Gaseous deposition of atmospheric elemental mercury in ecosystems – what we know and what is missing

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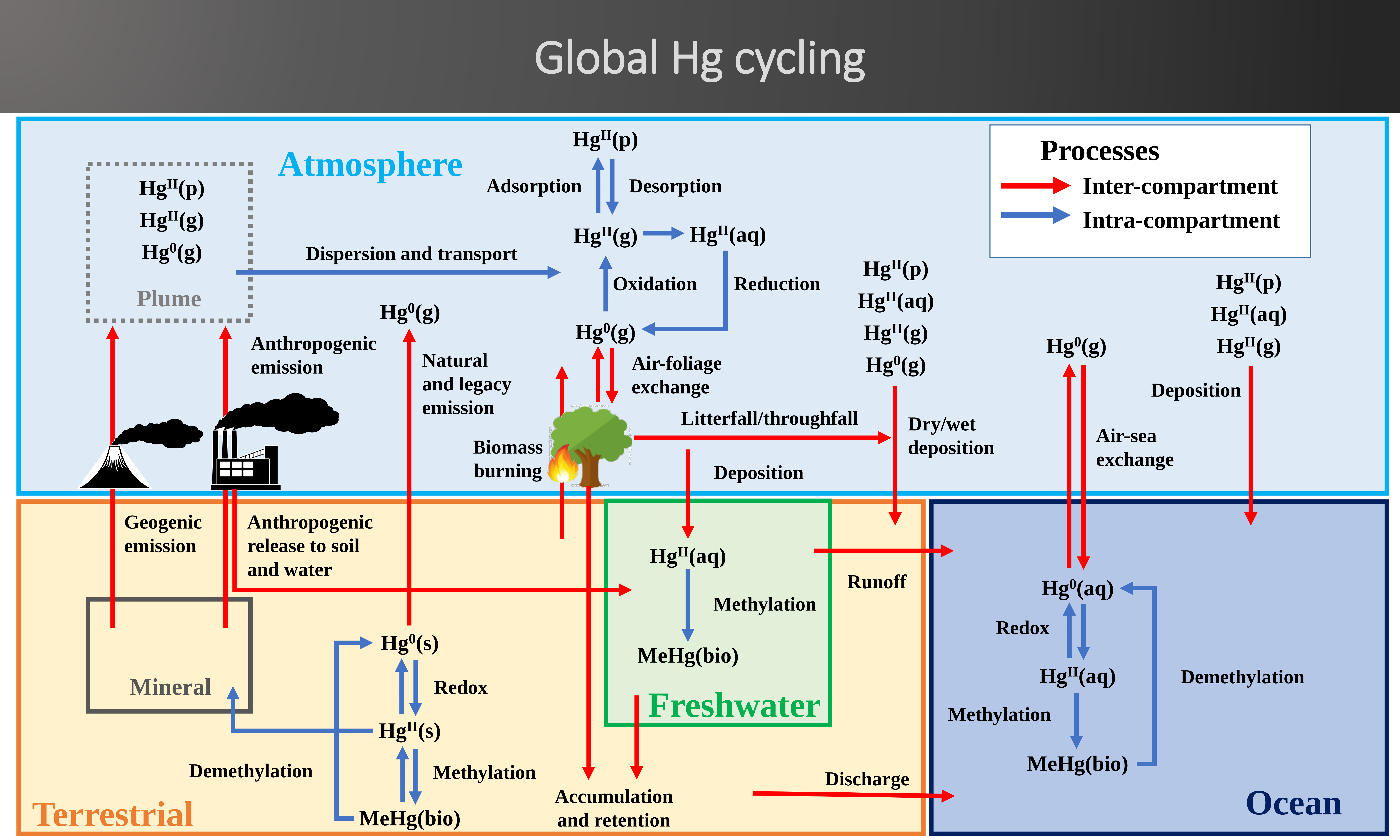

Global Hg cycling
Atmosphere
Processes
Inter-compartment
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Hg^II(p)
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Hg^0(g)
Plume
Hg^II(p)
Adsorption
Desorption
Hg^II(g)
Hg^II(aq)
Dispersion and transport
Oxidation
Reduction
Hg^0(g)
Hg^0(g)
Anthropogenic emission
Natural and legacy emission
Air-foliage exchange
Biomass burning
Litterfall/throughfall
Deposition
Hg^II(p)
Hg^II(aq)
Hg^II(g)
Hg^0(g)
Dry/wet deposition
Hg^0(g)
Air-sea exchange
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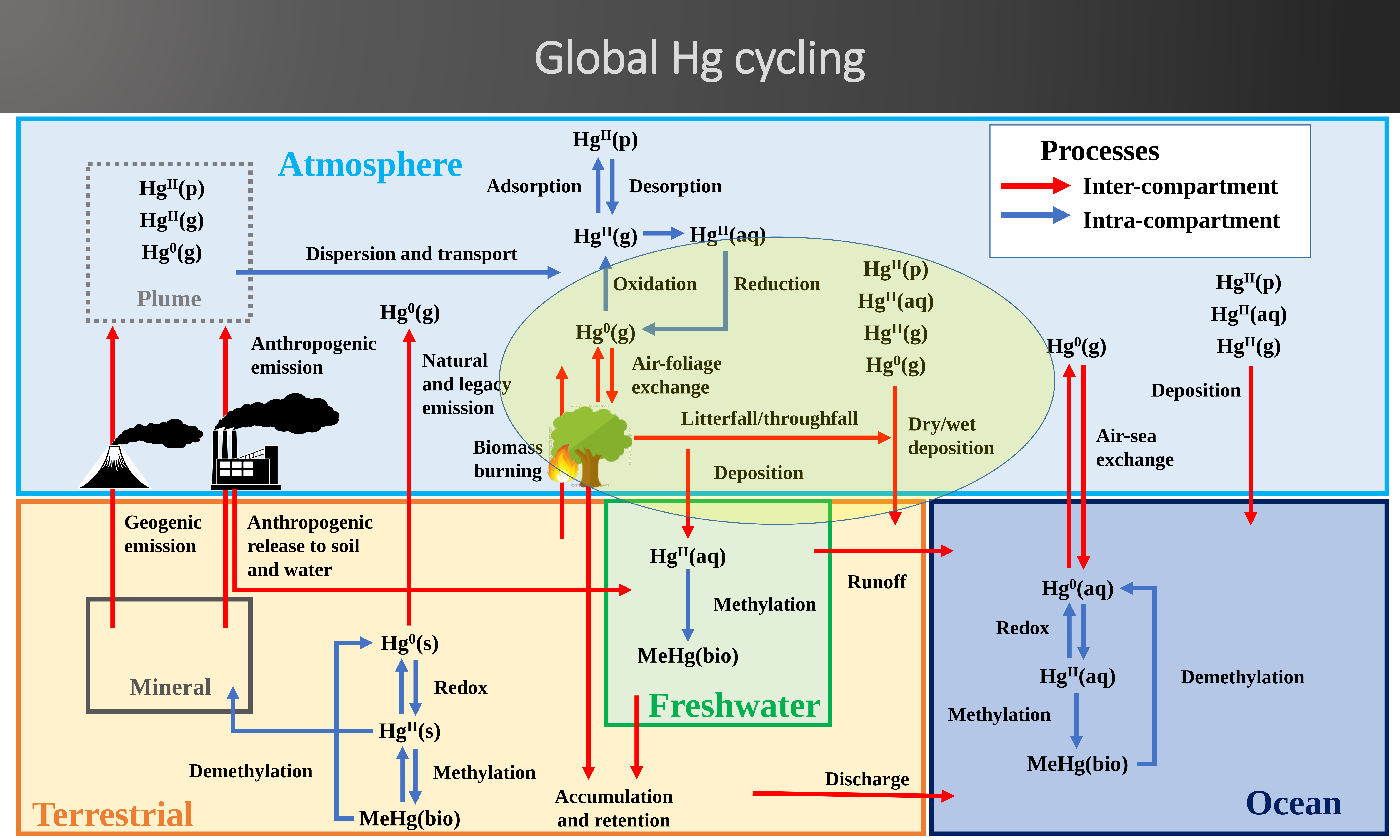
Hg^II(g)
Deposition
Geogenic emission
Anthropogenic release to soil and water
Mineral
Hg^0(s)
Redox
Hg^II(s)
Methylation
Demethylation
MeHg(bio)
Terrestrial
Accumulation and retention
Hg^II(aq)
Methylation
MeHg(bio)
Freshwater
Runoff
Discharge
Hg^0(aq)
Redox
Hg^II(aq)
Methylation
MeHg(bio)
Demethylation
Ocean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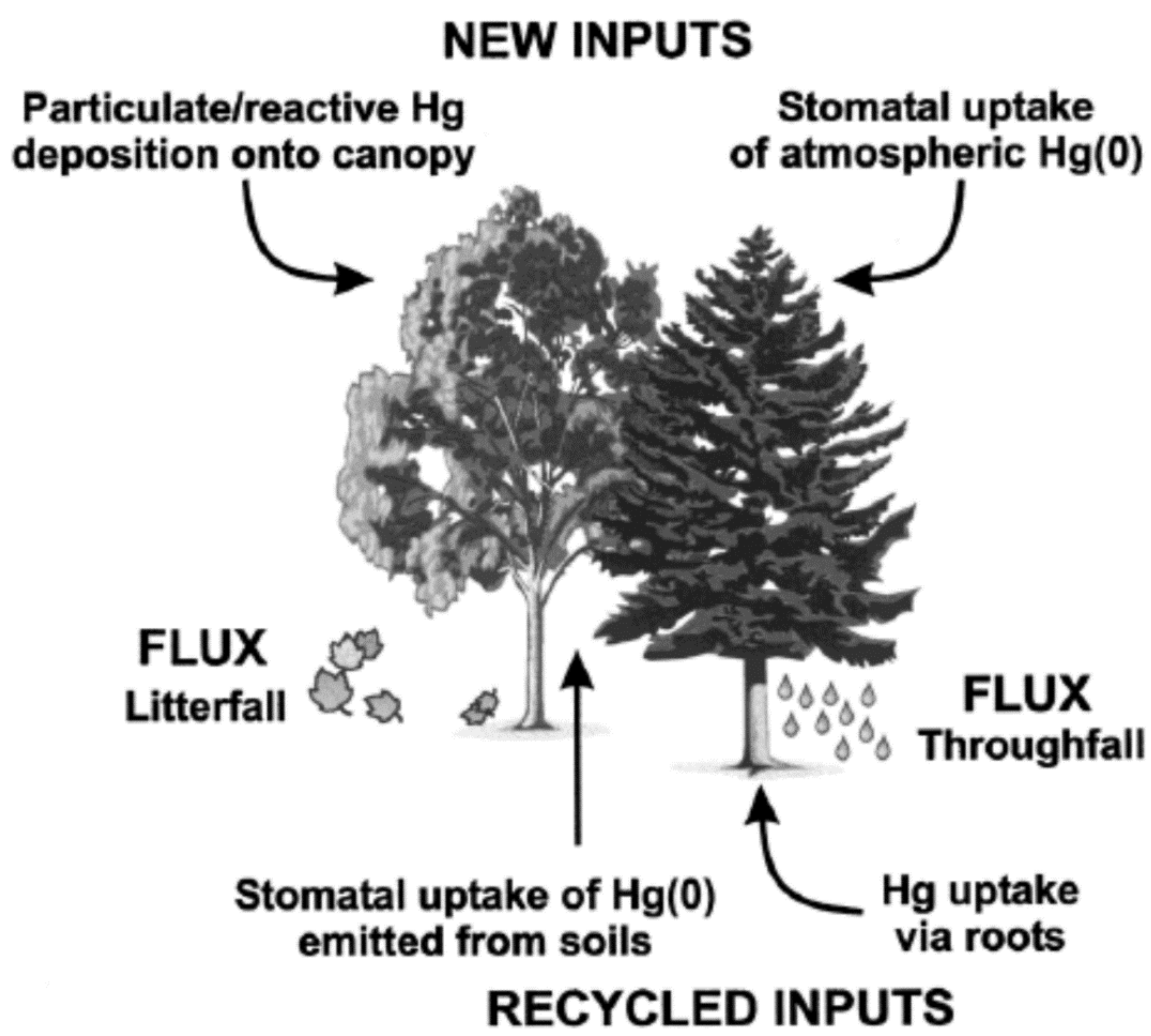
Plant litterfall and throughfall Hg deposition

