LOCAL FORMATION VERSUS REGIONAL CONTRIBUTIONS TO SECONDARY ORGANIC AEROSOLS

CLIMATE AND SPACE SCIENCES AND ENGINEERING
UNIVERSITY OF MICHIGAN

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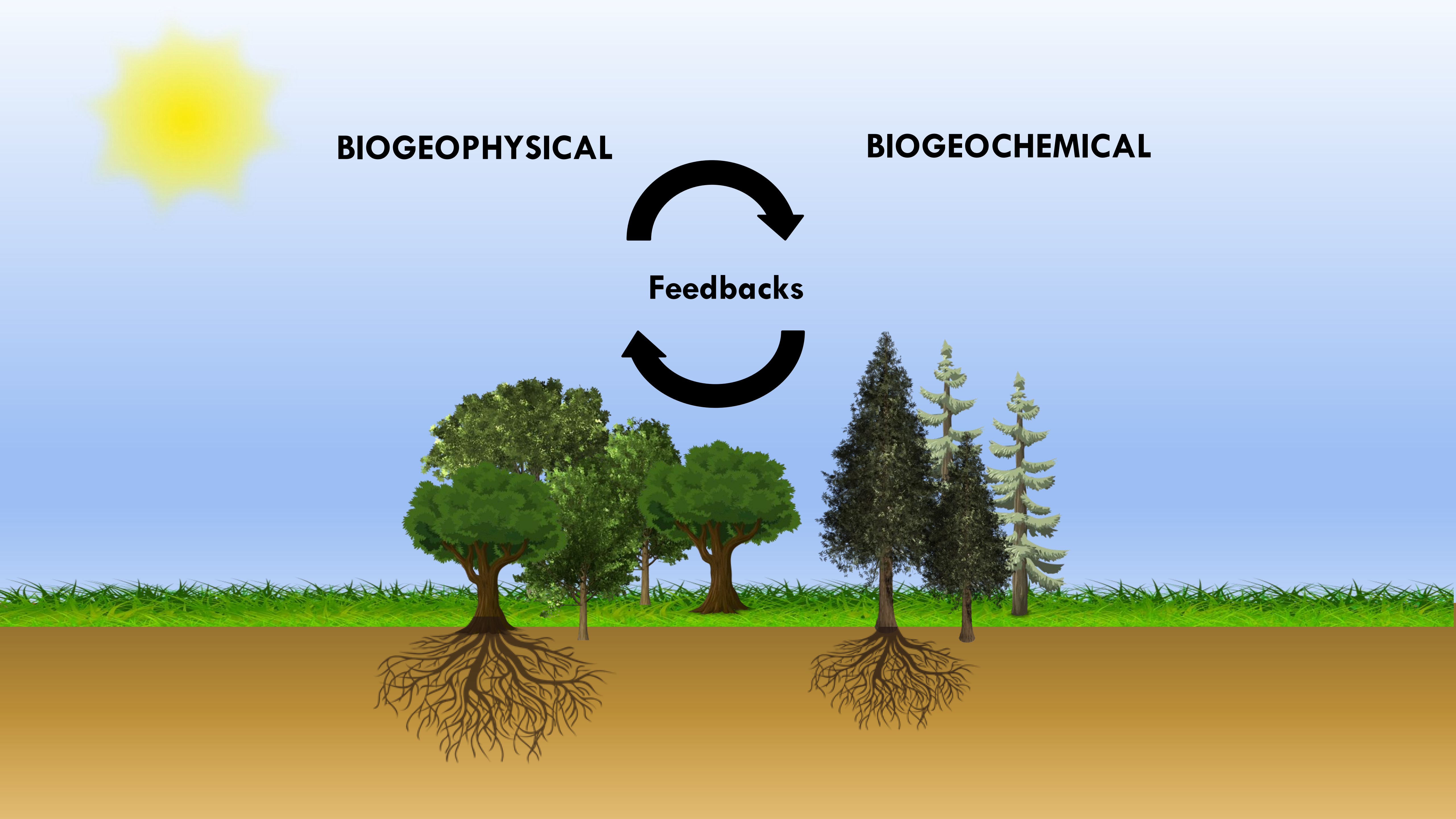
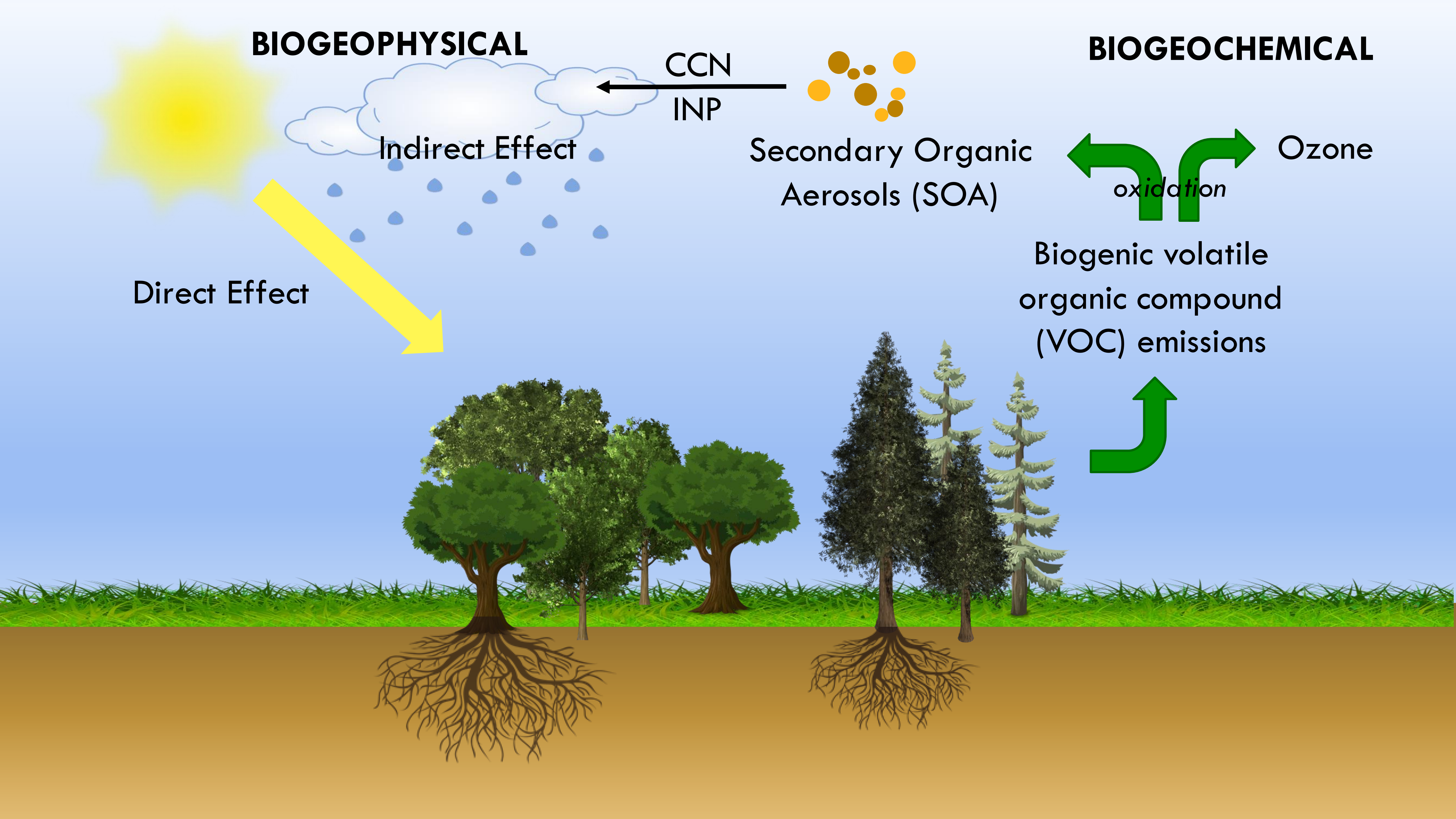


