Quantifying mercury in forest biomass

Theme 2:

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- Concentrations of Hg in wood are low but the massive amount of wood in forests makes this pool important for forest Hg cycling.
- Hg in wood is declining with time, likely due to reduced foliar uptake from a cleaner atmosphere.
- Future climate changes are likely to release more Hg from forest soils

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http://www.nsrcforest.org

Project Summary

The purpose of this project was to advance the understanding of Hg cycling in forests. Few studies had reported Hg in wood because concentrations were too low to be detected by commonly used methods. Thus our first goal was to establish a reliable method for analyzing Hg in wood. We compared different sample preparation methods and found that wood samples should be either freeze-dried or oven-dried at 65°C and then loaded in large quantities for analytical analysis due to their low concentrations. We found that oven drying at higher temperatures resulted in loss of Hg, while air-drying resulted in accumulation of Hg from the environment. Using these analytical methods, our next goal was to quantify pools of Hg in wood in the dominant species at four sites in the Northern Forest: Hubbard Brook, NH; Bear Brook, ME; Huntington Forest, NY; and Sleepers River, VT. Although concentrations of Hg in wood are low, the wood is the most massive part of a tree. Thus woody tissues can have an equal or larger Hg content than foliage. We were also interested in identifying the source of Hg in tree wood. Ten sugar maple clones planted in six blocks at the Heiberg Forest in New York State showed significant genetic control of sap Hg concentration, which was not related to soil Hg concentration differences across blocks. Clones could differ in stomatal uptake, root uptake, or translocation of Hg. We analyzed Hg in tree rings of four species collected at Hubbard Brook. Declining concentrations from the top to the bottom of the bole and from older to newer tree rings suggest that foliar uptake of Hg is more important than root uptake. Finally, to understand the climate change impacts on Hg cycling, we measured different pathways of Hg accumulation and loss in forests using climate-change manipulation experiments at the Hubbard Brook Experimental Forest, New Hampshire, USA: a combined growing-season warming and winter freeze-thaw cycle experiment, a throughfall exclusion to mimic drought, and a simulated ice storm experiment. We found that warmer soils in the growing season or more intense ice storms in winter may exacerbate Hg pollution by releasing Hg sequestered in forest soils via evasion and leaching. Understanding the source and fate of Hg in tree wood is important for predicting changes in Hg cycling in forests.

Background and Justification

Terrestrial ecosystems receive significant inputs of broadly toxic mercury (Hg) from atmospheric deposition of direct natural and human emissions as well as re-emissions (Grigal 2003; Driscoll et al. 2013). Trees are important to Hg cycling in forests, although their tissue concentrations are low (Obrist et al. 2011, 2012), because they are so massive that they contain significant amounts of Hg (Blackwell et al. 2014; Yang et al. 2018). Understanding the content of Hg in tree tissues would help predict the re-emission of Hg associated with losses of aboveground carbon pools (Obrist 2007); biomass burning is a potentially important but poorly characterized source of Hg emissions (Friedli et al. 2009). The pathways of Hg uptake and translocation in bole wood are uncertain and difficult to study, especially in field settings.

Litterfall and throughfall (Hg washed from foliar surface during rain events) have been shown to dominate the input of Hg to forest soils (Grigal et al. 2000; St. Louis et al. 2001; Demers et al. 2007; Bushey et al. 2008). Forest soils sequester Hg but can re-emit Hg (Hg^0) back to the atmosphere (Graydon et al. 2009; Denkenberger et al. 2012). Forest soils can also release Hg^{2+} to receiving waters via drainage waters (Driscoll et al. 2007). The dissolved Hg leached from soils to nearby streams and lakes can be methylated and bioaccumulate up food chains, resulting in exposure to wildlife or humans (Chan et al. 2003; Chen et al. 2008). However, all these fluxes can be altered under warmed temperatures, droughts, or ice storm events. Measurements are needed to quantify the magnitude of changes in soil Hg retention due to expected changes in climate for forest ecosystems (Obrist et al. 2018).

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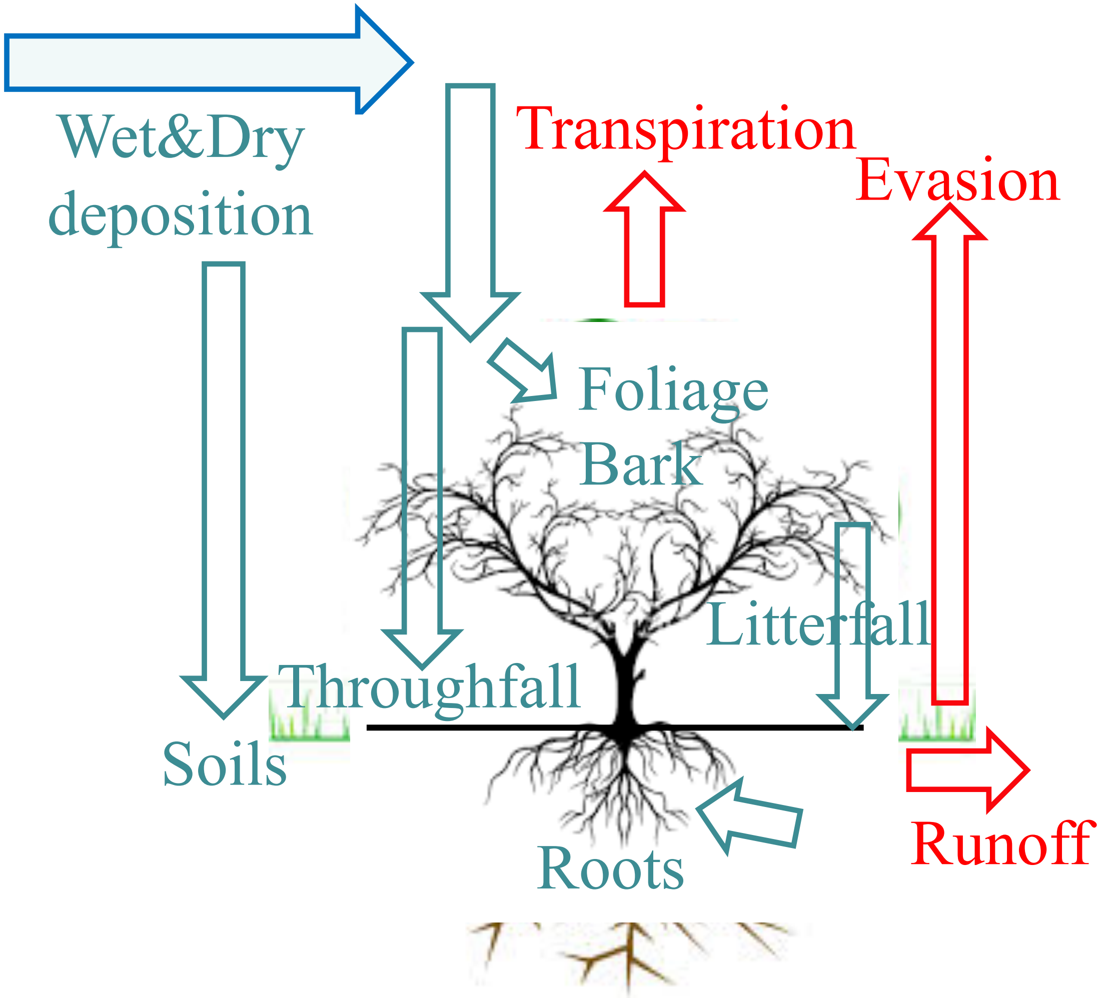
Sources