We've known about the role of plants for dry Hg deposition (Hg^0 and Hg^{II}) for decades



St. Louis et al. (2001) Environmental Science & Technology 2001

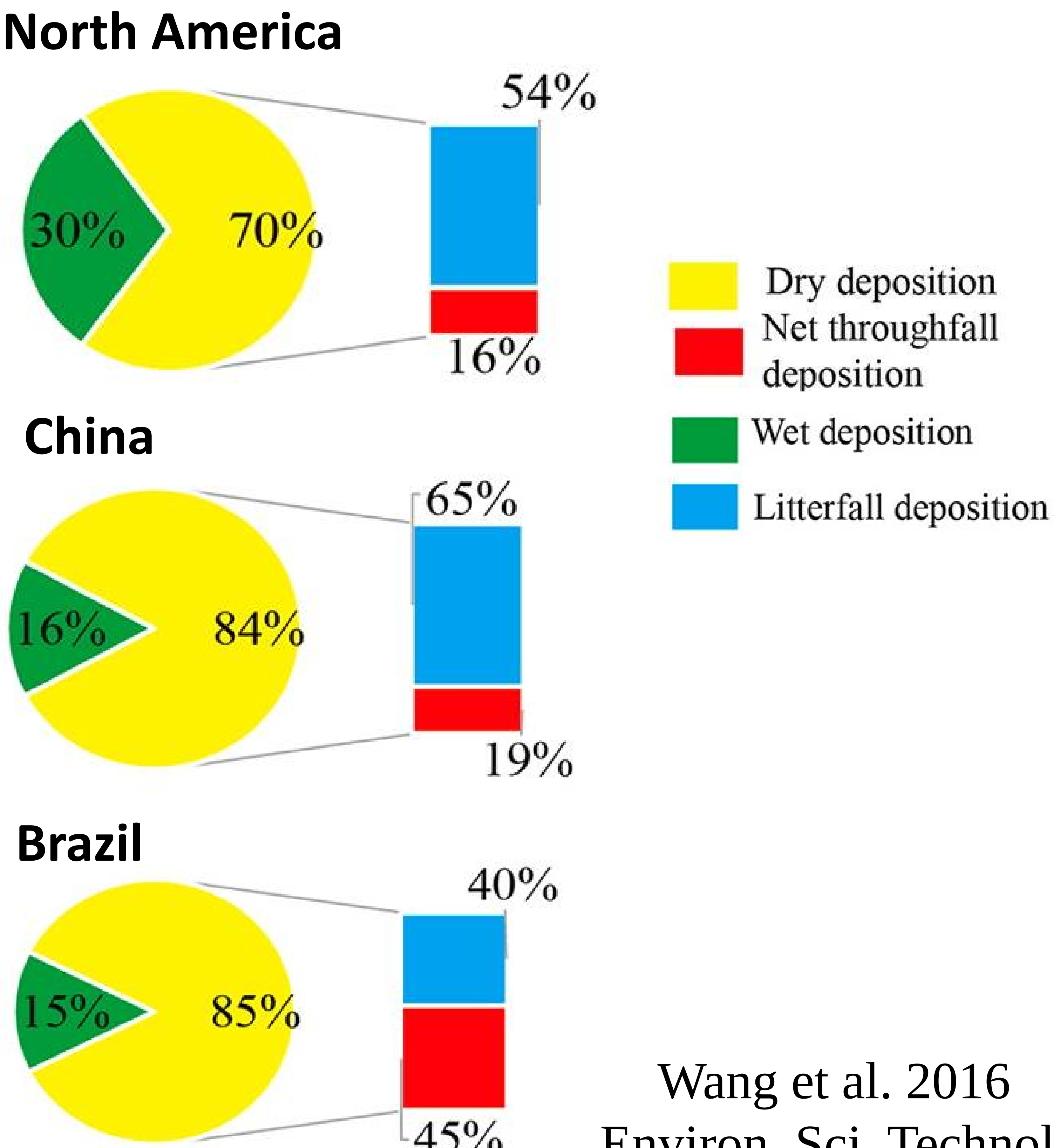
Table II.
Fluxes of Hg in LF, TF and OF precipitation for the period 3/93 to 8/93.

	Litterfall	Troughfall µg m $^{-2}$.	Open Field
Hg deposition	10.4	9.4	5.3

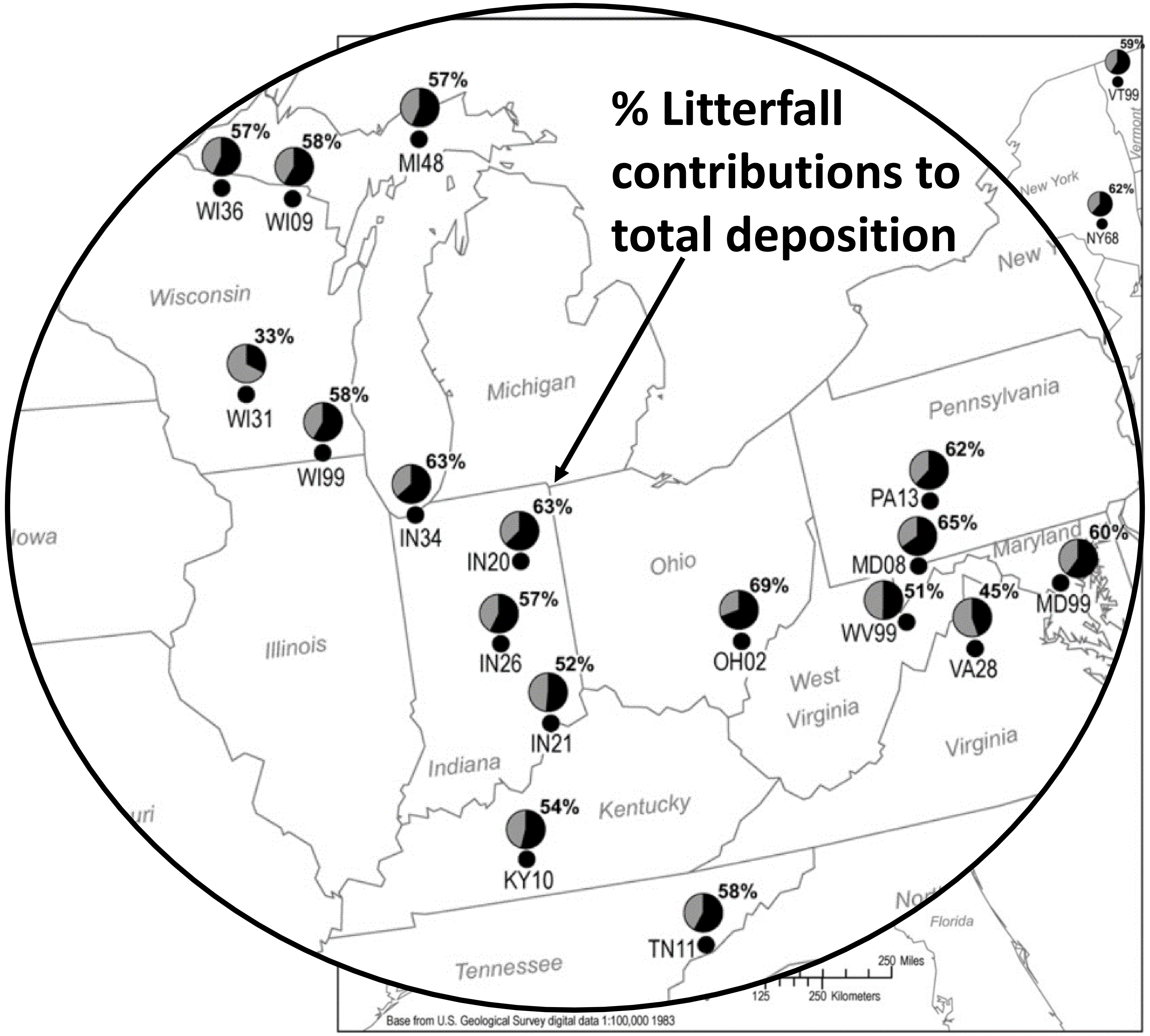
Munthe et al. Water Air and Soil Pollution 1995

