A proposed coupling of natural mercury emissions and deposition in CMAQ

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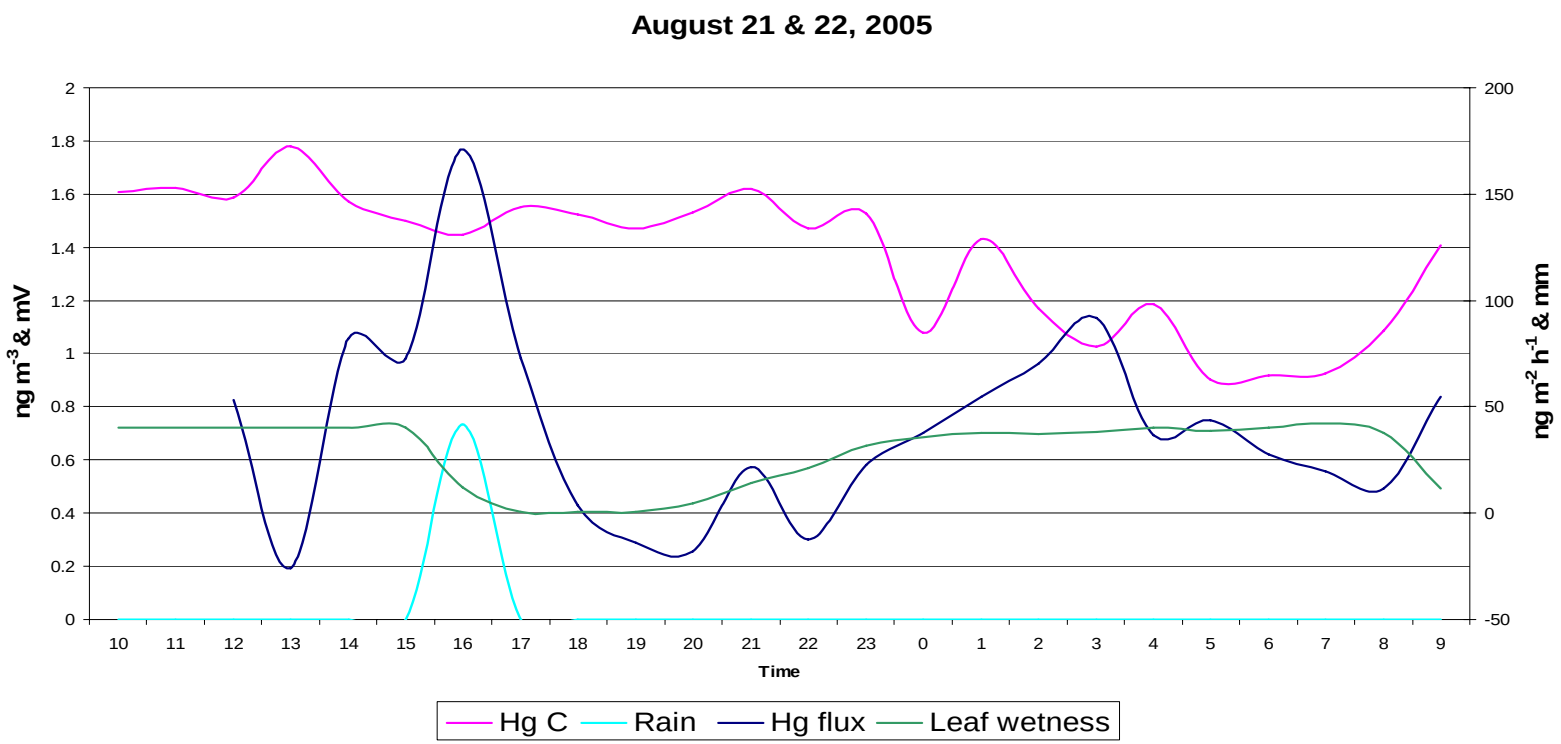
HgSim (Bash et al. 2004)

