Seasonal Variation in Pathways of Atmosphere-Land Exchange of Mercury in a Northern Hardwood Forest

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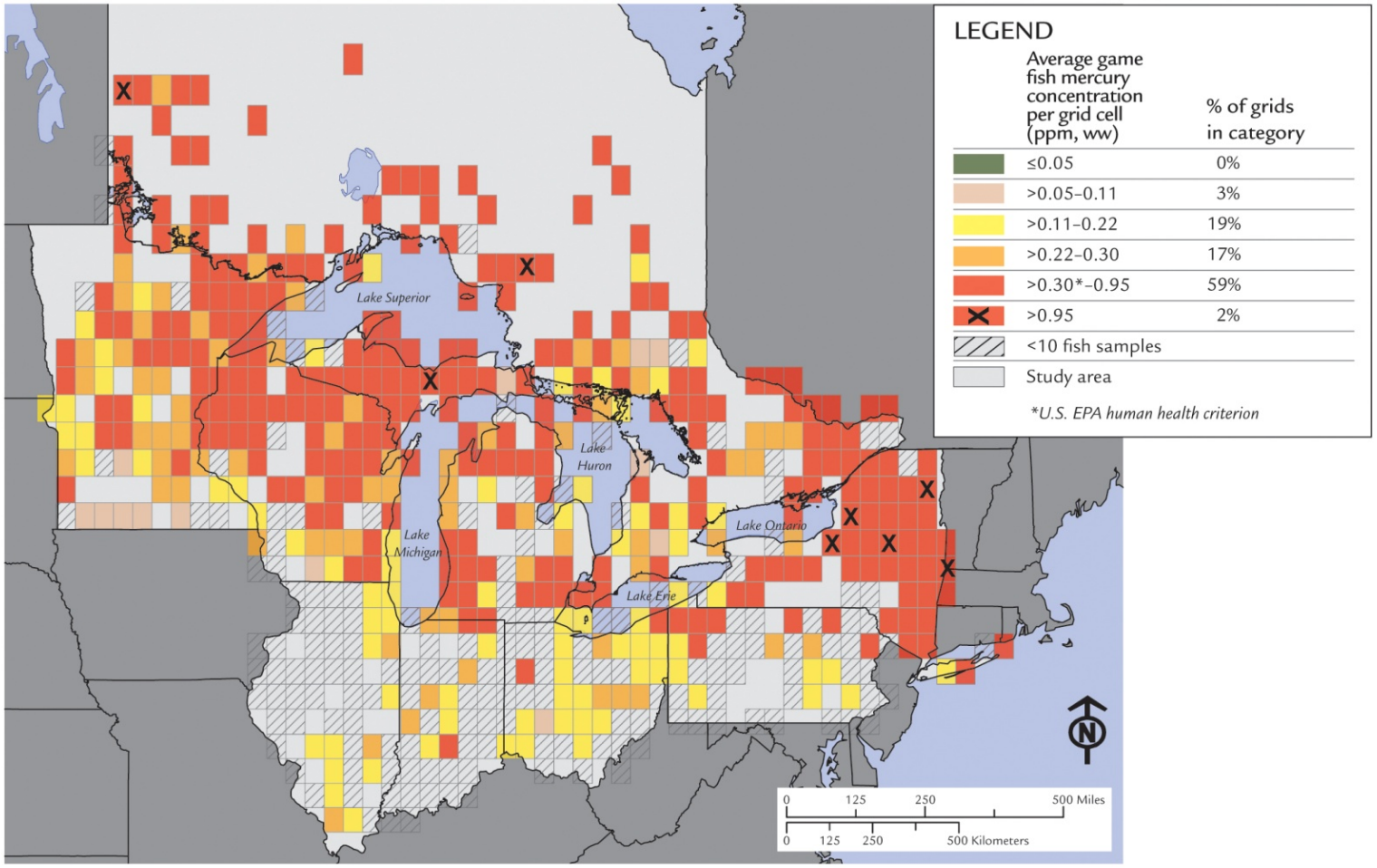
Introduction

- Mercury (Hg) is a toxic pollutant
- Exposure to Hg:
  - Consumption of contaminated aquatic organisms (fish)
- Emission sources of Hg:
  - Natural emissions (direct and secondary emissions): 5207 Mg/yr
  - Anthropogenic emissions (mostly industrial emissions): 2320 Mg/yr (Pirrone et al., 2010)
- Forms of Hg in the atmosphere:
  - GEM (gaseous elemental mercury); >90%, 0.5-2 yrs (readily transported throughout the atmosphere & a global pollutant)
  - GOM (gaseous oxidized mercury, as known as RGM, reactive gaseous mercury); hrs to days
  - PBM (particulate-bound mercury); hrs to days