Plant litterfall and throughfall Hg deposition

Global litterfall Hg deposition: 1,020–1,230 Mg yr^{-1}



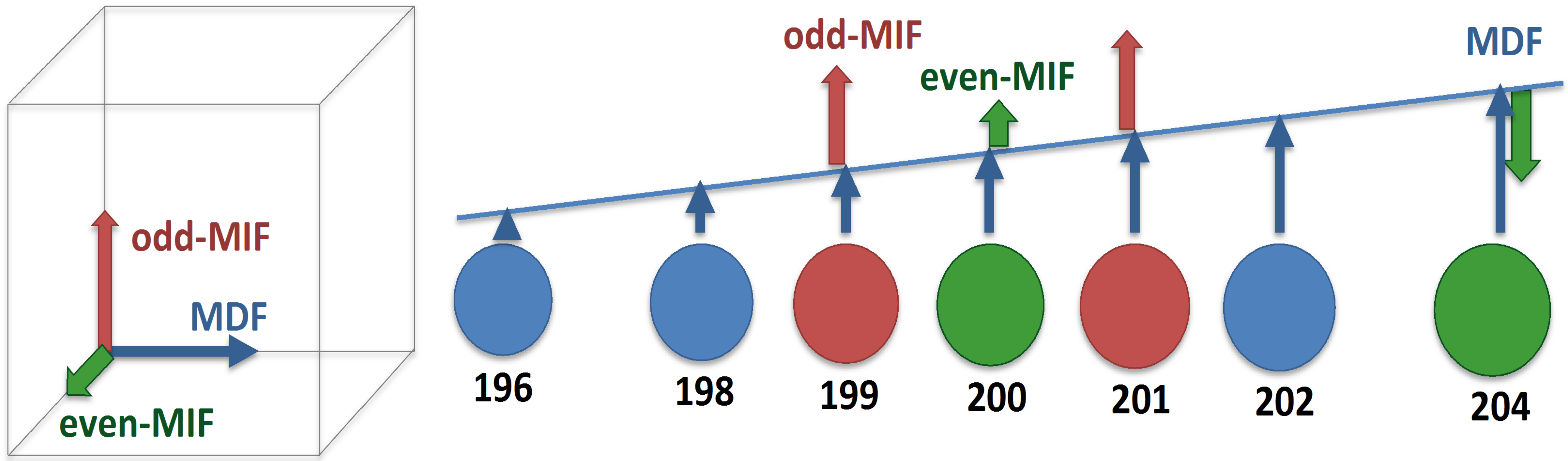
Wang et al. 2016
Environ. Sci. Technol.



Risch et al. Environmental Pollution 2012

Plant litterfall and throughfall Hg deposition

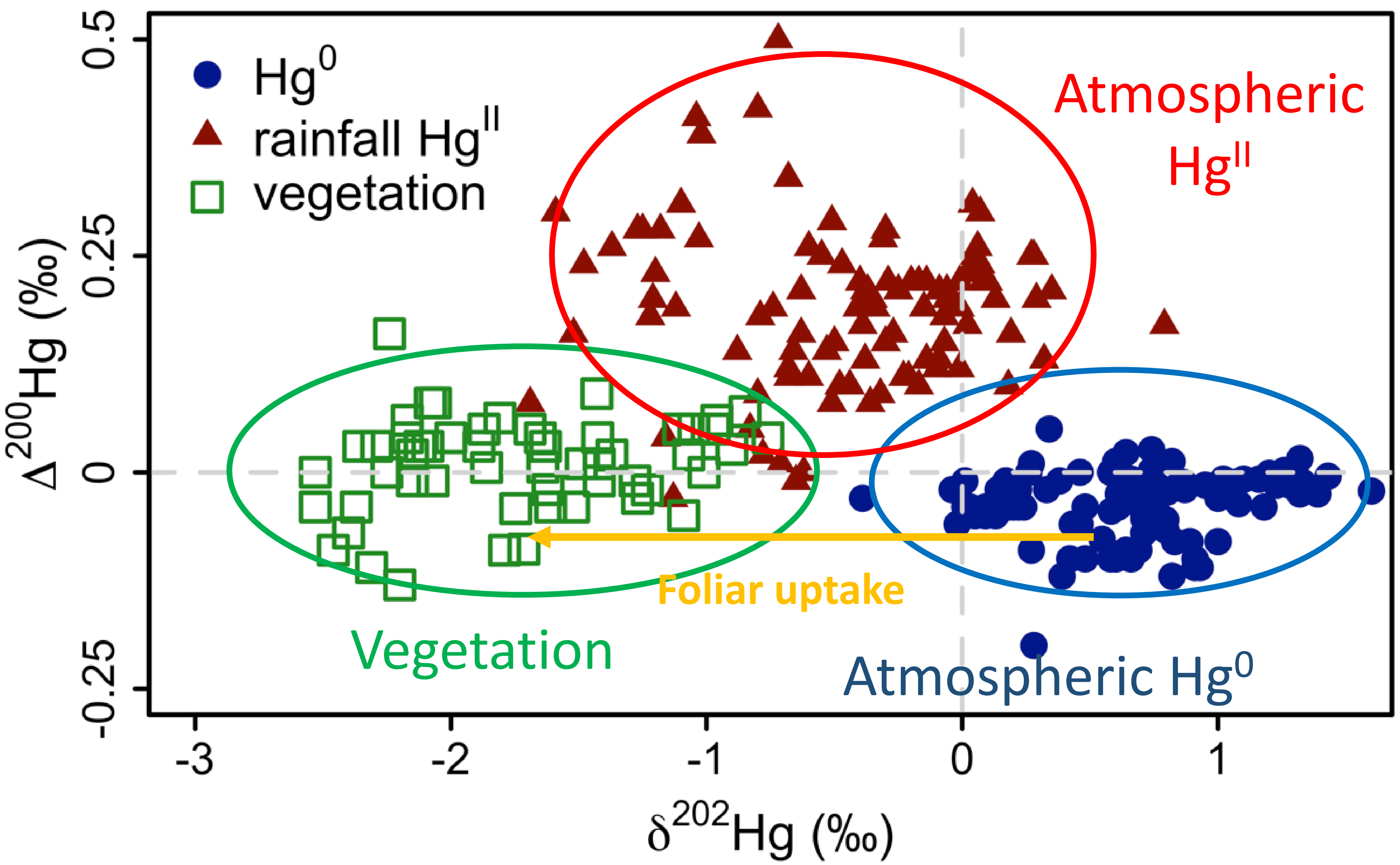
Stable Hg isotopes: new and powerful tool to fingerprint processes and sources



Obrist et al. 2018 Ambio (adopted from Wiederhold et al. 2010 Environ Sci Technol)

Plant litterfall and throughfall Hg deposition

Stable isotope signatures in plant and soils show a dominant atmospheric Hg^0 source in terrestrial ecosystems