- Current natural emissions model
- Land surface emissions
  - Function of evapotranspiration, land cover and temperature
- Emissions from water
  - Function of concentration gradient, surface friction velocity and temperature
- Not currently coupled to CMAQ

Mercury flux REA system

- Mercury flux measurements were taken over a hardwood forest canopy
- The relaxed eddy accumulation (REA) technique was used
- Co-located meteorological instruments
- Sampling of environmental media
  - Leaves, soil, precipitation, soil water, and atmospheric Hg concentrations

Mercury flux time series



Mercury flux observations

- A dynamic system
  - Stomata, temperature/solar radiation, and precipitation influences on the flux
- Different dynamics during periods of wet and dry leaf surfaces
  - Higher net flux for a dry canopy (51 ng m^{-2} h^{-1} vs 12.4 ng m^{-2} h^{-1})

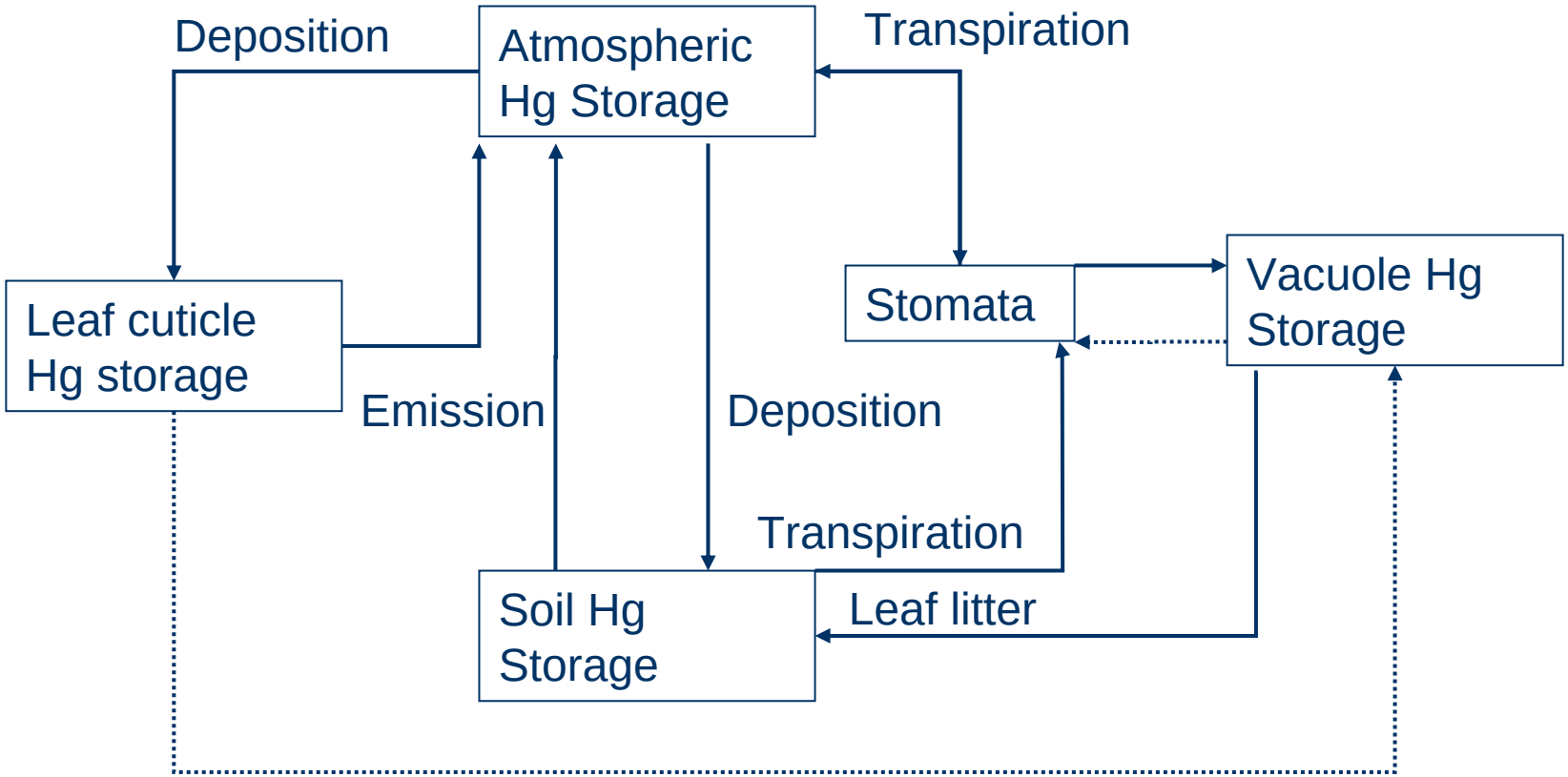
Mercury flux observation

- Highest emission rates seen following precipitation (a mean of 59.8 ng m^{-2} h^{-1})
  - Similar results seen in emissions from soil (Gillis and Miller, 1999)
  - Source of the spikes could be from the forest soil
- Deposition and emissions at the leaf cuticle surface
  - Enhanced by the dew on the canopy
  - Accumulation of Hg on the Cuticle was not observed

Mercury flux observation

- Seasonal trends
  - Deposition in the spring to strong emissions in the fall
  - Accumulation of mercury in forest foliage
- Compensation point observed in time series
  - Stronger emissions during times of low atmospheric concentrations
  - Stronger deposition during times of high atmospheric concentrations

Flow chart of hypothesized surface-air exchange



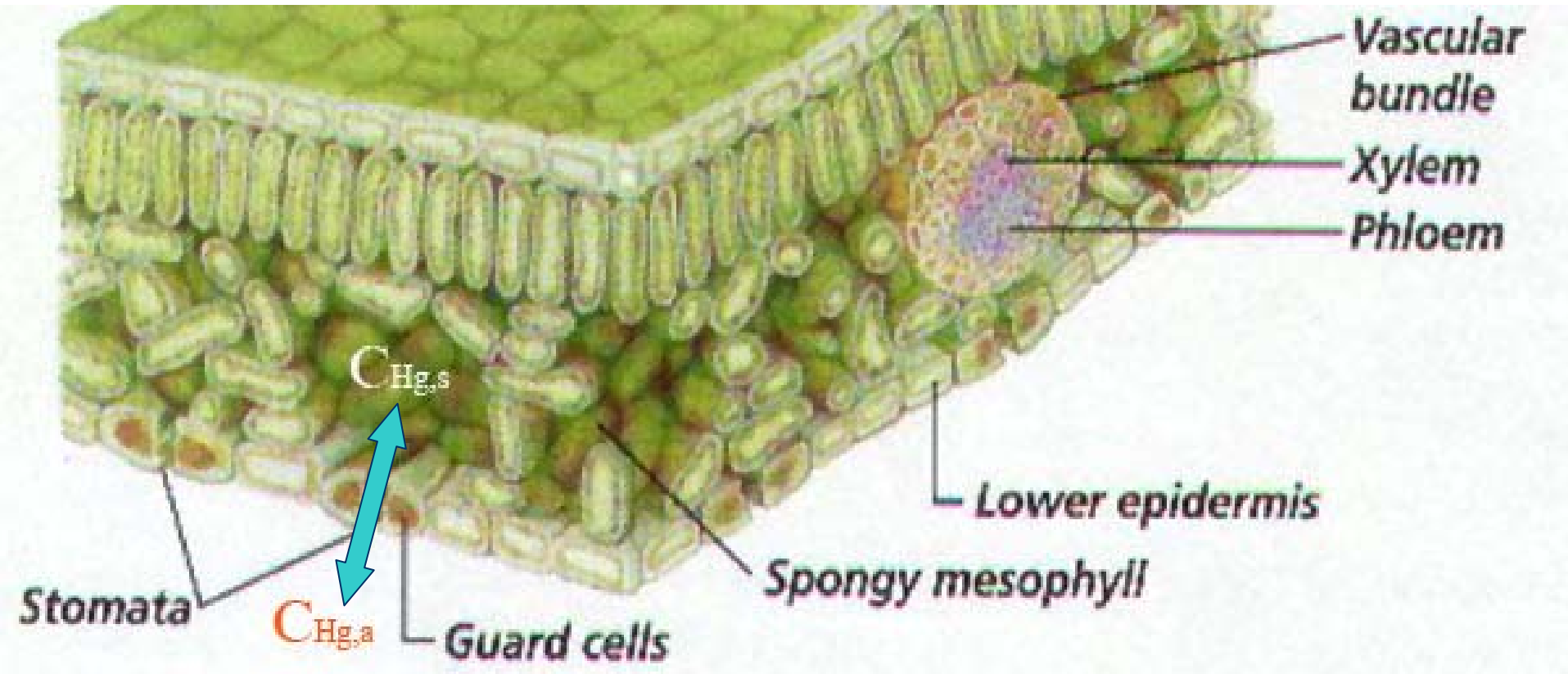
Modeling the mercury flux over a forest canopy