Mercury in game fish



after Evers et al. 2011

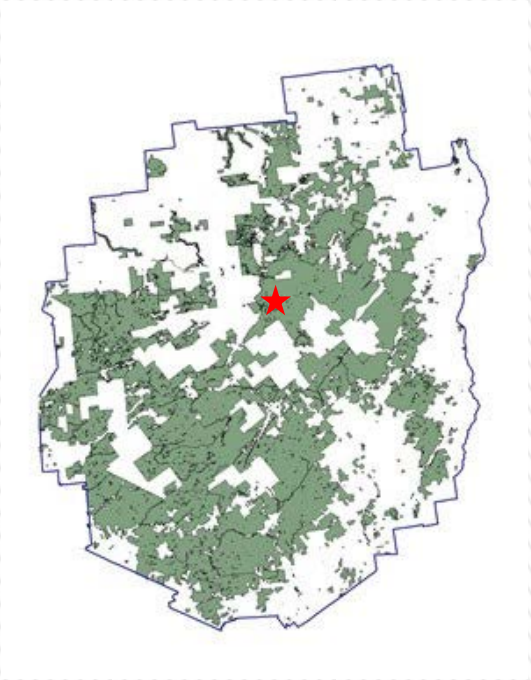
Objective

- Examine the transport and fate of mercury in a northern hardwood forest of the Adirondacks by developing a detailed mercury mass balance.

Site description

Huntington Wildlife Forest (HWF):

- Located in the central Adirondacks of New York State (43.97°N, -74.22°W);
- 6000 ha experimental northern hardwood forest;
- Cool, moist and continental climate;
- Northern hardwood species are dominated by American beech (AB), sugar maple (SM), and yellow birch (YB), with some red maple.



http://www.nrri.umn.edu/worms/team/ongoing_state.html

Data sources

Datasets	Source	Time-interval	Measurement period
Hg fluxes in precipitation	MDN (NY20)	~1 week	2004.1 – 2011.4
Hg concentrations in the atmosphere	Clarkson University	3 hours	2009.1-2011.12
Hg fluxes in throughfall	Choi et al. (2008)	~1 month	2004.1 – 2006.12
Hg concentrations in leaf tissues and litter	Bushey et al. (2008)	~1 month	2004. 5 - 2004.10, 2005. 5 - 2004.10 (fresh leaf); 2004. 5 - 2004. 10, 2005. 10 - 2005.12 (litter)
Hg concentrations in soil water	Syracuse University	~1 month	2004.7 - 2006.6
Hydrological data (stream flow)	SUNY-ESF: Arbutus Lake inlet data	1 day	2004.1 -2006.6
Meteorological data	The Clean Air Status and Trends Network (CASTNET) (HWF 187)	1 hour	2004.1 - 2006.12, 2009.1 – 2011.12

Computation methods

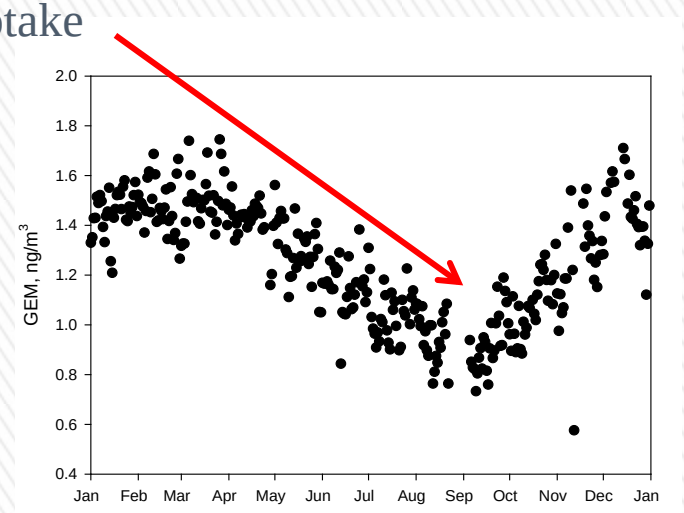
- Wet Hg deposition
  - Weekly data obtained from MDN (Mercury Deposition Network) of NADP
- Dry Hg deposition
  - Dry deposition of GEM, GOM, PBM: Inferential method (measured air Hg conc. × CMAQ deposition velocities)
  - Measured summer GEM depletion
  - Foliar uptake of GEM: foliar mercury accumulation or litterfall mercury – (mercury in soil transpiration)
- Throughfall Hg
  - Monthly data obtained from Choi et al. (2008)
- Litterfall Hg
  - Data obtained from Bushey et al. (2008)
- Soil Hg evasion
  - Choi and Holsen (2009) & datasets of temperature, solar radiation
- Hg in soil water
  - BROOK 90 simulated soil water movement (surface runoff, infiltration, vertical flow, soil transpiration) and measured Hg concentrations in soil solutions at the HWF