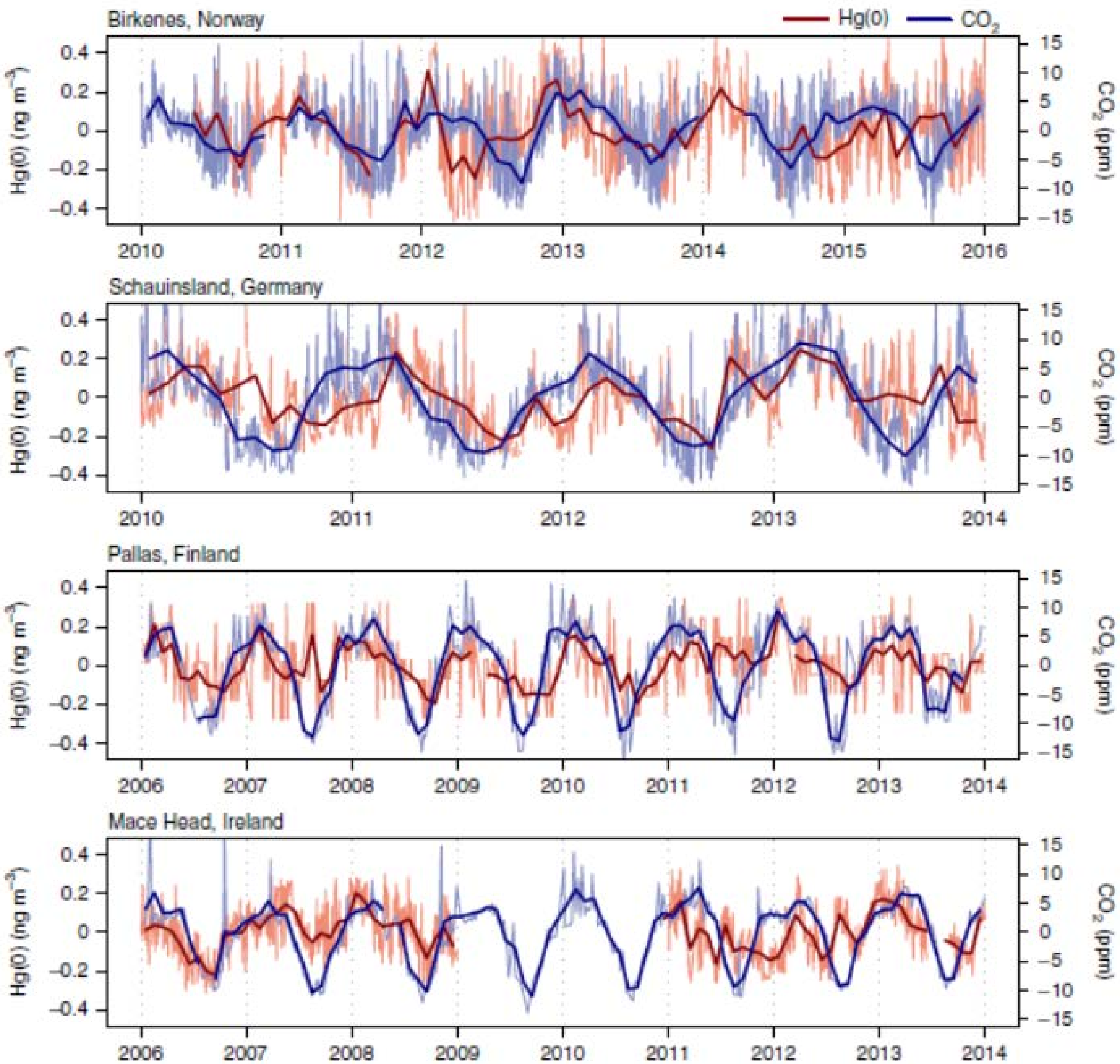
Global literature compilation by M. Jiskra, unpublished

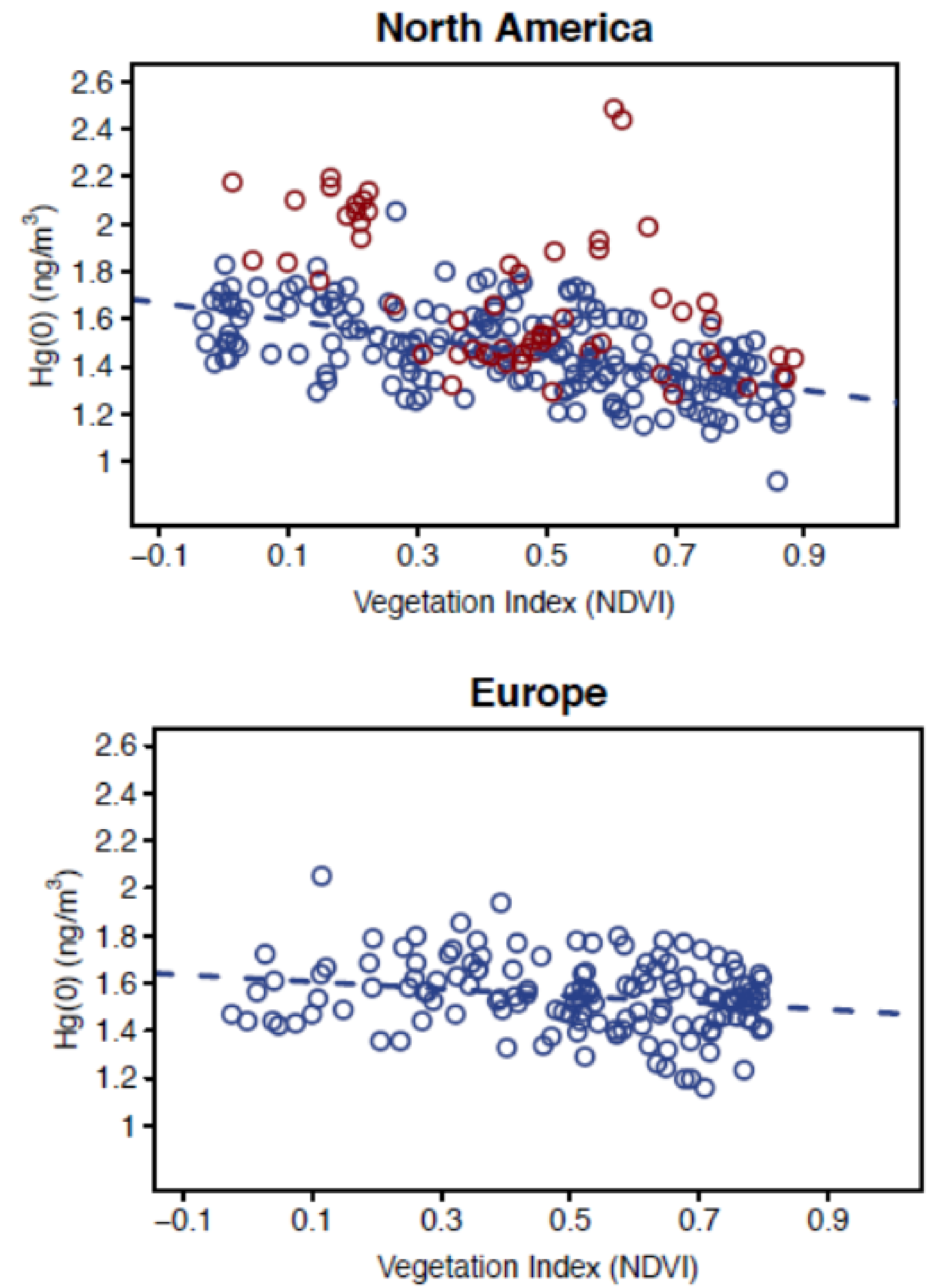
Percentage Hg^0 source of total Hg in soils:

- Central North America: 57–94% (Demers et al. 2013; Zheng et al. 2016)
- Alaskan tundra soils: 71% (Obrist et al. 2017)
- Central European peat soils: 79% (Enrico et al. 2016),
- Boreal forest soils in Sweden: 90% (Jiskra et al. 2015)