BIOGEOPHYSICAL
BIOGEOCHEMICAL
Feedbacks

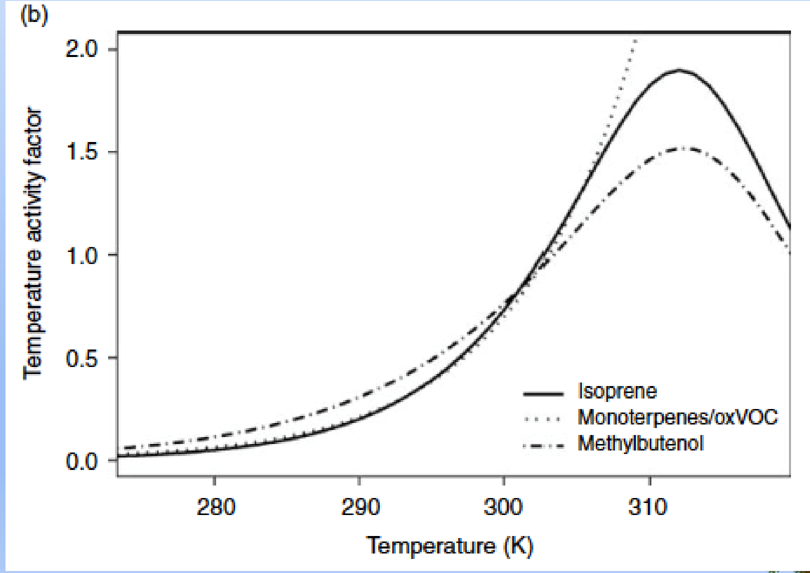

BIOGEOPHYSICAL
BIOGEOCHEMICAL
CCN
INP
Indirect Effect
Secondary Organic Aerosols (SOA)
Ozone
oxidation
Direct Effect
Biogenic volatile organic compound (VOC) emissions