Elemental Hg
reactive gaseous Hg
particulate Hg



Wet&Dry deposition

Transpiration

Evasion

Foliage

Bark

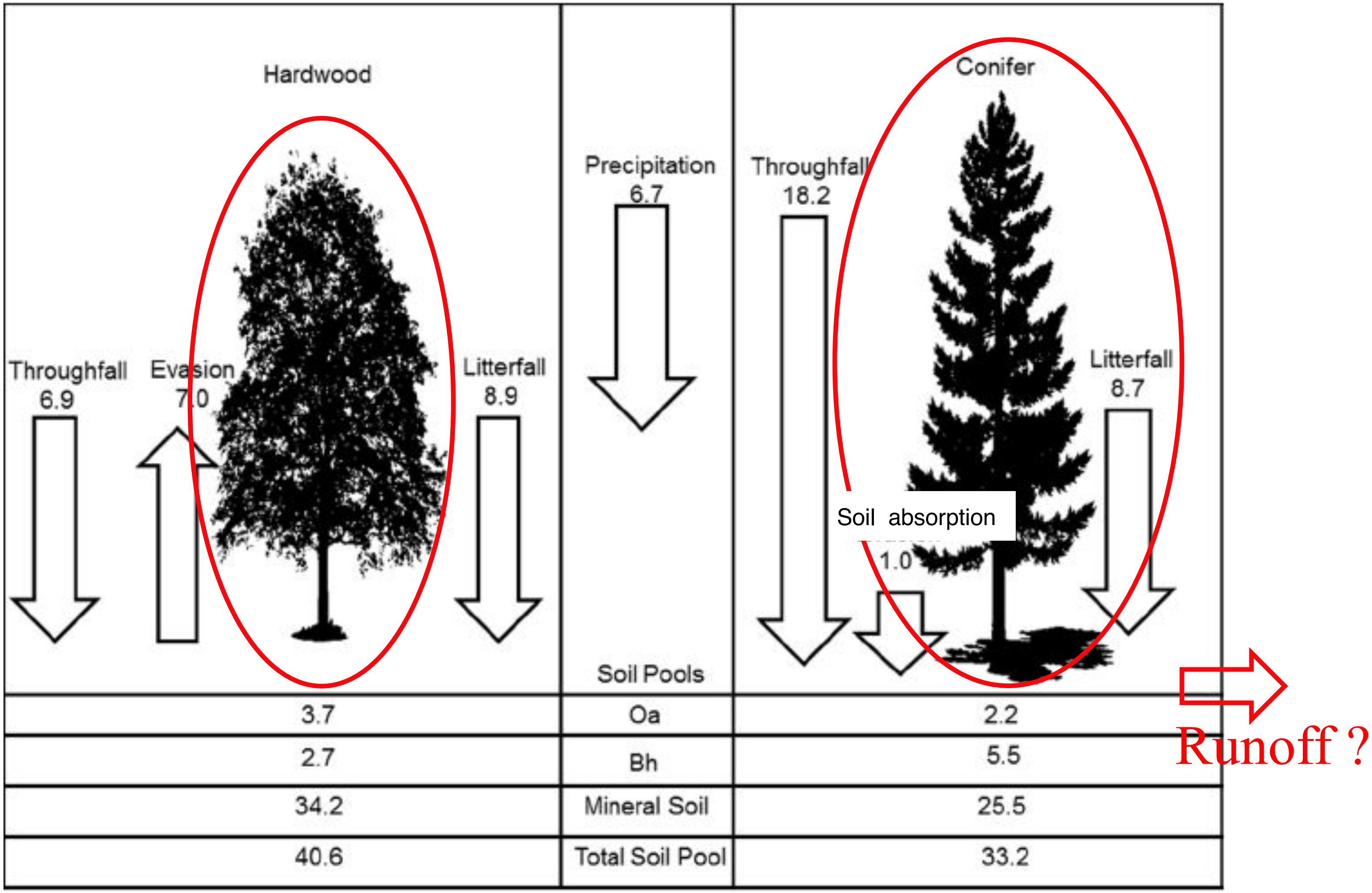
Litterfall

Throughfall

Soils

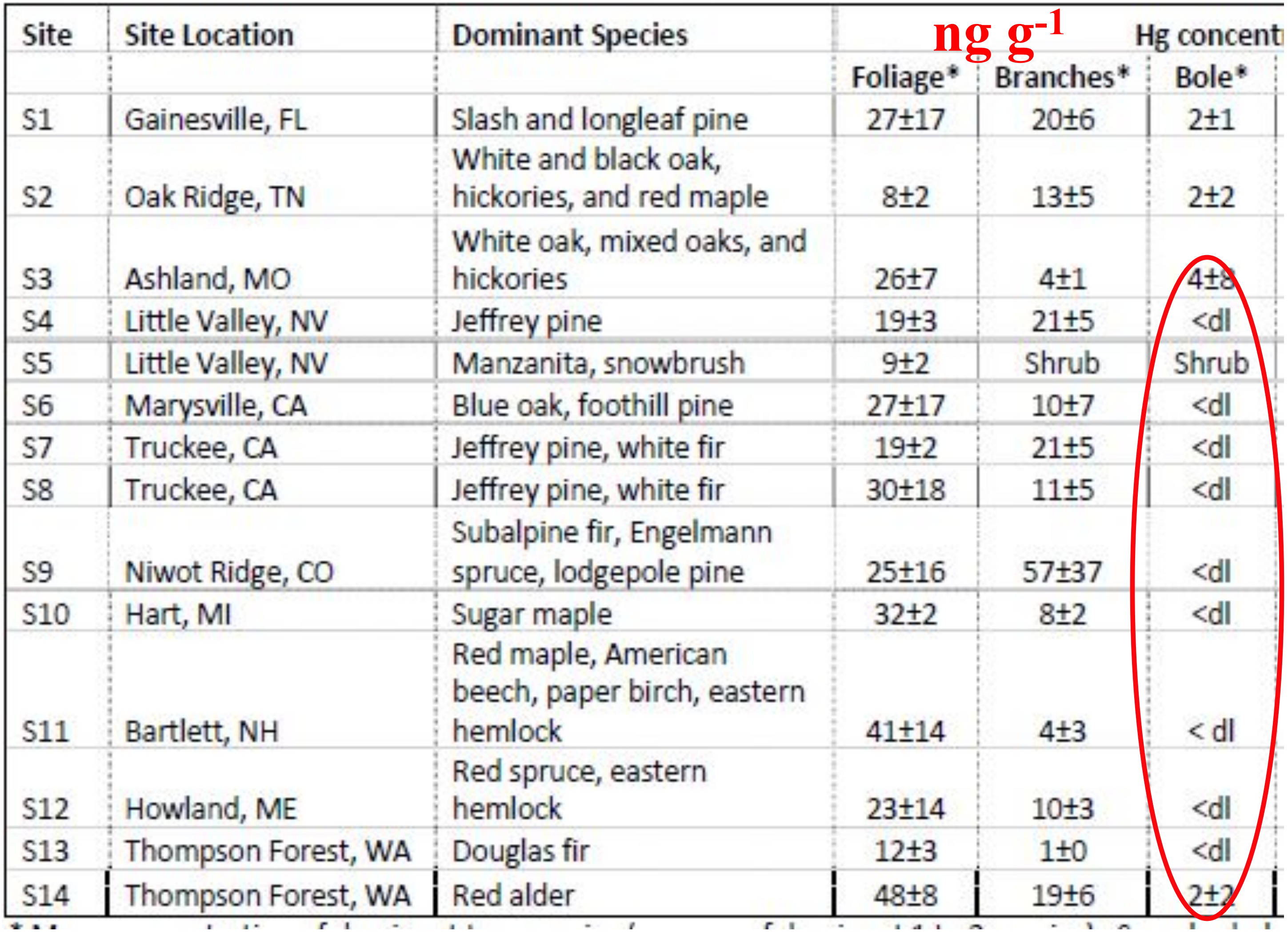
Roots

Runoff



Hardwood	Soil Pools	Conifer
3.7	Oa	2.2
2.7	Bh	5.5
34.2	Mineral Soil	25.5
40.6	Total Soil Pool	33.2

Adapted from Blackwell et al., 2014



ng g^{-1}

Site	Site Location	Dominant Species	Hg concent		
			Foliage*	Branches*	Bole*
S1	Gainesville, FL	Slash and longleaf pine	27±17	20±6	2±1
S2	Oak Ridge, TN	White and black oak, hickories, and red maple	8±2	13±5	2±2
S3	Ashland, MO	White oak, mixed oaks, and hickories	26±7	4±1	4±8
S4	Little Valley, NV	Jeffrey pine	19±3	21±5	<dl
S5	Little Valley, NV	Manzanita, snowbrush	9±2	Shrub	Shrub
S6	Marysville, CA	Blue oak, foothill pine	27±17	10±7	<dl
S7	Truckee, CA	Jeffrey pine, white fir	19±2	21±5	<dl
S8	Truckee, CA	Jeffrey pine, white fir	30±18	11±5	<dl
S9	Niwot Ridge, CO	Subalpine fir, Engelmann spruce, lodgepole pine	25±16	57±37	<dl
S10	Hart, MI	Sugar maple	32±2	8±2	<dl
S11	Bartlett, NH	Red maple, American beech, paper birch, eastern hemlock	41±14	4±3	< dl
S12	Howland, ME	Red spruce, eastern hemlock	23±14	10±3	<dl
S13	Thompson Forest, WA	Douglas fir	12±3	1±0	<dl
S14	Thompson Forest, WA	Red alder	48±8	19±6	2±2

Obrist et al., 2011

Methods

Project 1. We collected bole wood samples from the dominant species, American beech (*Fagus grandifolia* Ehrh.), sugar maple (*Acer saccharum* Marshall.), red spruce (*Picea rubens* Sarg.) and balsam fir (*Abies balsamea* (L.) Mill.), at the Hubbard Brook Experimental Forest. We prepared saw-dust size samples in five different processing methods: fresh, air-drying, freeze-drying and oven-drying at 65 and 103°C. For air-drying samples, we further examined the possibility of contamination by putting samples in each of eight locations: a clean room, a drying room, two dirty rooms, a garage, a barn, a storage room, and a closed chamber with liquid Hg. We then analyzed Hg concentrations using the EPA combustion method.