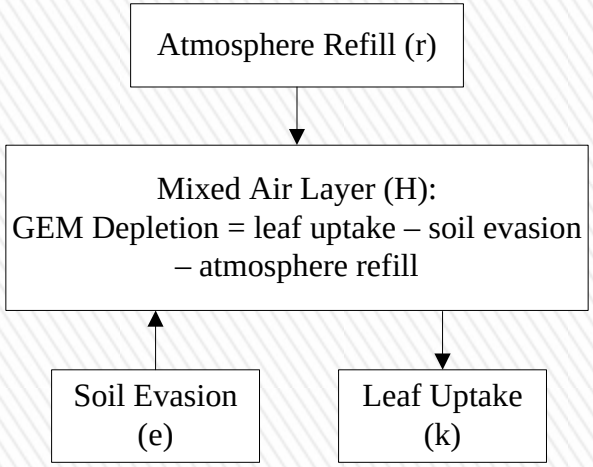
Foliar mercury uptake

Comments on stomatal uptake (via GEM depletion) and soil transpiration:

1. Underestimate of stomatal uptake of GEM via GEM depletion

GEM depletion associated with foliar uptake

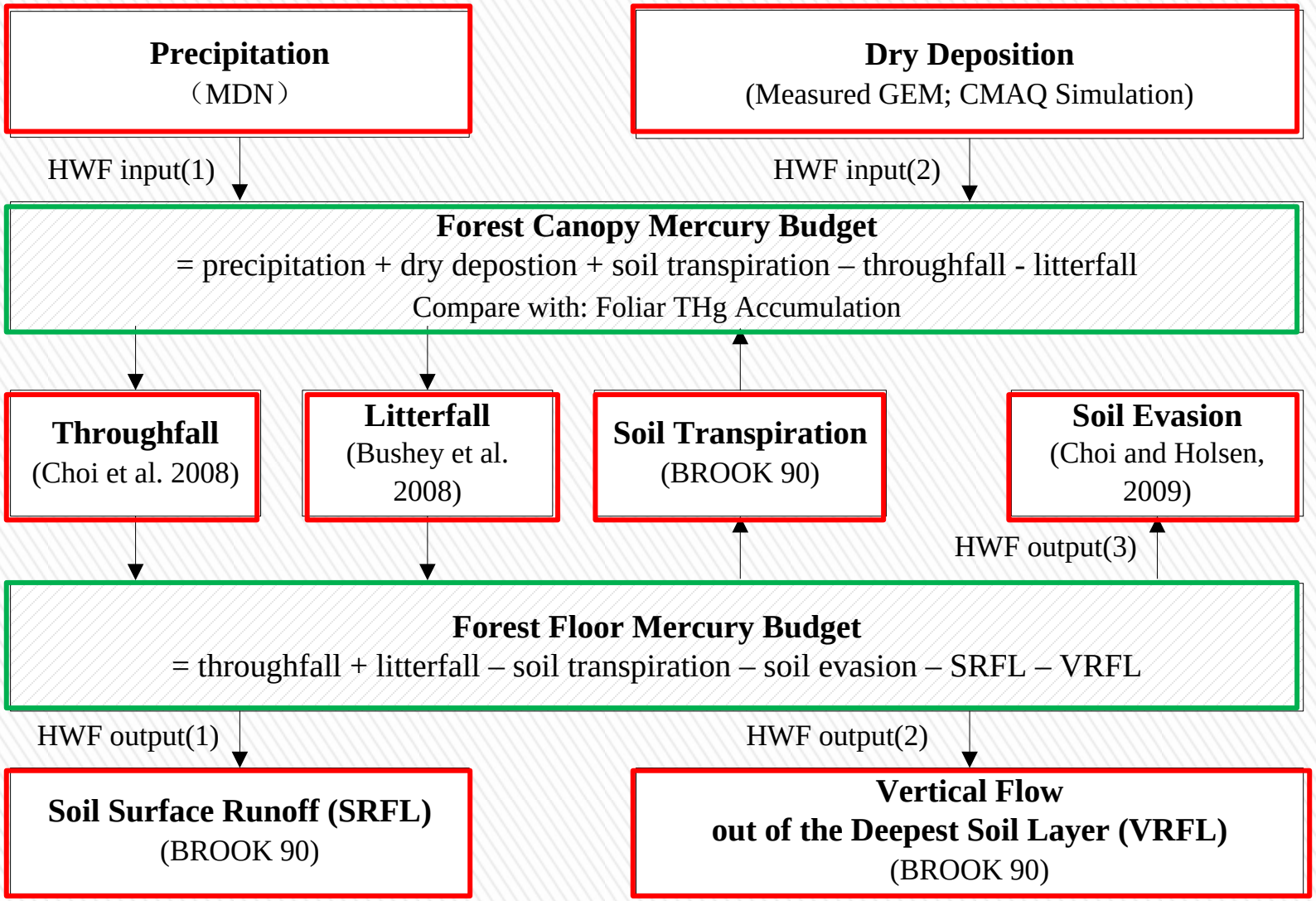




r, H, k were assumed to be constant (unlikely scenario).