- The emissions model should be coupled with an air quality model
  - Gradient dependent fluxes
  - Deposition and emission processes
  - Soil flux under the canopy
- Coupling via the concentration gradient across the stomata and storage in the canopy (leaf surfaces and vacuoles) and soil

Stomatal Pathway

- Important in deposition and emissions
  - Transpiration of mercury
- Accumulation in forest foliage
  - Storage in the vacuoles
  - Unclear if the source is atmospheric or terrestrial
- Gradient dependent effects on the flux
  - Compensation point seen in literature

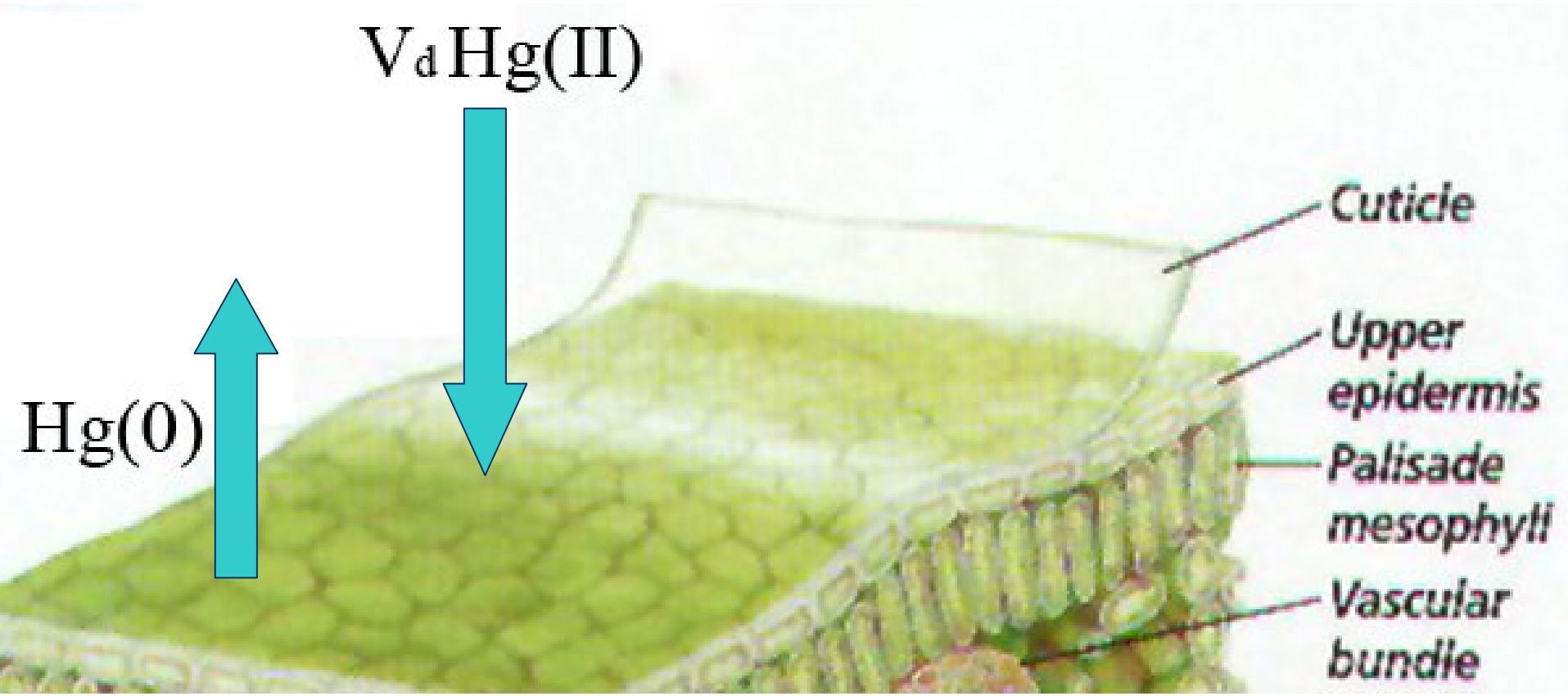
Stomatal Pathway



Cuticle Pathway

- Can be modeled as deposition to the cuticle and volatization from the cuticle
- Measurements of mercury on leaf surface
  - Shown deposition but little accumulation
- Hypothesized that deposition is Hg(II) and Particulate
- Emissions could be modeled as a photoreduction
- Hg possibly stored in the leaf vacuoles