Implications of vegetation Hg uptake/deposition

Plant pump strongly controls seasonality of atmospheric Hg^0

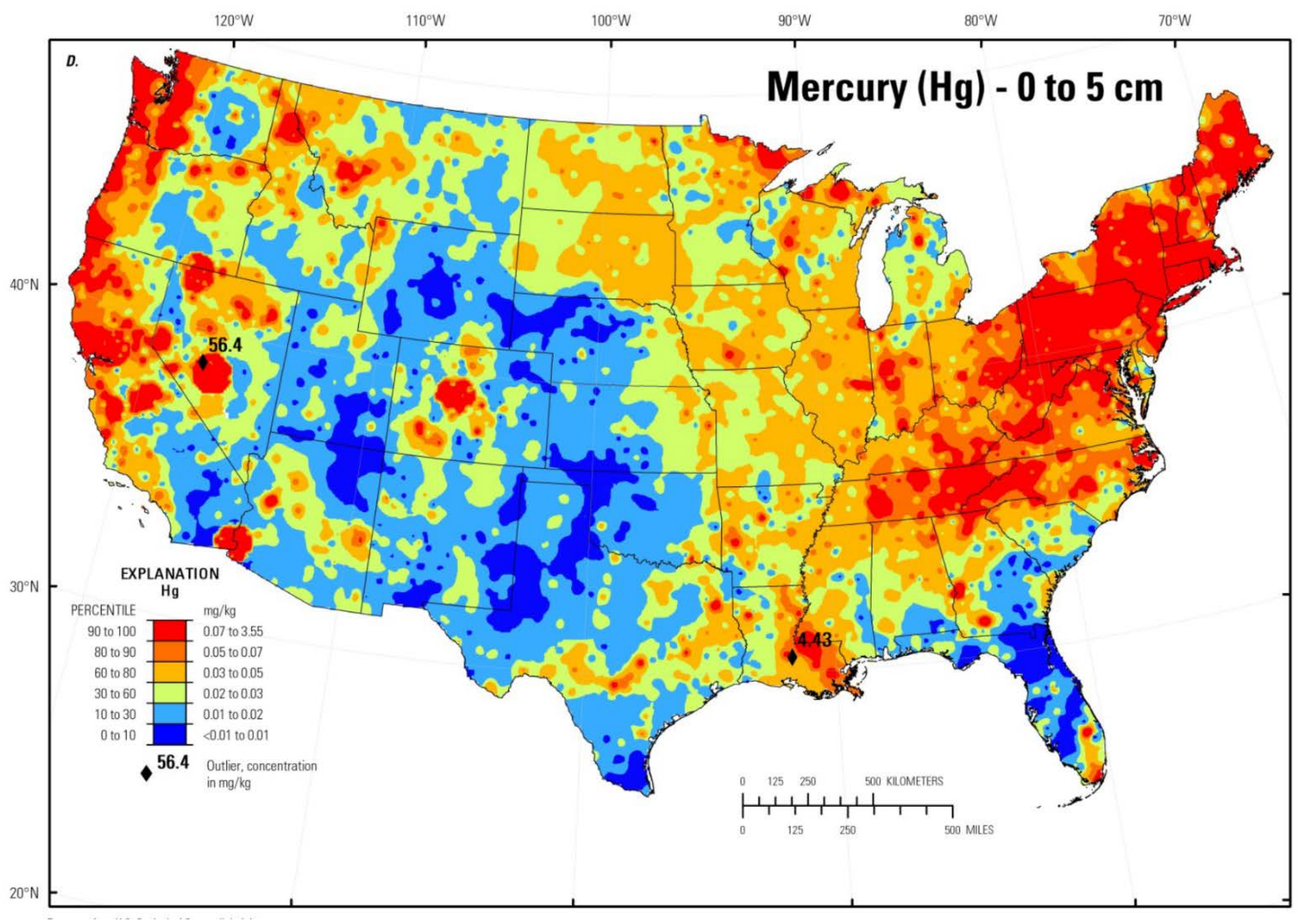




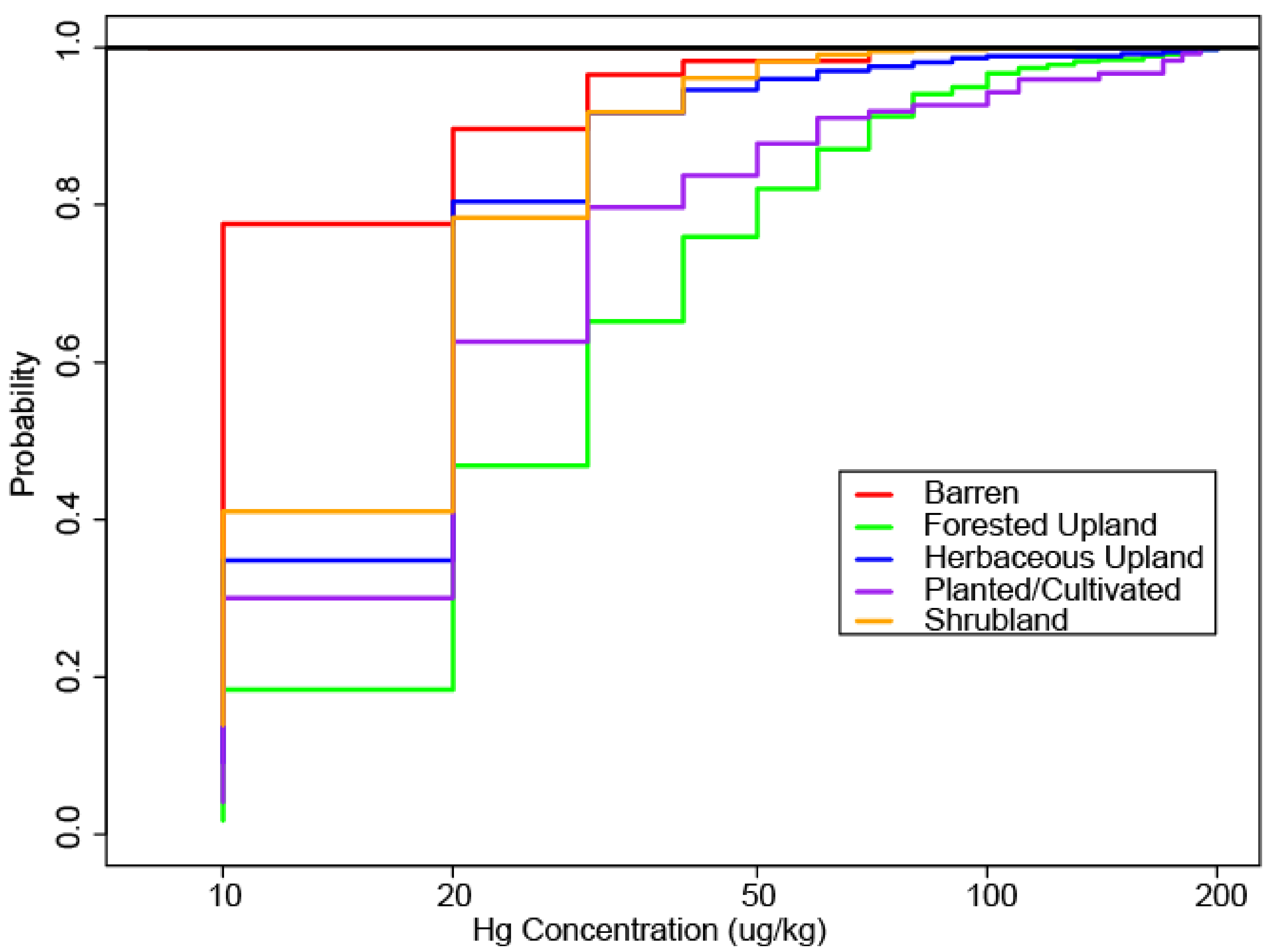
Jiskra et al. 2018. Nature Geoscience

Implications of vegetation Hg uptake/deposition

Plant pump strongly controls concentrations of soil Hg across landscapes



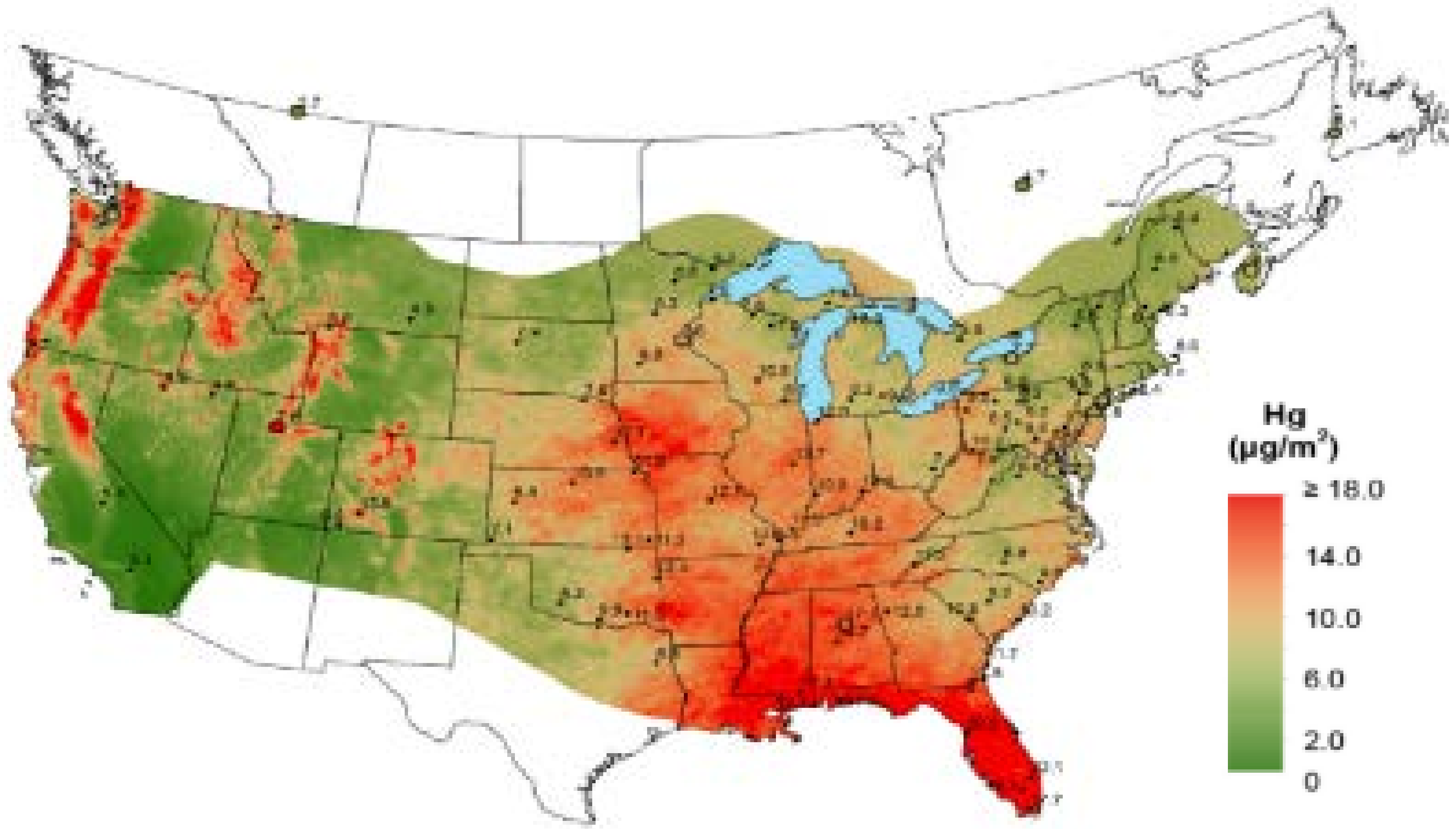
Smith et al. 2014, US. Geol. Survey Data Series