2. Overestimate of Hg fluxes in soil transpiration:
assumed 100% uptake of mercury during soil transpiration (unlikely scenario).

Approach


Precipitation
（MDN）
Dry Deposition
(Measured GEM; CMAQ Simulation)
HWF input(1)
HWF input(2)
Forest Canopy Mercury Budget
= precipitation + dry depostion + soil transpiration – throughfall - litterfall
Compare with: Foliar THg Accumulation
Throughfall
(Choi et al. 2008)
Litterfall
(Bushey et al. 2008)
Soil Transpiration
(BROOK 90)
Soil Evasion
(Choi and Holsen, 2009)
HWF output(3)
Forest Floor Mercury Budget
= throughfall + litterfall – soil transpiration – soil evasion – SRFL – VRFL
HWF output(1)
HWF output(2)
Soil Surface Runoff (SRFL)
(BROOK 90)
Vertical Flow
out of the Deepest Soil Layer (VRFL)
(BROOK 90)

Dry mercury deposition (in μg m^{-2} yr^{-1})

	Growing Season		Non-growing Season	
Litterfall – soil transpiration	GEM	15.2	NA	
Foliar accumulation – soil transpiration	GEM	14.7	NA	
GEM depletion	GEM	7.6	NA	
Inferential method (measured atmospheric mercury concentrations × CMAQ deposition velocities)	GEM	10.4	GEM	6.6
	GOM +PBM	~ 0	GOM+PBM	~ 0.2
Throughfall - precipitation	GOM+PBM	1.3	GOM+PBM	0.8

Inferential method:

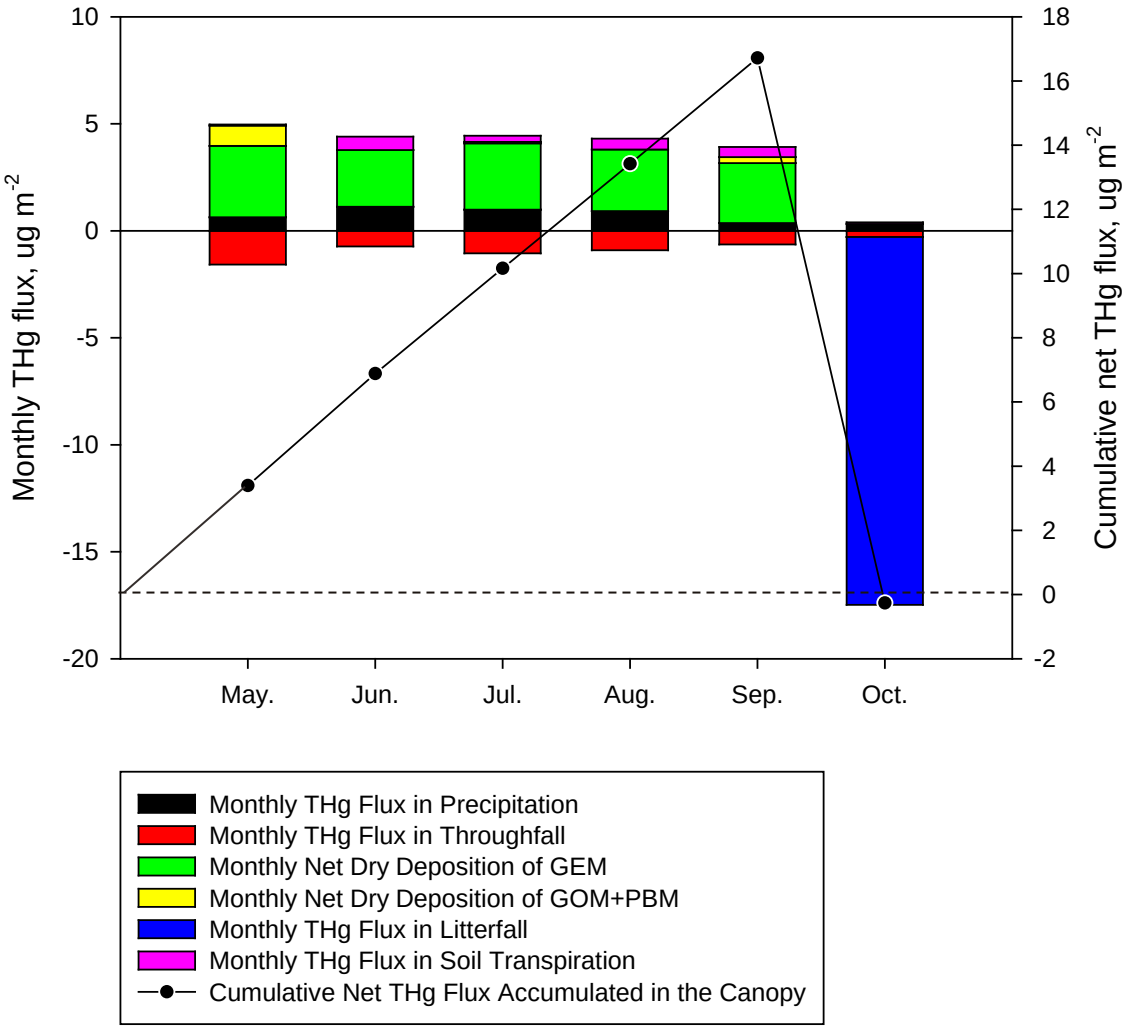
one-directional mercury deposition↓ :