Project 2.

We collected bark, bole wood and foliage samples from dominant hardwood and conifer species at four forested sites in the northeastern USA. Dominant trees included American beech, white ash (*Fraxinus americana* L.), yellow birch (*Betula alleghaniensis* Britt.), sugar maple, red maple (*Acer rubrum* L.), red spruce, balsam fir and white pine (*Pinus strobus* L.). We analyzed samples using combustion methods for Hg concentrations, and then calculated Hg pools using the measured concentrations times the estimated biomass.

Project 3.

We collected wood disc samples from the bottom, middle, and top of trees of American beech, sugar maple, yellow birch, and red spruce at the Hubbard Brook Experimental Forest. We analyzed each 5-year growth increment using bottom disc samples to examine radial patterns of Hg. We analyzed the most recent 5-year growth increment for disc samples collected at three heights to examine vertical patterns of Hg.

We collected xylem sap samples using a mini-tap procedure from sugar maple trees that varied by clone and location (block) at the Heiberg Forest. Clones had been propagated by selecting current year shoots in the spring of 1971, 1972, and 1973 just after elongation. Cuttings were placed in media under a mist system to establish roots. Following root establishment, young cuttings were potted and overwintered in a cold frame. Clonal trees were planted at the Heiberg Forest in 1973 and 1974 at a spacing of 3–4 m, interspersed with wild type sugar maples grown from seed. Clonal trees were distributed among six adjoining replicate blocks in a randomized complete block design. We also collected foliage and soil samples selectively for Hg analysis.