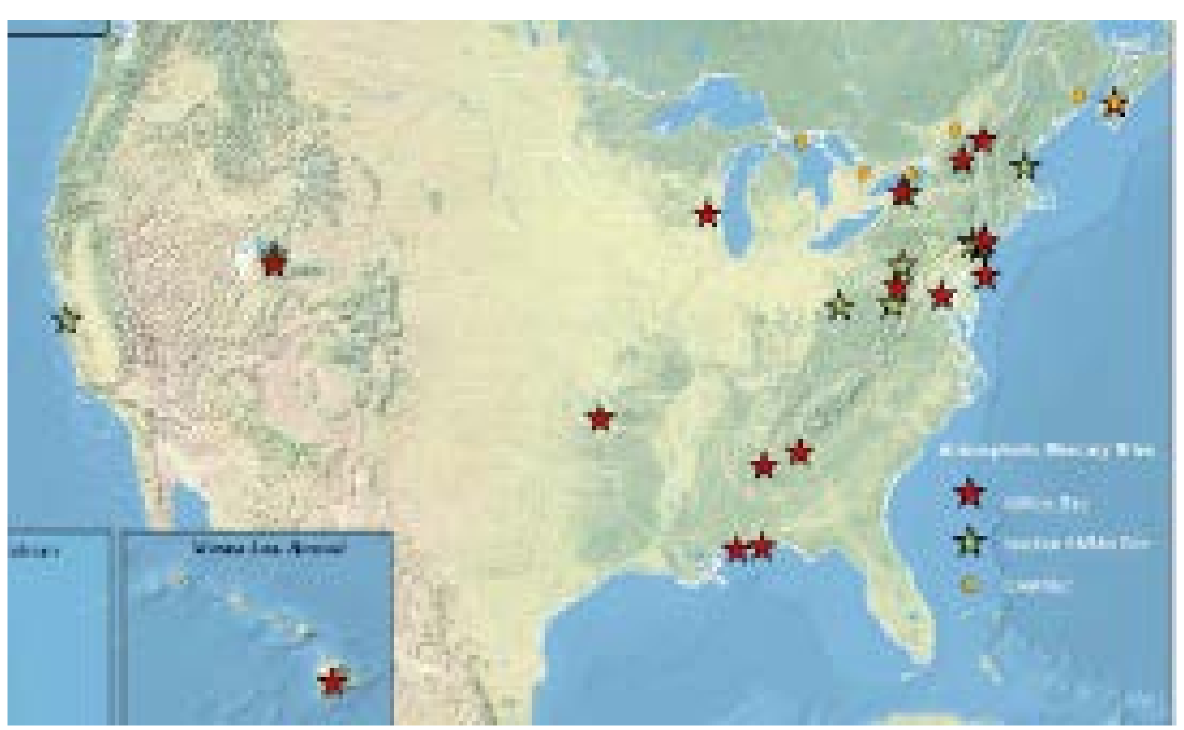
Obrist et al., 2014, STOTEN

NADP Mercury Deposition Network

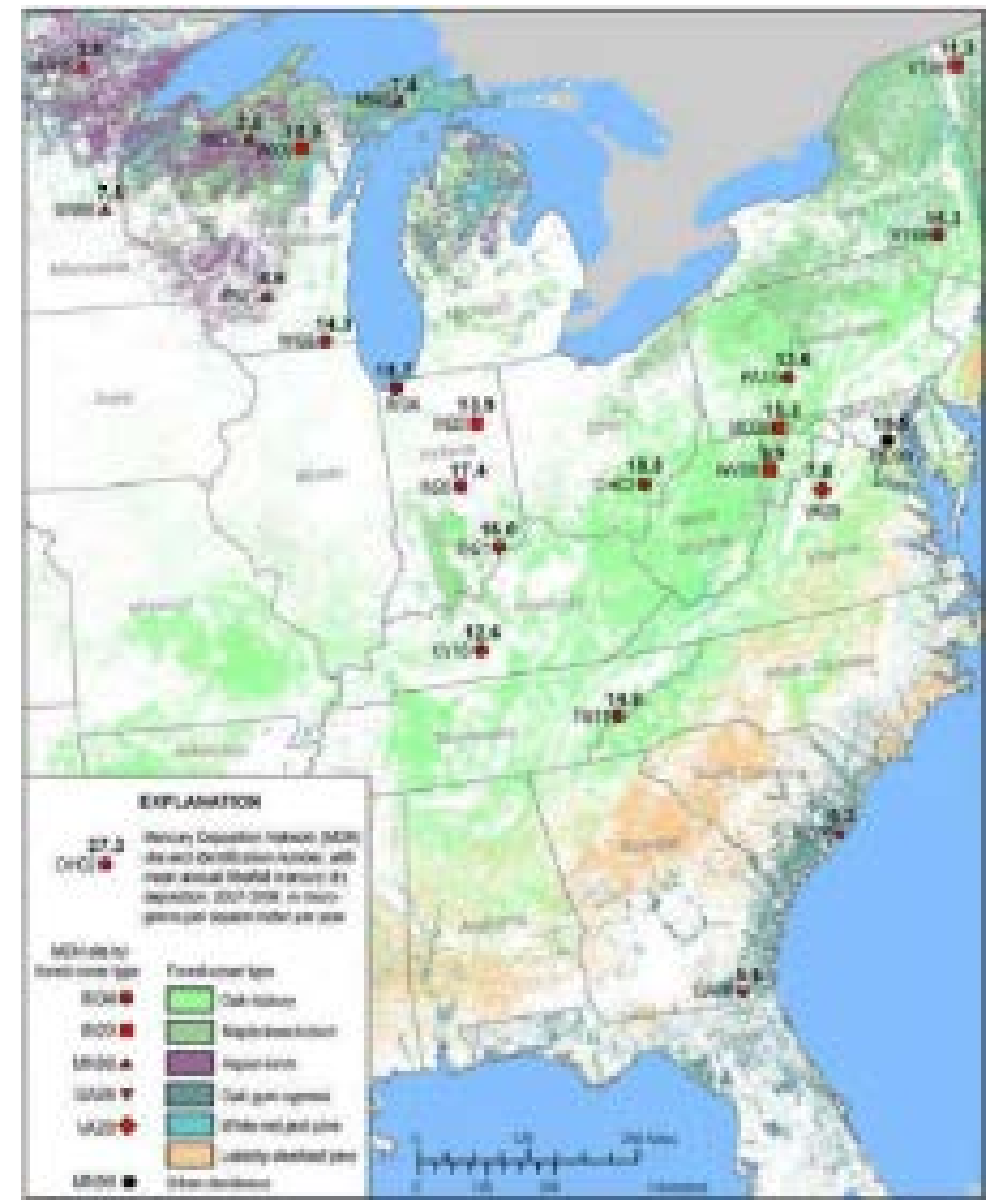
NADP MDN (Wet Hg^{II} deposition)



NADP AMNet (Hg^0 and Hg^{II} conc.)



NADP Litter Deposition Network



Why measuring litterfall deposition is not enough:

- Misses deposition of woody tissues – can equal foliar deposition (Obrist et al. 2012, Melendez-Perez et al. 2014, Yang et al. 2014, Richardson and Friedland 2015, Yang et al. 2017)
- Contains Hg^0 and oxidized Hg^{II}, with relative contributions varying (Zhenget al. 2016)
- Misses re-emissions of Hg^0 – photochemical processes (Agnan et al. 2016, Eckley et al. 2016, Zhu et al. 2016)
- Misses direct Hg^0 exchange to/from soils and snow (Gustin, Lindberg et al. 2000, Choi and Holsen 2009, Fain, Helmig et al. 2013, Obrist, Pokharel et al. 2014, Obrist, Agnan et al. 2017)
- Does not provide temporal patterns of Hg^0 deposition

$\Rightarrow$ **Need for direct assessment of net Hg^0 deposition: gross deposition minus (re-)emission**

Using micrometeorological methods to assess net Hg^0 deposition in Alaska

Using micrometeorological methods to assess net Hg^0 deposition in Alaska

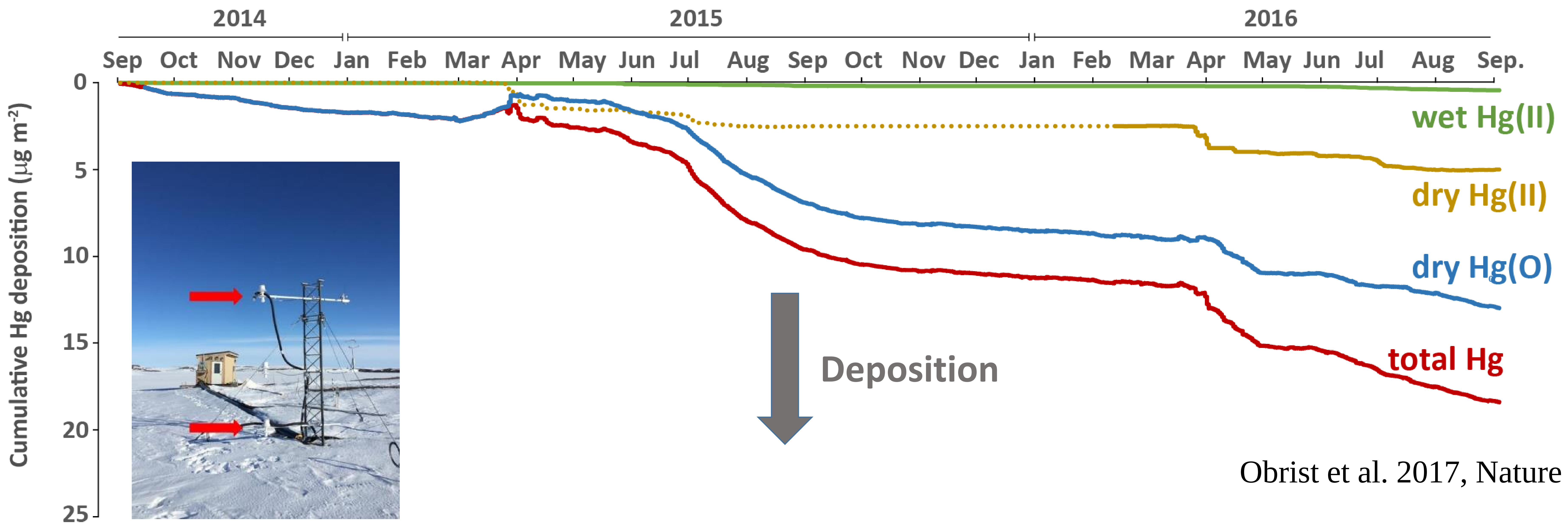
Net Hg^0 exchange (flux-gradient method)