underestimates (deposition velocities by CMAQ & measured air Hg conc.)

The other methods:

net mercury deposition ↑↓

Mercury mass balance for the canopy



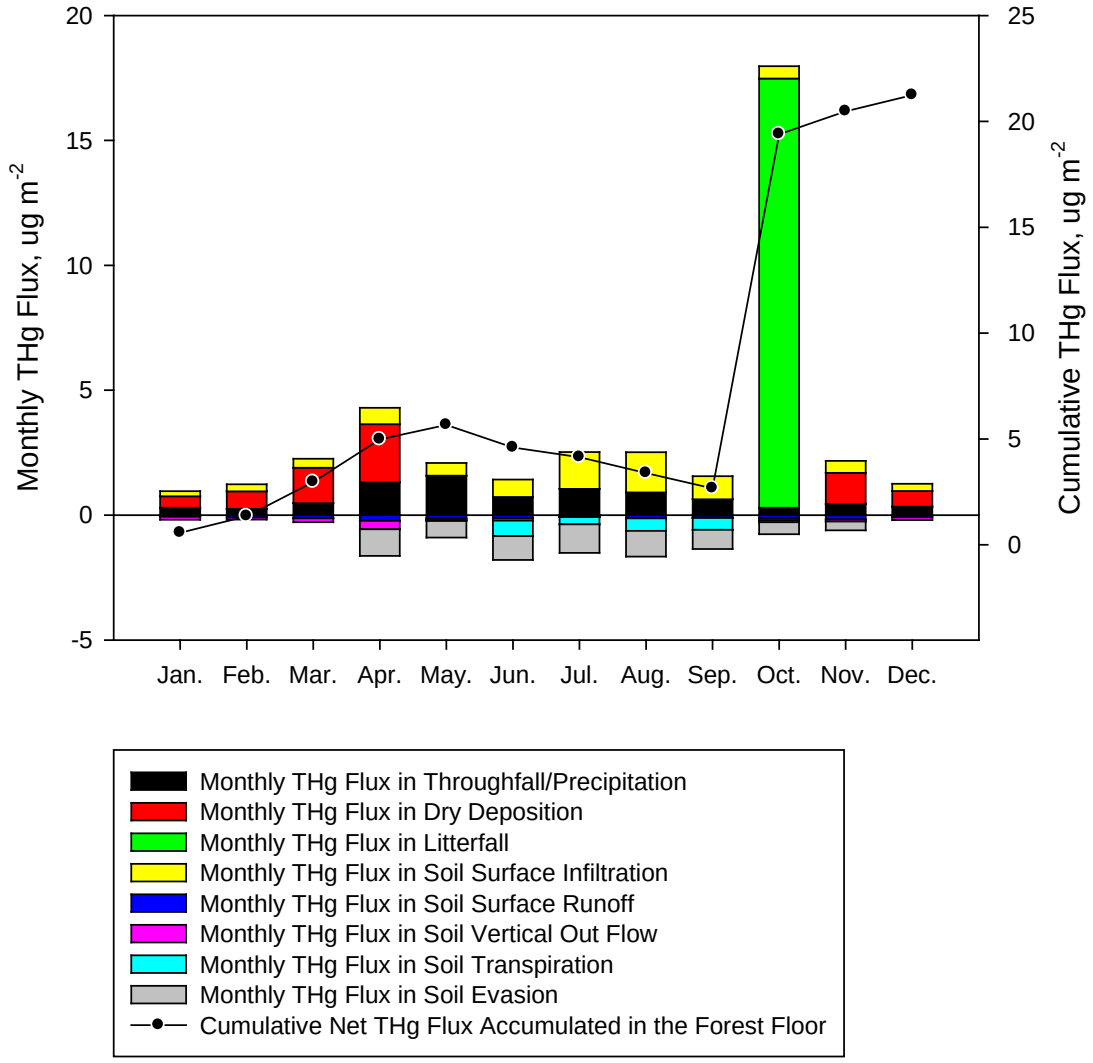
Growing season:

Inputs: 22.1 µg m^{-2} yr^{-1}

Outputs: 22.4 µg m^{-2} yr^{-1}

- GEM uptake by foliage was the major input (14.7 µg m^{-2} yr^{-1}; 67%).
- Hg accumulated in the canopy until litterfall
- Canopy was a temporary sink for Hg during leaf-on period (net accumulation ~17 µg m^{-2} yr^{-1}).