Project 4.

We conducted measurements at experimental plots associated with three climate change manipulation experiments located at the Hubbard Brook Experimental Forest in the White Mountain National Forest in Central New Hampshire, USA. To examine the effects of soil warming in the growing season and soil freeze-thaw cycles in winter, three plots were selected (each 11 m × 13.5 m): one control plot, one plot with soils warmed by ~ 5 °C via heating cables during the growing season (warming), and one plot warmed in the growing season and also subjected to four 3-day freezing episodes induced by removing snow by shoveling, separated by 3 days of soil warming (warming + freeze-thaw cycles). These treatments had been applied for 4 years (since December 2013) at the time of our study. To examine the effect of drought, one plot was selected from the drought experiment (15 m × 15 m) in which ~ 50% of throughfall was removed by placing gutters 2 m above the ground to cover 50% of the surface area in spring 2015. To examine the effect of ice storm events, we selected two plots (each 20 m × 30 m, 10 m apart) from the simulated ice storm experiment: one control plot and one high-ice plot, which received 0.75 inches of glaze ice as one event in February 2016, one and a half years prior to our study. We measured soil Hg^0 evasion rates in three seasons and collected leaf litterfall, throughfall, soil, and soil solution for Hg analysis in above plots.

Processing in the laboratory

Clean samples

Analytical methods for analyzing mercury

Method	Method detection limit (ng g^{-1})	Sample size (mg)	Preparation procedure	EPA method
AFS (Atomic fluorescence spectroscopy)	0.0005	> 500	Digestion	1631E
ICP-MS (Inductively coupled plasma mass spectrometry)	0.1	> 500	Acid digestion	6020A
Manual cold vapor atomic absorption spectrometry	0.2	500 – 600	Heating or digestion	7471B
Thermal decomposition, amalgamation, and atomic absorption spectrophotometry	1.0	2 - 1000	None	7473
Microwave digestion	1.75	> 500	Acid digestion	3051A
ICP-AES (Inductively coupled plasma atomic emission spectroscopy)	17	> 500	Acid digestion	6010B

Results/Project outcomes

Project 1. Samples that were freeze-dried or oven-dried at 65°C were suitable for determination of Hg, whereas oven-drying at 103°C resulted in Hg losses, and air-drying resulted in Hg gains (Figure 1), presumably due to sorption from indoor air. Mean (±SE) concentrations of Hg tree bole wood were 1.75 ± 0.14 ng g^{-1} for American beech, 1.48 ± 0.23 ng g^{-1} for sugar maple, 3.96 ± 0.19 ng g^{-1} for red spruce and 4.59 ± 0.06 ng g^{-1} for balsam fir. Based on these concentrations and estimates of wood biomass by species based on stand inventory, we estimated the Hg content of wood in the reference watershed at Hubbard Brook to be 0.32 g ha^{-1}, twice the size of the foliar Hg pool (0.15 g ha^{-1}).

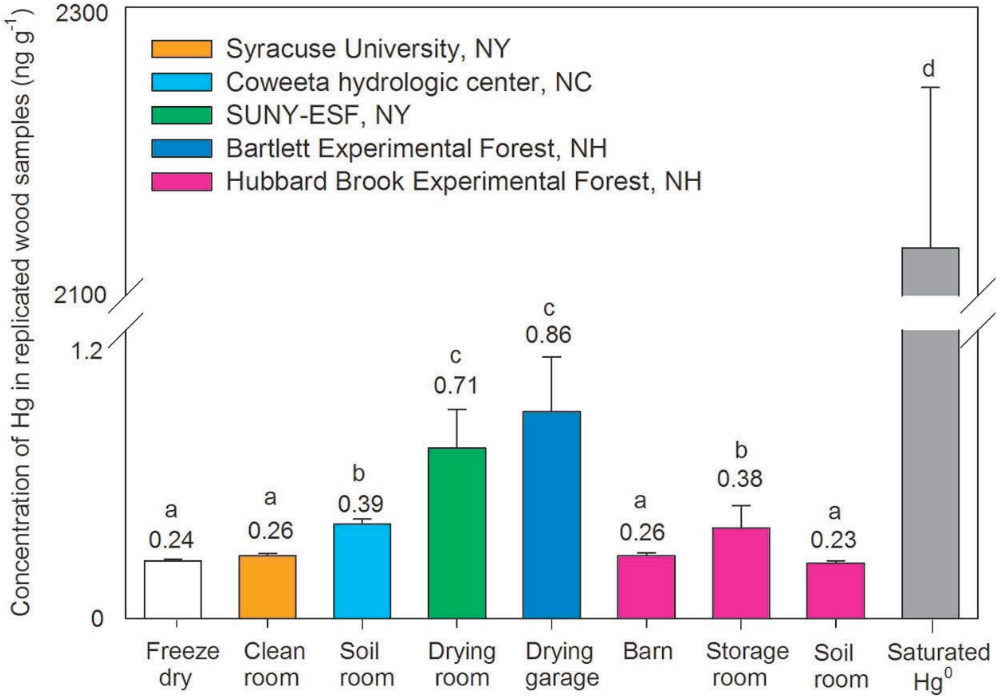
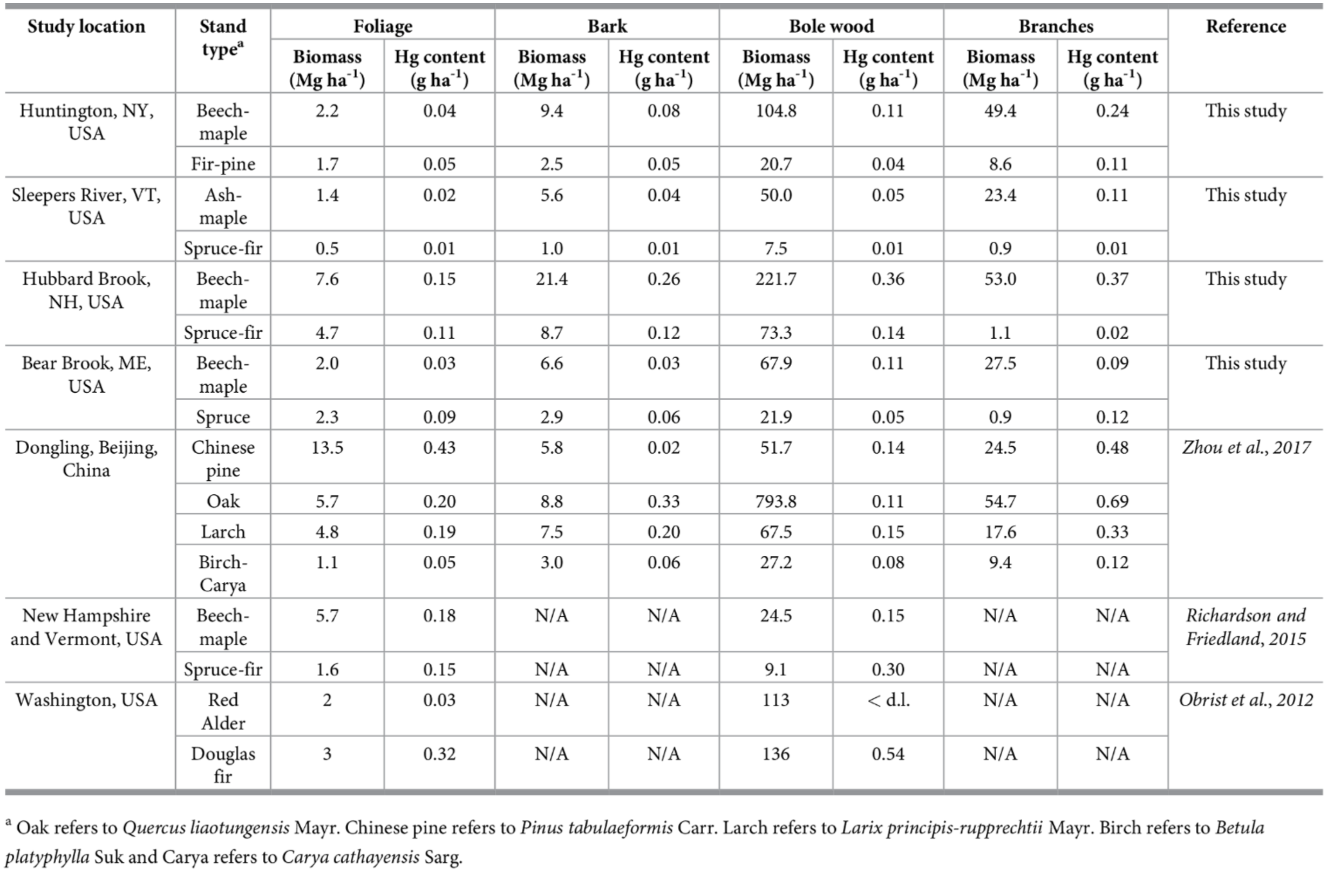