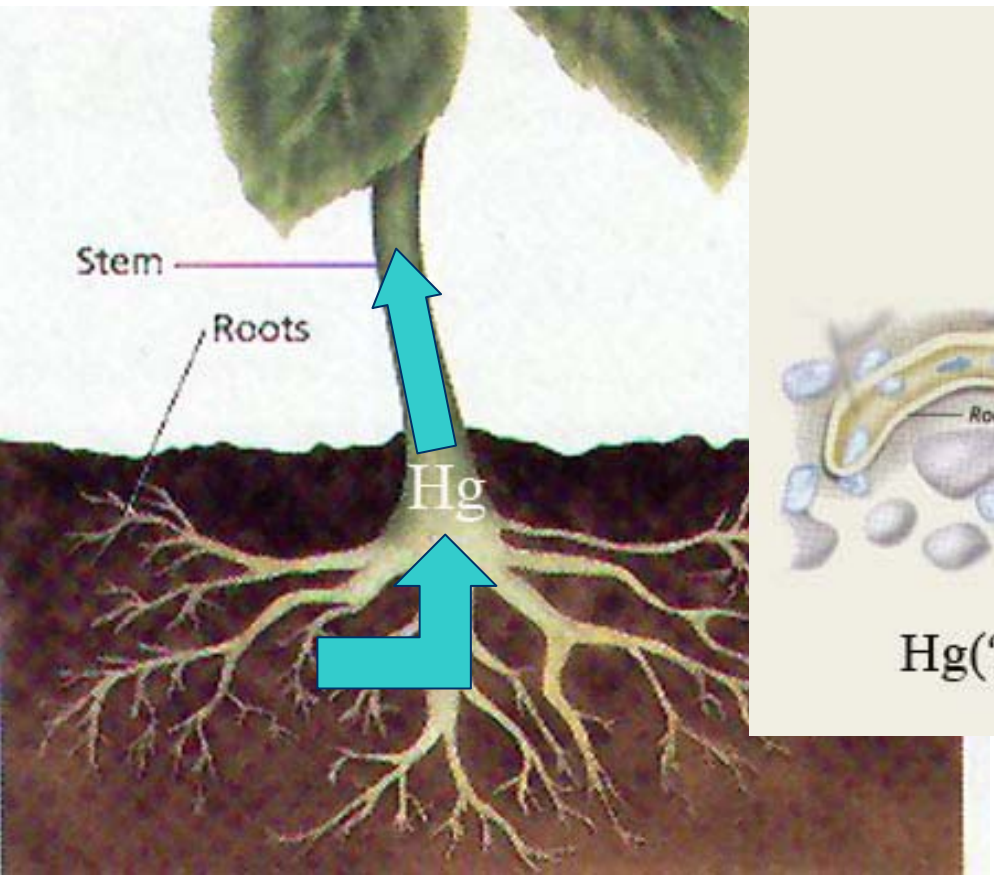
Cuticle Pathway

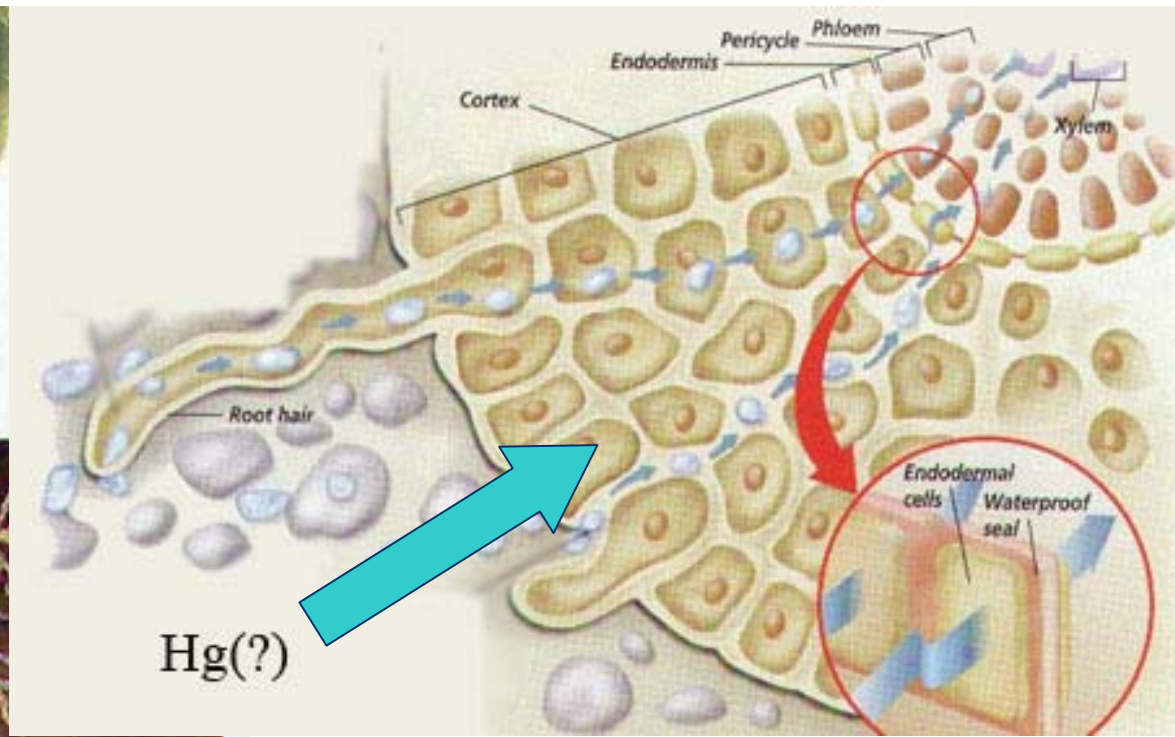


Forest Soil Pathway

- Can be modeled as a deposition and emission process from soil surface
- Storage will couple deposition and emissions
- Largest influx of Hg is from leaf litter
- Spikes after precipitation
  - Escaping of interstitial soil air?
  - Release of Hg bound to soil particles into soil water solution and subsequent volatization?

Forest Soil Pathway





Conclusions

- Multi-layer canopy model suited to this approach
  - Useful in partitioning of storages
    - In the vacuoles, on the leaf cuticles, and soil
  - Improved deposition and emissions over a big leaf model
  - Better representation of non-linearities in canopy (Radiation, Stomatal resistance, Dew formation, Hg Concentration gradient, etc.)
- More measurements with model comparisons are needed

Thank you!

Questions?

UConn REA System