Mercury mass balance for the forest floor



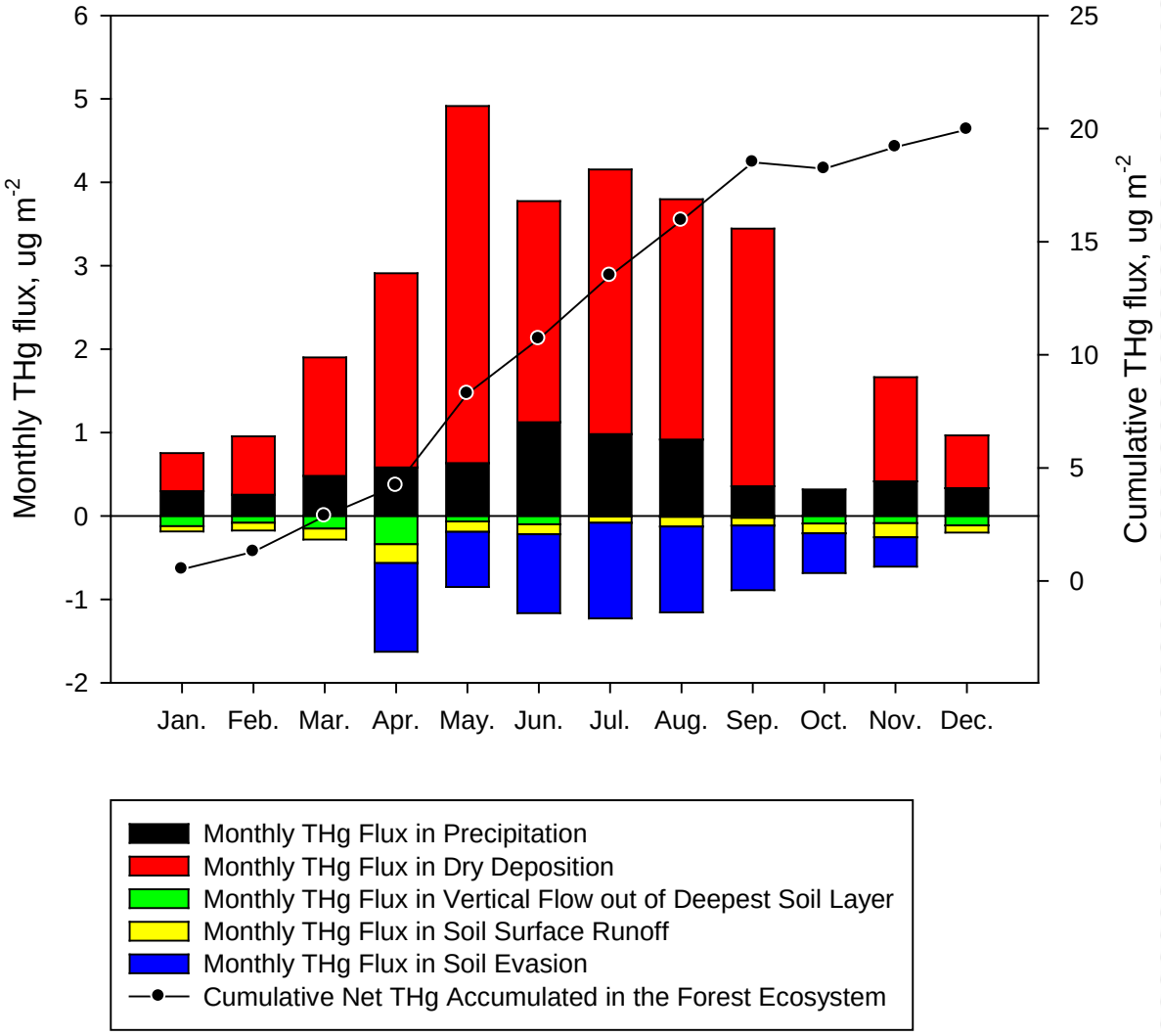
Inputs: 32.2 µg m^{-2} yr^{-1}

Outputs: 11.2 µg m^{-2} yr^{-1}

Net: ~ 21 µg m^{-2} yr^{-1}

- Litterfall: largest input (17.2 µg m^{-2} yr^{-1}; 53%)
- Soil evasion: largest output (6.5 µg m^{-2} yr^{-1}; 57%)
- Soil drainage mercury (surface runoff + vertical out flow) was a small export pathway (8.6% of total mercury inputs), especially for the growing season.