Figure 1. Mercury in sugar maple wood air-dried in different locations. For comparison, samples were prepared without any contamination (freeze-dried) and fully contaminated (in air exposed to liquid Hg). Different letters indicate significant differences (Tukey's honestly significant difference test).

Project 2. Foliar concentrations of Hg averaged 16.3 ng g^{-1} among the hardwood species, which was significantly lower than values in conifers, which averaged 28.6 ng g^{-1} ($p < 0.001$). Similarly, bark concentrations of Hg were lower ($p < 0.001$) in hardwoods (7.7 ng g^{-1}) than conifers (22.5 ng g^{-1}). For wood, concentrations of Hg were higher in yellow birch (2.1–2.8 ng g^{-1}) and white pine (2.3 ng g^{-1}) than in the other species, which averaged 1.4 ng g^{-1} ($p < 0.0001$). Sites differed significantly in Hg concentrations of foliage and bark ($p = 0.02$), which are directly exposed to the atmosphere, but the concentration of Hg in wood depended more on species ($p < 0.001$) than site ($p = 0.60$). The Hg contents of tree tissues in hardwood stands, estimated from modeled biomass and measured concentrations at each site, were higher in bark (mean of 0.10 g ha^{-1}) and wood (0.16 g ha^{-1}) than in foliage (0.06 g ha^{-1}, Table 1). In conifer stands, because foliar concentrations were higher, the foliar pool tended to be more important.



Study location	Stand type[a]	Foliage		Bark		Bole wood		Branches		Reference
		Biomass (Mg ha^{-1})	Hg content (g ha^{-1})	Biomass (Mg ha^{-1})	Hg content (g ha^{-1})	Biomass (Mg ha^{-1})	Hg content (g ha^{-1})	Biomass (Mg ha^{-1})	Hg content (g ha^{-1})	
Huntington, NY, USA	Beech-maple	2.2	0.04	9.4	0.08	104.8	0.11	49.4	0.24	This study
	Fir-pine	1.7	0.05	2.5	0.05	20.7	0.04	8.6	0.11	
Sleepers River, VT, USA	Ash-maple	1.4	0.02	5.6	0.04	50.0	0.05	23.4	0.11	This study
	Spruce-fir	0.5	0.01	1.0	0.01	7.5	0.01	0.9	0.01	
Hubbard Brook, NH, USA	Beech-maple	7.6	0.15	21.4	0.26	221.7	0.36	53.0	0.37	This study
	Spruce-fir	4.7	0.11	8.7	0.12	73.3	0.14	1.1	0.02	
Bear Brook, ME, USA	Beech-maple	2.0	0.03	6.6	0.03	67.9	0.11	27.5	0.09	This study
	Spruce	2.3	0.09	2.9	0.06	21.9	0.05	0.9	0.12	
Dongling, Beijing, China	Chinese pine	13.5	0.43	5.8	0.02	51.7	0.14	24.5	0.48	*Zhou et al., 2017*
	Oak	5.7	0.20	8.8	0.33	793.8	0.11	54.7	0.69	
	Larch	4.8	0.19	7.5	0.20	67.5	0.15	17.6	0.33	
	Birch-Carya	1.1	0.05	3.0	0.06	27.2	0.08	9.4	0.12	
New Hampshire and Vermont, USA	Beech-maple	5.7	0.18	N/A	N/A	24.5	0.15	N/A	N/A	*Richardson and Friedland, 2015*
	Spruce-fir	1.6	0.15	N/A	N/A	9.1	0.30	N/A	N/A	
Washington, USA	Red Alder	2	0.03	N/A	N/A	113	< d.l.	N/A	N/A	*Obrist et al., 2012*
	Douglas fir	3	0.32	N/A	N/A	136	0.54	N/A	N/A	

[a] Oak refers to *Quercus liaotungensis* Mayr. Chinese pine refers to *Pinus tabulaeformis* Carr. Larch refers to *Larix principis-rupprechtii* Mayr. Birch refers to *Betula platyphylla* Suk and Carya refers to *Carya cathayensis* Sarg.

