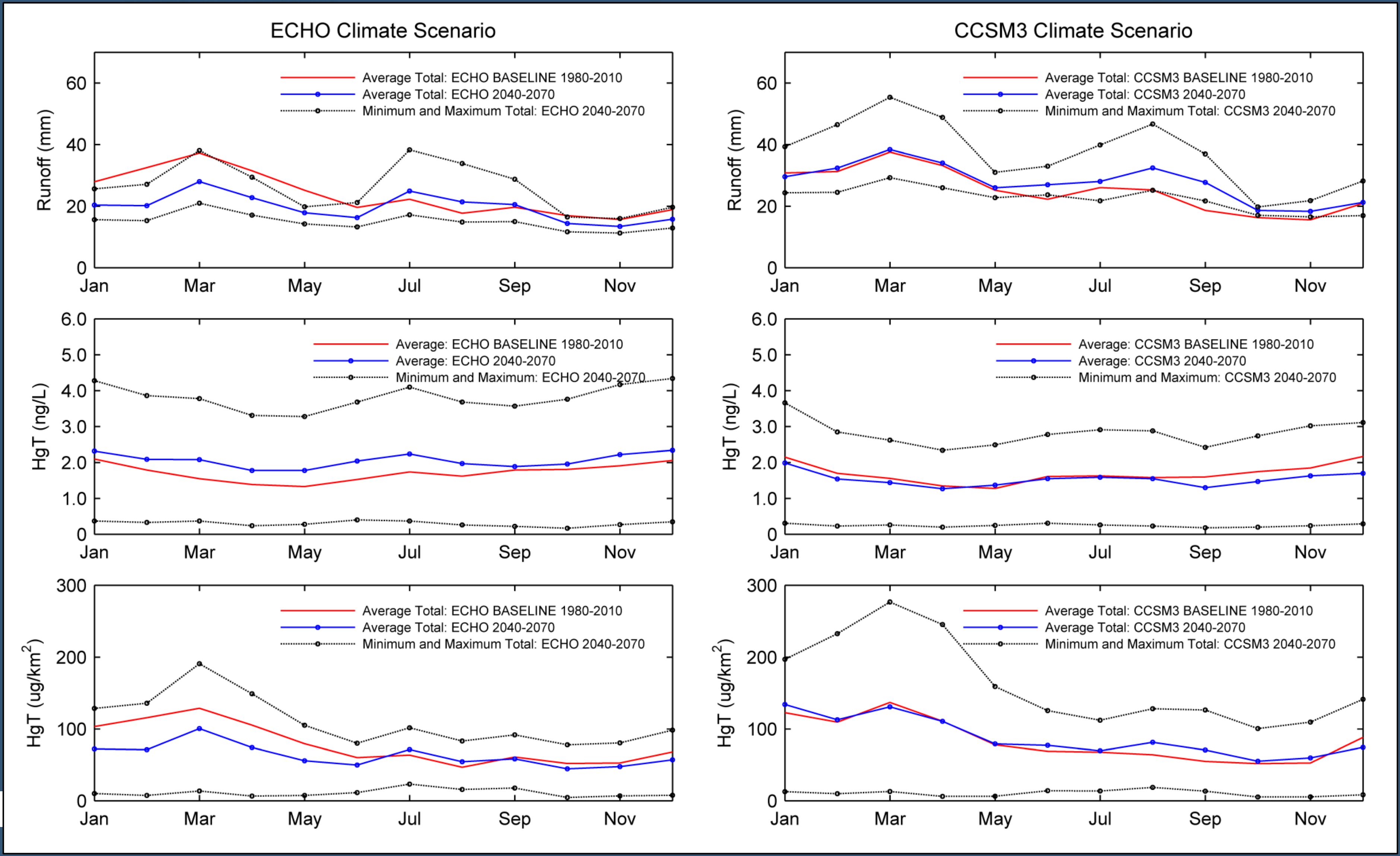
Multiple Watershed Models: Experiment with Climate Change Scenario Analyses



Watershed Hg Modeling: Implications and Future Advances

- GBMM use in highly erodible landscape (e.g., agricultural)
- VELMA-Hg use in settings where DOC-bound Hg important (e.g., forests)
- Additional processes needed in models: methylation, sulfur dynamics, variables that increase availability of other Hg species (e.g, pH, Fe, size/quality of OM), wetland cycling, in-stream processes
- Hydrological model improvements – links to groundwater models and use of newest advances in hydrologic modeling

Summary: Key Findings

- $Hg_{F,OBS}$ approximately two-thirds average $Hg_{T,OBS}$
- *GBMM*: Shallow, subsurface flow and overland flow potentially important transport mechanisms of particulate Hg following high rainfall events
- *GBMM*: Other in-stream processes could also be important (bank erosion, sediment resuspension) but not currently part of models
- *VELMA-Hg*: Dissolved Hg likely directly transported from VSAs in watershed following high rainfall events
- *VELMA-Hg*: DOC-bound Hg more important during baseflow conditions
- *TOPLOAD*: Saturated subsurface flow important for HgT fluxes during baseflow
- Many advancements needed in science of watershed Hg modeling

Thank you!

Questions?

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For further details see: Golden, HE, CD Knightes, CA Conrads, GM Davis, TD Feaster, CA Journey, ST Benedict, MA Brigham, and PM Bradley. 2012. *Characterizing mercury concentrations and fluxes in a Coastal Plain watershed: Insights from dynamic modeling and data*. Journal of Geophysical Research: Biogeosciences: 117, doi:10.1029/2011JG001806