Mercury mass balance for the overall ecosystem



Inputs: 29.2 µg m^{-2} yr^{-1}

Outputs: 9.2 µg m^{-2} yr^{-1}

Net: 20 µg m^{-2} yr^{-1}

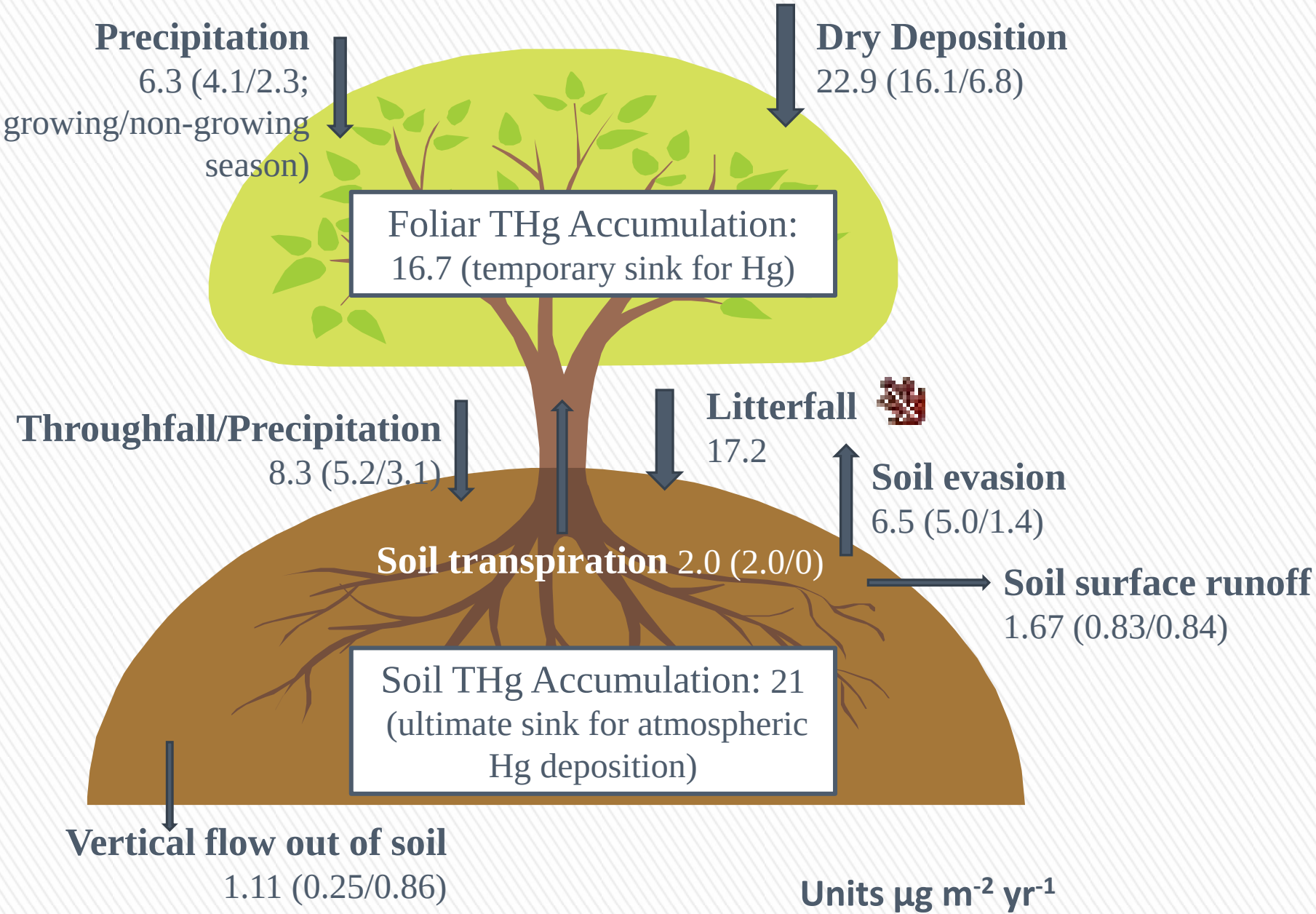
Inputs:

Dry deposition (22.9 µg m^{-2} yr^{-1}) > wet deposition (6.3 µg m^{-2} yr^{-1})

Outputs:

Soil evasion (6.5 µg m^{-2} yr^{-1}) > soil drainage (2.8 µg m^{-2} yr^{-1})

The northern forest mediates mercury transport



Conclusions

- Foliar GEM uptake was the major Hg input (67%) to the HWF.
- The forest canopy was a temporary sink for Hg over the growing season.
- Most (68%) of the atmospheric Hg deposition to the HWF was retained in the ecosystem; the losses of Hg were from predominantly from soil evasion (22%) with a small fraction (10%) lost via watershed drainage.
- The HWF was a substantial net sink for atmospheric Hg as well as an important Hg filter between atmosphere and hydrosphere.