Wet Hg^{II} deposition

Hg^{II} pyrolyzer

GEM: Tekran 2537 Air Hg Analyzers, CO_2, CH_4, O_3, NOx, O_2)

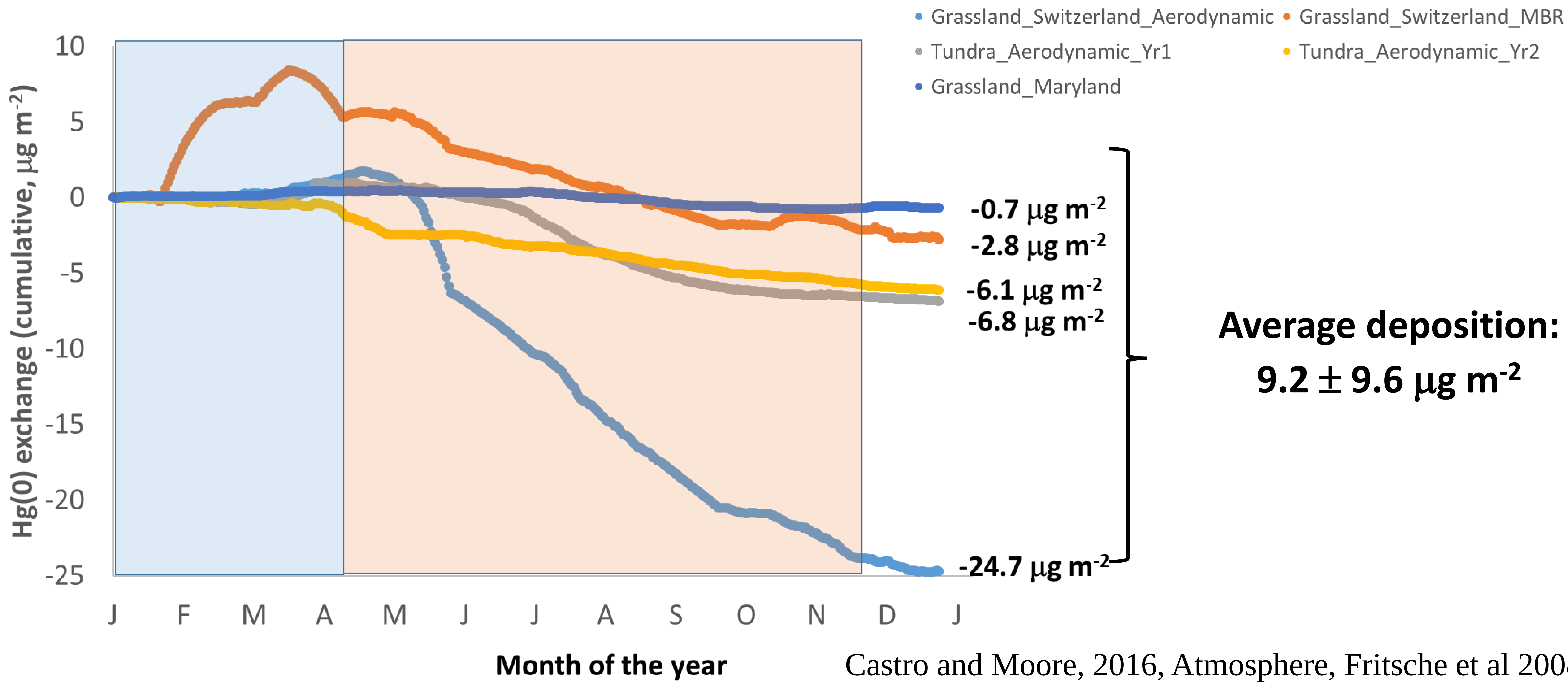
Using micrometeorological methods to assess net Hg^0 deposition in Alaska



71% of atmospheric Hg deposition in the Arctic tundra is Hg^0

Annual records of annual Hg^0 net exchange (deposition)

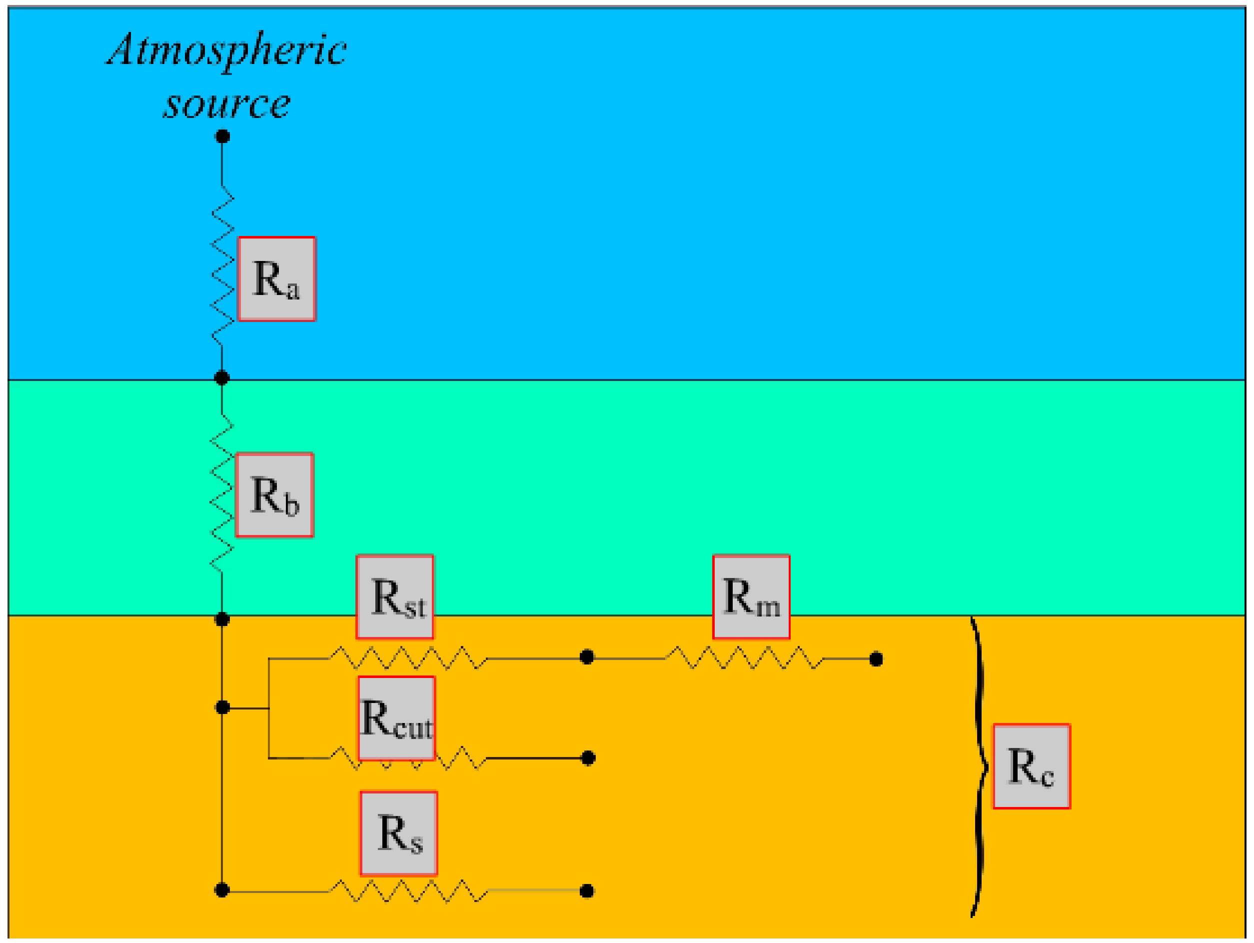
Only a few annual data sets of net Hg^0 exchange exist (upland sites only)