Table 1. Biomass and Hg content of foliage, bark and bole wood in hardwood and conifer stands in this study and three published studies

Project 3. Declining concentrations from the top to the bottom of the bole ($p < 0.001$) and from older to newer tree rings ($p = 0.001$; Figure 2) suggest that foliar uptake of Hg is more important than root uptake. Ten sugar maple clones planted in six blocks at the Heiberg Forest in New York State showed significant genetic control of sap Hg concentration ($p = 0.02$; Figure 3), which was not related to soil Hg concentration differences across blocks.

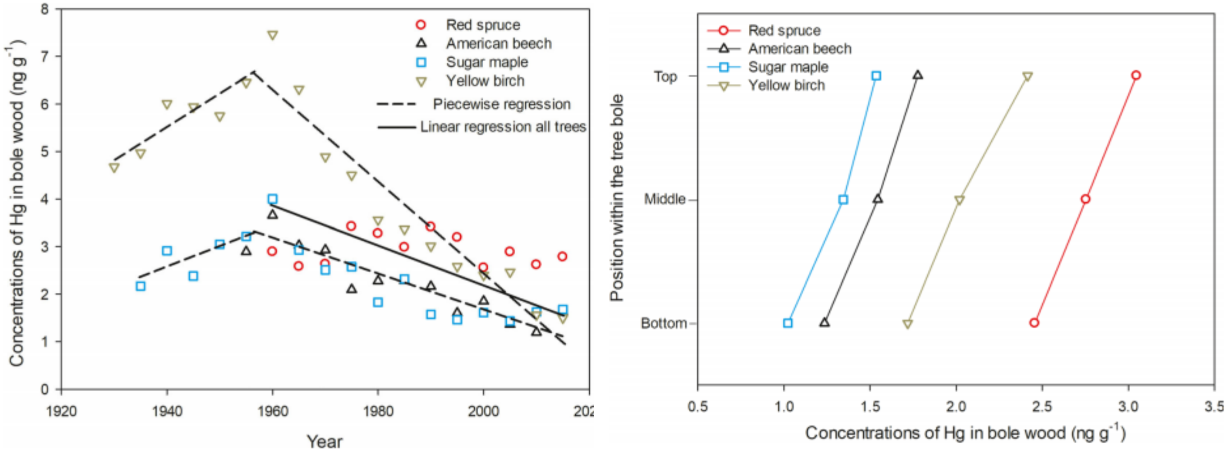


Figure 2. Radial (left) and vertical (right) pattern of Hg concentrations in bole wood of four trees sampled at the Hubbard Brook Experimental Forest in New Hampshire. Samples were taken in 5-year increments of growth rings.

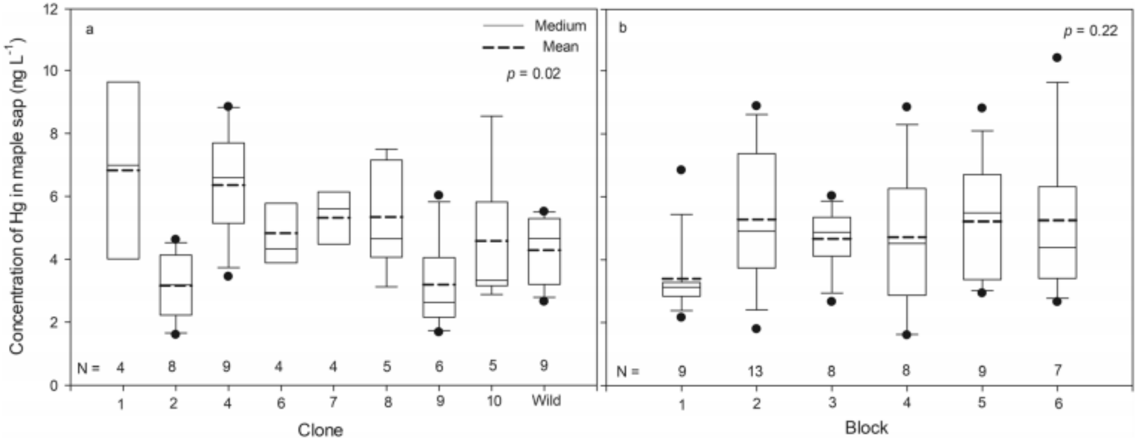


Figure 3. Concentrations of Hg in maple sap by clone (a) and block (b). Aligned rank testing found significant effects of clone ($p = 0.02$) but not block ($p = 0.22$).

Project 4. Comparing Hg inputs to the soil (litterfall and throughfall) with Hg outputs (soil evasion), soil Hg retention was 16–60% lower at the soil warming (6.4 μg m^{-2} year^{-1}), soil warming + freeze-thaw (3.4 μg m^{-2} year^{-1}), and drought plots (6.8 μg m^{-2} year^{-1}) than in the control (8.1 μg m^{-2} year^{-1}; Figure 5). In the simulated ice storm plot, where we estimated Hg in woody materials as an additional input and Hg leaching as an additional output, soil Hg retention was 41% lower (3.9 μg m^{-2} year^{-1}) than in the control (6.6 μg m^{-2} year^{-1}).