Foliar mercury uptake

Comparison of foliar mercury uptake at the HWF

Direct estimate (accumulation rate of foliar Hg concentration × leaf mass per ground area)	16.7 μg m^{-2} yr^{-1}
Litterfall mercury deposition (litter Hg concentration × litter mass per ground area)	17.2 μg m^{-2} yr^{-1}
GEM depletion (stomatal uptake of GEM)+ **Hg uptake via soil transpiration**	9.6 μg m^{-2} yr^{-1} (stomatal uptake of GEM: 7.6; Hg in soil transpiration: 2.0)

Foliar mercury uptake

Uncertainties of the estimate methods

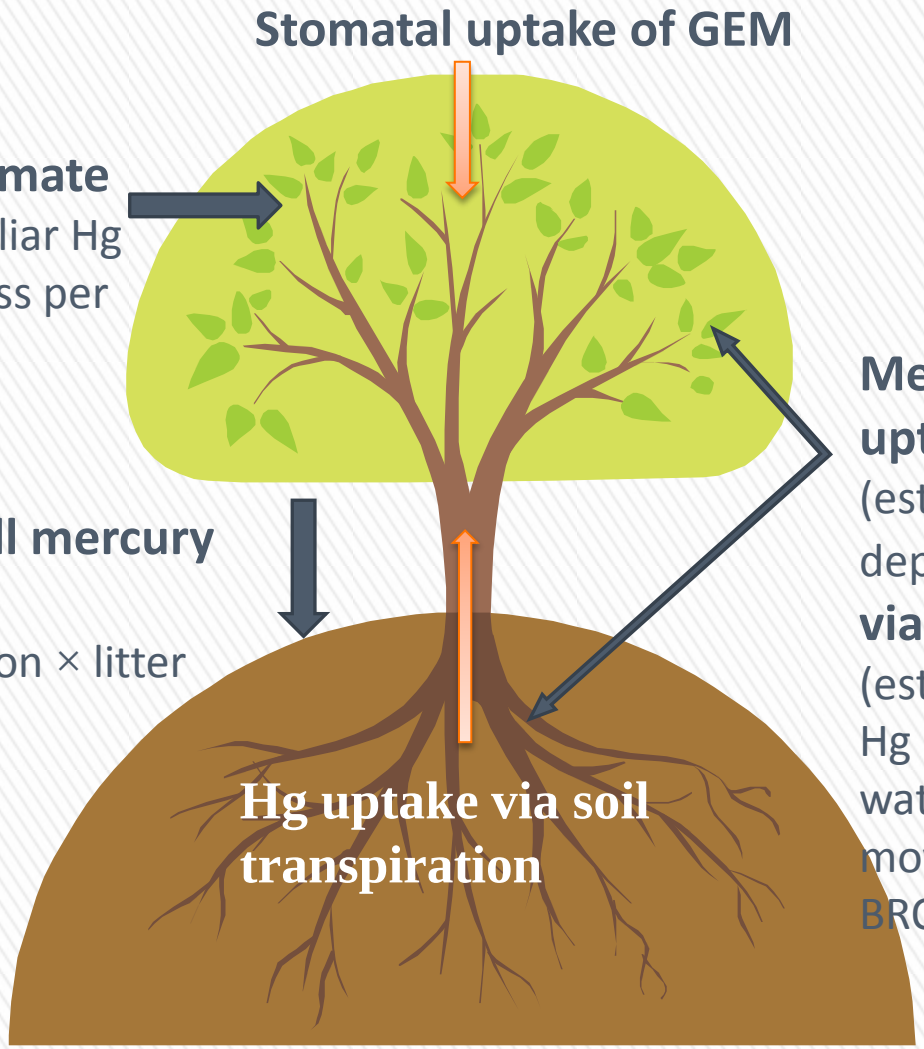
Direct estimation of foliar mercury & litterfall mercury:

- limited knowledge of leaf biomass of major tree species

Investigator/Source	YB	SM	AB	Overall Forest	Basis
Smith and Martin (2001), IFS (1992)	35.5	131.8	86.7	248.0	canopy biomass
Blair (unpublished data)	NA	NA	NA	341.4	
IFS (1992)	NA	NA	NA	354.4	
Bushey et al. (2008)	3.8	209	145.5	358.3	litter biomass
IFS (1992)	NA	NA	NA	313.1	
Blair (unpublished data)	NA	NA	NA	348.0	

unit: g/m^2

Foliar mercury uptake



Method 1: **Direct estimate** (accumulation rate of foliar Hg concentration × leaf mass per land area)

Method 2: **Litterfall mercury deposition** (litter Hg concentration × litter mass per land area)

Method 3: **Stomatal uptake of GEM** (estimated by GEM depletion) **+ Hg uptake via soil transpiration** (estimated by measured Hg concentrations in soil water and soil water movement simulated by BROOK90)