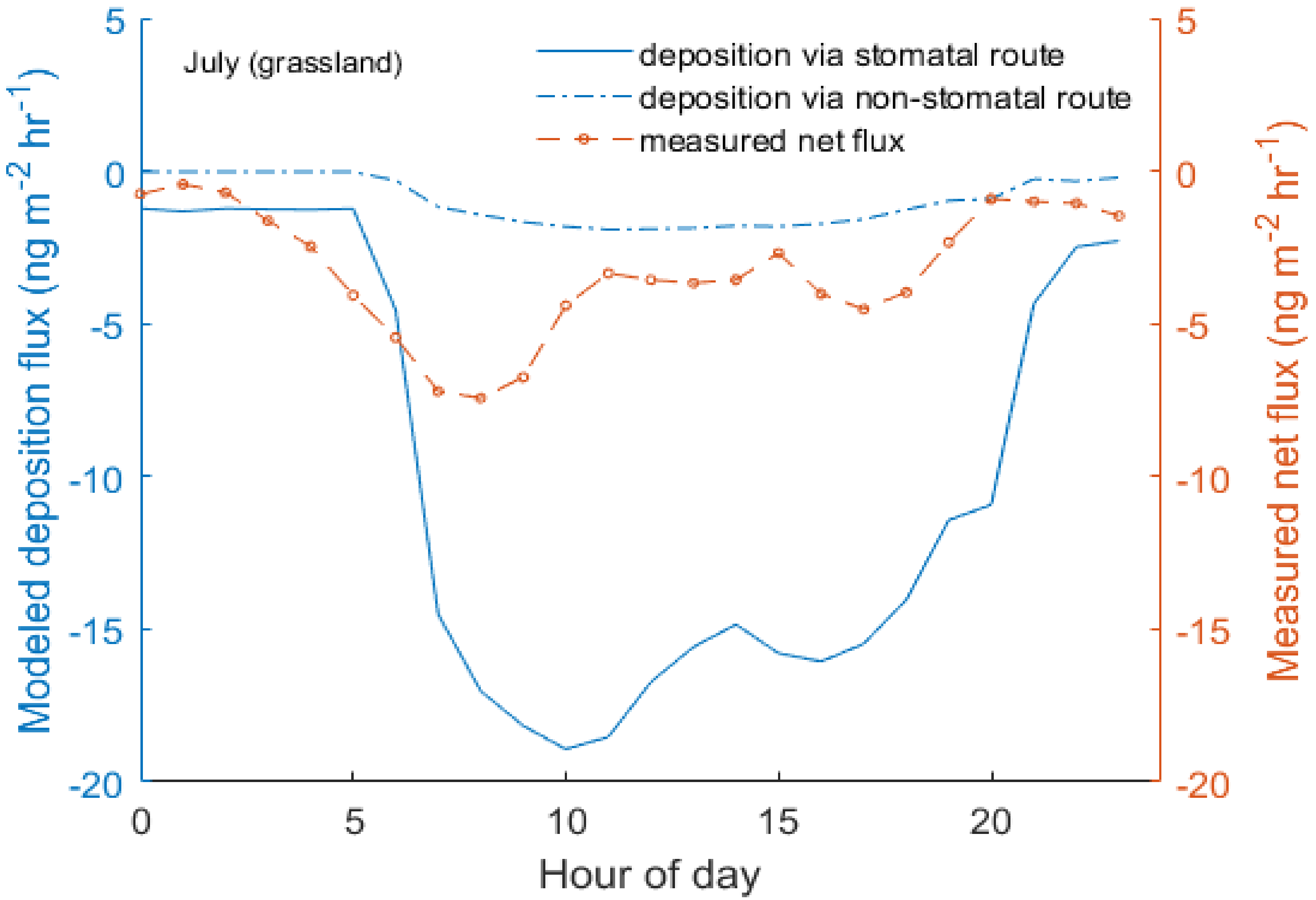
Castro and Moore, 2016, Atmosphere, Fritsche et al 2008, Atmos. Environ., Obrist et al., 2017, Nature

Constraining models using time series Hg^0 deposition measurements

CTMs use resistance-based modeling approaches for Hg^0 exchange

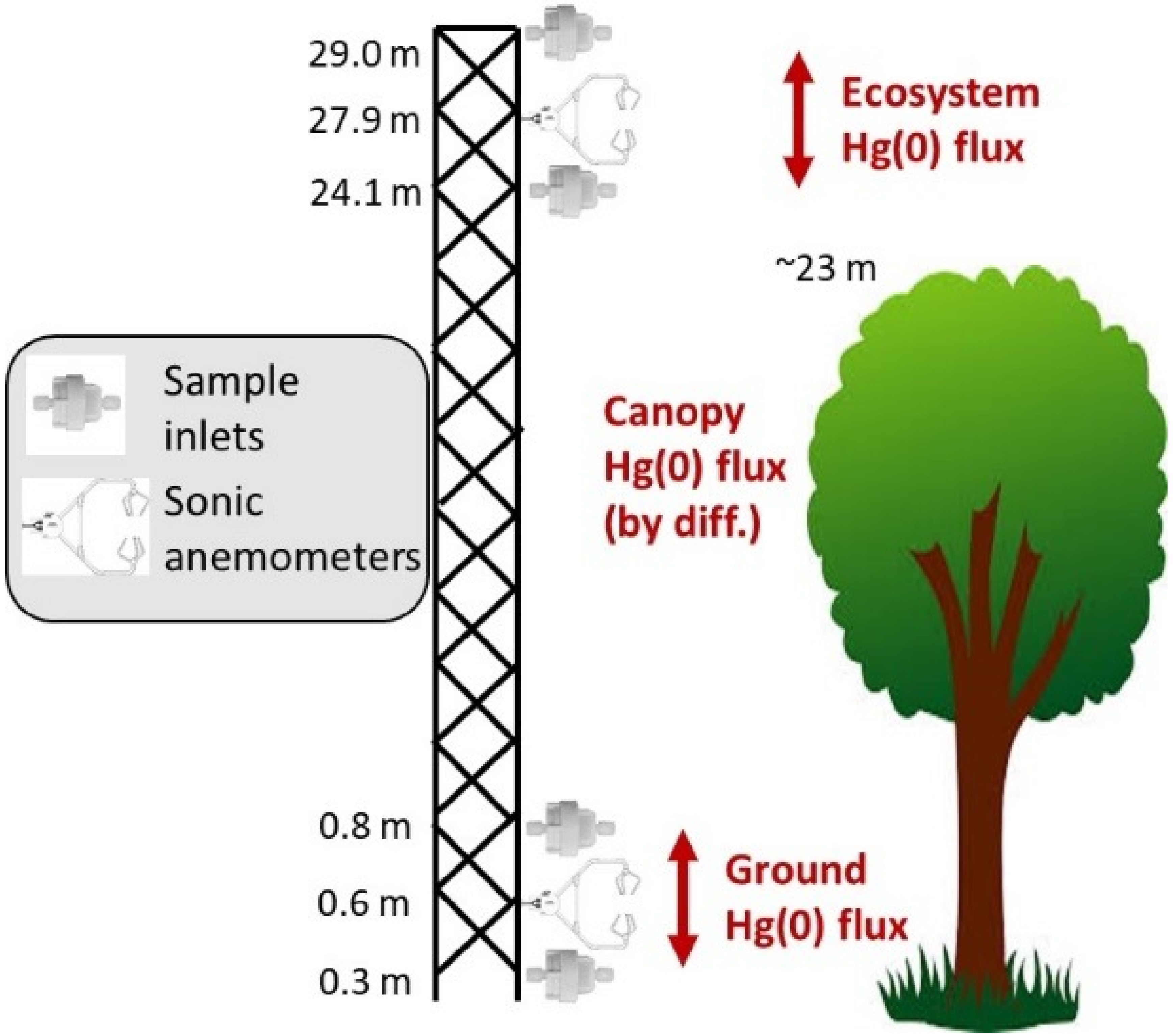


Khan et al. (in review)



What about forests?

We need direct Hg^0 flux measurements over forests

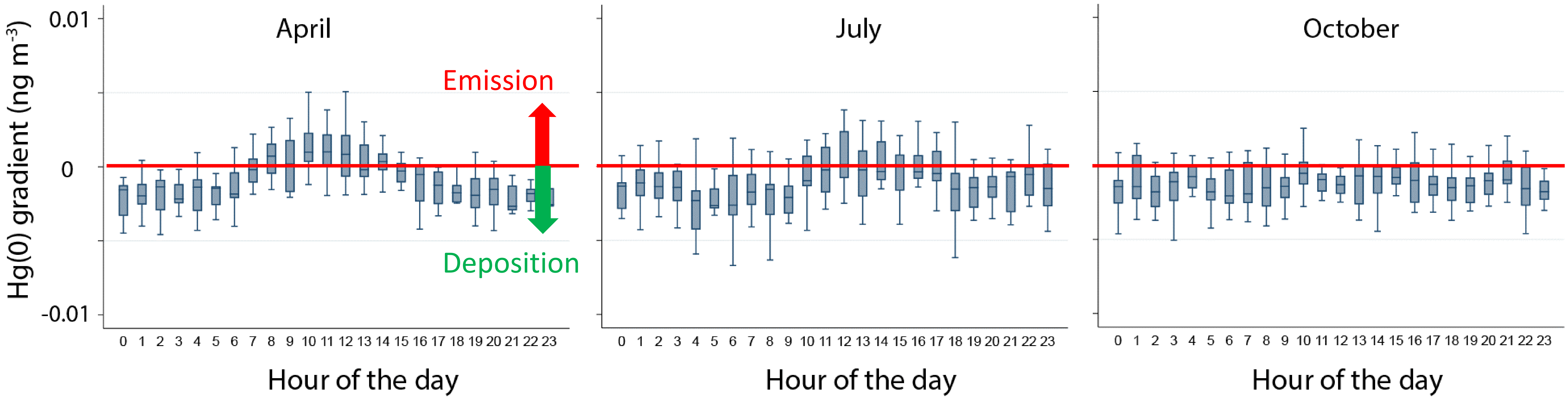


Harvard Forest Measurement Tower

NSF ATC Project: 2019-2022: PI: Obrist, Co-PI: Commane

What about forests?

Hg^0 concentration gradients (~ fluxes) measured over Harvard Forest



Howard et al., in preparation
Poster Session: Howard et al.

Summary

Dominant source of Hg in (most) terrestrial ecosystems derived from atmospheric Hg^0 deposition, largely derived from plant inputs

Direct net Hg^0 deposition measurements lacking over most ecosystems, in spite of applicable techniques

Diel and seasonal Hg^0 deposition patterns needed to constrain Hg^0 deposition parameterization in CTMs

Current measurements support net annual Hg^0 deposition over grasslands/tundra. Measurements over forests lacking (but feasible)