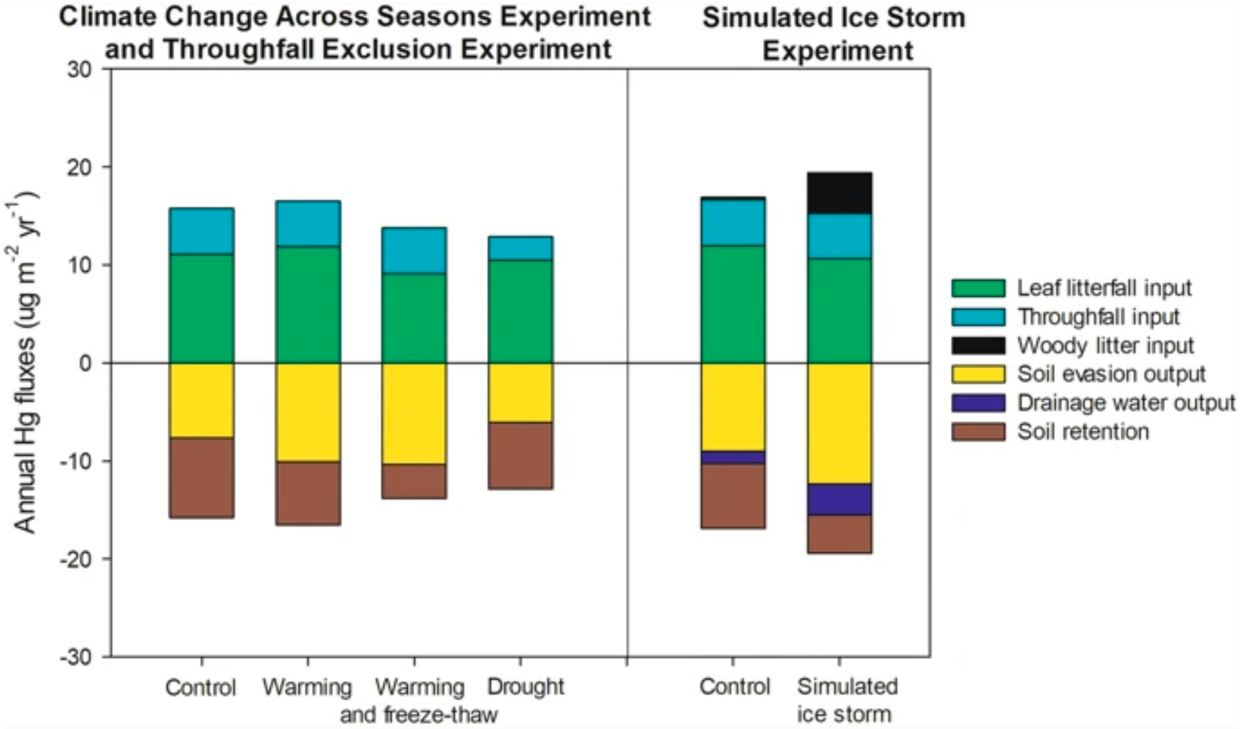
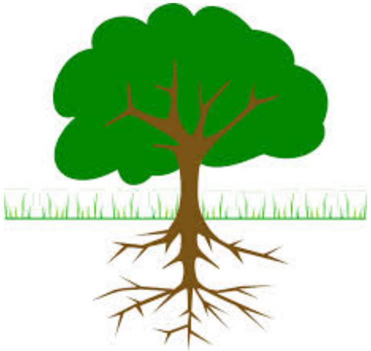


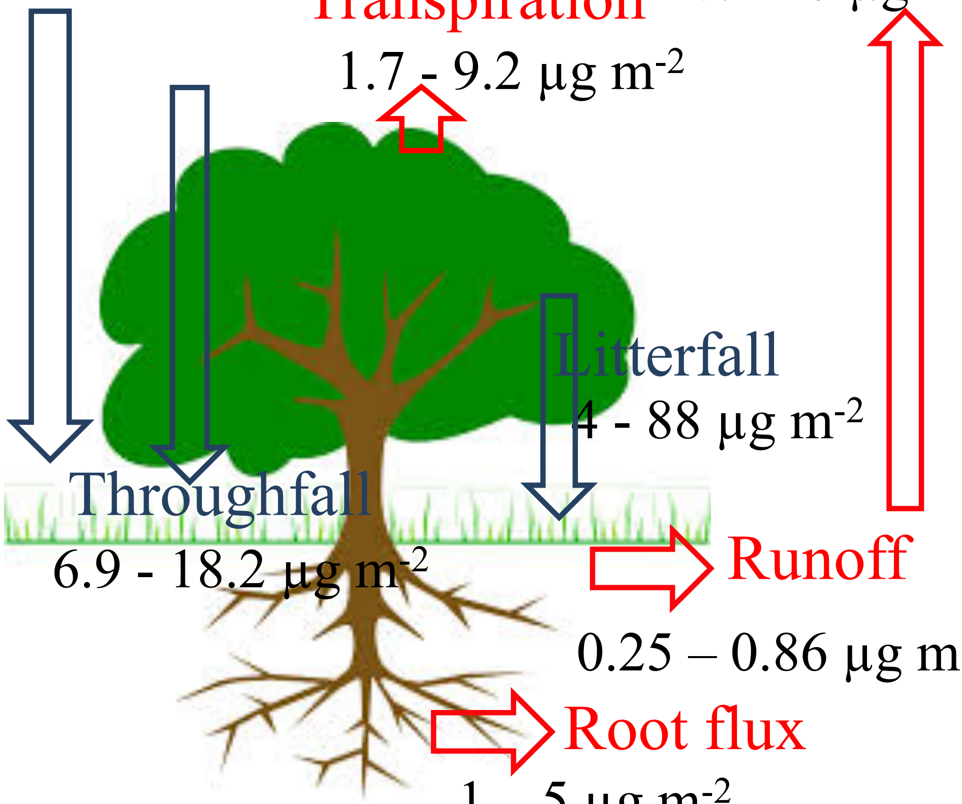
Figure 5. Measured annual Hg inputs (litterfall and throughfall) and outputs (soil evasion and drainage water) for experiments depicting three climate change manipulations in northern hardwood at the Hubbard Brook Experimental Forest, NH. Soil Hg retention was calculated by difference. Input of Hg in twigs and branches and output of Hg via soil water discharge were measured only in the simulated ice storm plots; soil Hg retention was overestimated in the other plots by the amount of Hg in the drainage water and underestimated by the amount of Hg in woody litter.

Implications and applications in the Northern Forest region

Mercury in wood deserves more attention and is feasible to measure using appropriate techniques. Better understanding of the source of Hg in wood is needed to forecast future changes in Hg cycling in forested ecosystems. Climate changes such as warmer air temperatures and intense ice storms are likely to exacerbate Hg pollution by releasing Hg sequestered in forest soils. Monitoring efforts should be focused on areas with high forest cover that are most sensitive to climate change.



Soils	9,000-22,000 µg m^{-2}
Branches	1-69 µg m^{-2}
Wood	1-54 µg m^{-2}
Foliage	1-43 µg m^{-2}
Bark	1-33 µg m^{-2}



Wet&dry deposition
4.4 - 37 µg m^{-2}

Transpiration
1.7 - 9.2 µg m^{-2}

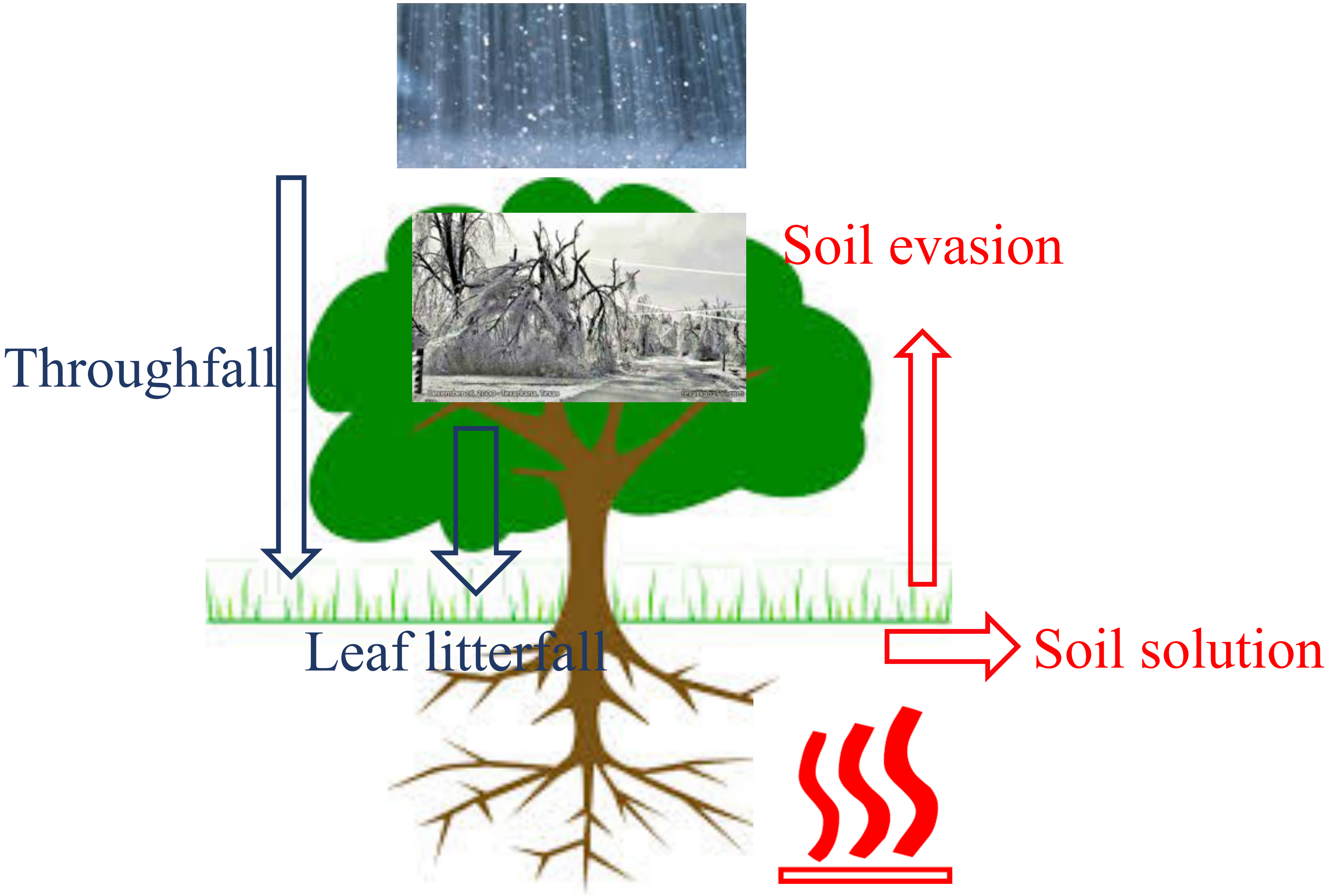
Evasion
-0.1 - 7 µg m^{-2}

Litterfall
4 - 88 µg m^{-2}

Throughfall
6.9 - 18.2 µg m^{-2}

Runoff
0.25 – 0.86 µg m^{-2}

Root flux
1 – 5 µg m^{-2}



Soil evasion

Throughfall

Leaf litterfall

Soil solution

Future directions

The mechanisms of Hg uptake and translocation within the tree are still not well understood. Future studies might make use of Hg stable isotope analysis. Dendrochemistry is also promising. Species differences are intriguing and might shed light on mechanisms. We found that yellow birch accumulated more Hg than the other species we studied.

The impacts of climate change on Hg fluxes will vary with the intensity and frequent of natural disturbances such as hurricanes, ice storms, and wildfires. How altered Hg fluxes due to climate change would influence the retention of Hg in forested ecosystems remains an important area of study. Pathways of atmospheric Hg deposition, leaf litterfall Hg deposition, and throughfall Hg deposition are the main inputs of Hg to the forest floor and have been often reported. Pathways of soil Hg evasion and Hg runoff and leaching are rarely studied.

List of products

Journal Publications

Yanai, R.D., Yang, Y., Wild, A.D., Smith, K.T. and Driscoll, C.T., 2020. New approaches to understand mercury in trees: radial and longitudinal patterns of mercury in tree rings and genetic control of mercury in maple sap. Water, Air, & Soil Pollution, 231:1-10. DOI: 10.1007/s11270-020-04601-2

Yang, Y., L. Meng, R.D. Yanai, M. Montesdeoca, P.H. Templer, H. Asbjornsen, L.E. Rustad, and C.T. Driscoll. 2019. Climate change may alter mercury fluxes in northern hardwood forests. *Biogeochemistry*, 146:1-16. DOI: 10.1007/s10533-019-00605-1

Yang, Y., R.D. Yanai, C.T. Driscoll, M. Montesdeoca and K. Smith. 2018. Concentrations and content of mercury in bark, wood, and leaves in hardwoods and conifers in four forested sites in the northeastern USA. *PLOS One*. DOI: 10.1371/journal.pone.0196293

Yang, Y., Yanai, R.D., Montesdeoca, M., and Driscoll, C.T. 2017. Measuring mercury in wood: Important but challenging. International Journal of Environmental Analytical Chemistry, 97: 456-467. DOI: 10.1080/03067319.2017.1324852

Conference Presentations

Yang, Y., Meng, L., Yanai, R.D., Driscoll, C.T., Montesdeoca, M., Templer, P.H., Rustad, L.E., and Absbjornsen, H. Climate change may worsen mercury pollution in northern hardwood forests. American Geophysical Union Fall Meeting, Washington, D.C. December 14, 2018.

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