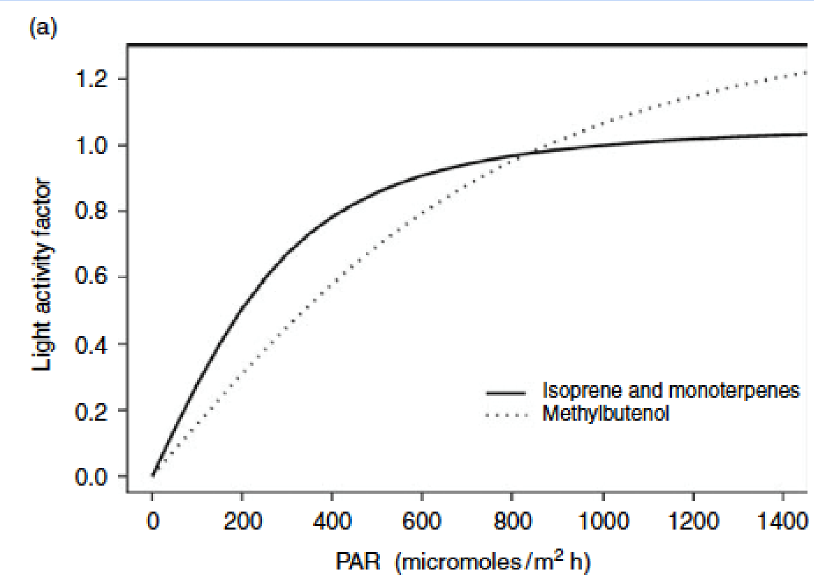
ENVIRONMENTAL DRIVERS OF BVOC EMISSIONS

TEMPERATURE

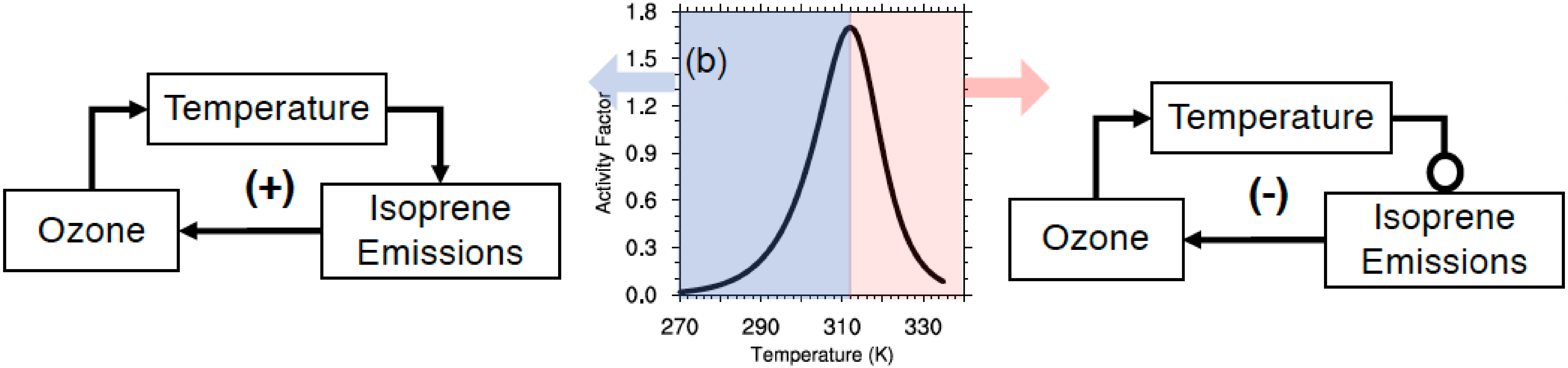
LIGHT

Feedbacks





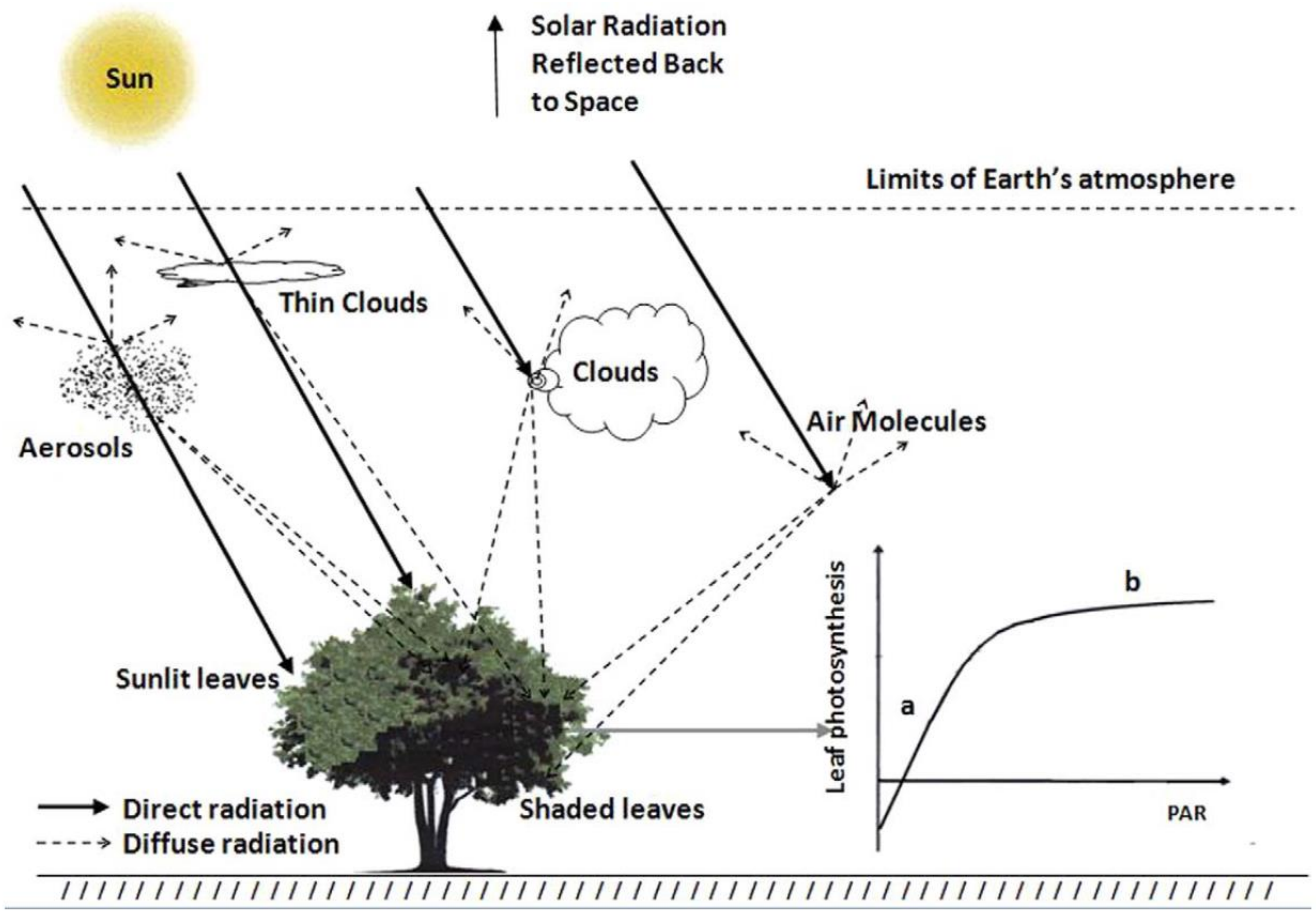
ISOPRENE FEEDBACKS WITH TEMPERATURE



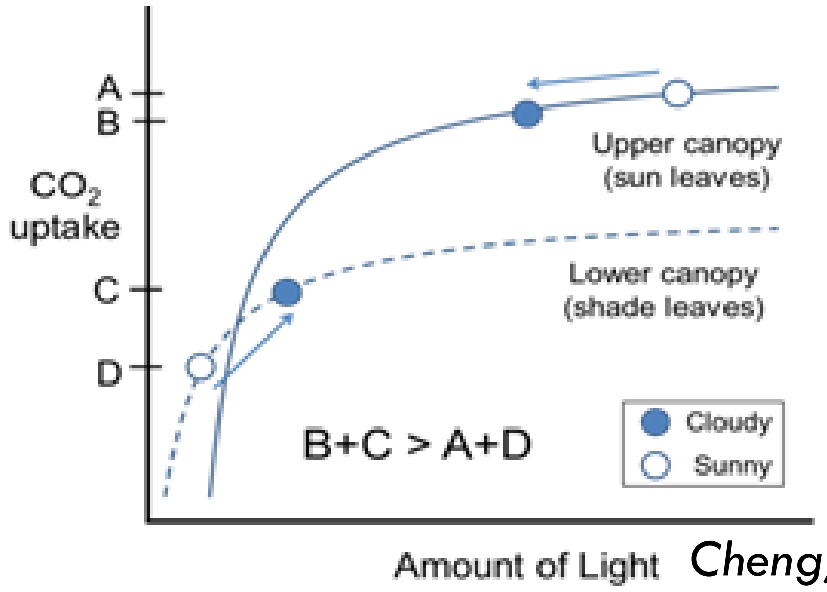
- Positive feedback loop when below the temperature peak
- Negative feedback loop when above the temperature peak
- How dynamic and responsive is the temperature maximum?

Steiner et al. 2010; PNAS
Steiner, 2020; Acc Chem Res

BIOGEOCHEMICAL FEEDBACKS: AEROSOL-CANOPY

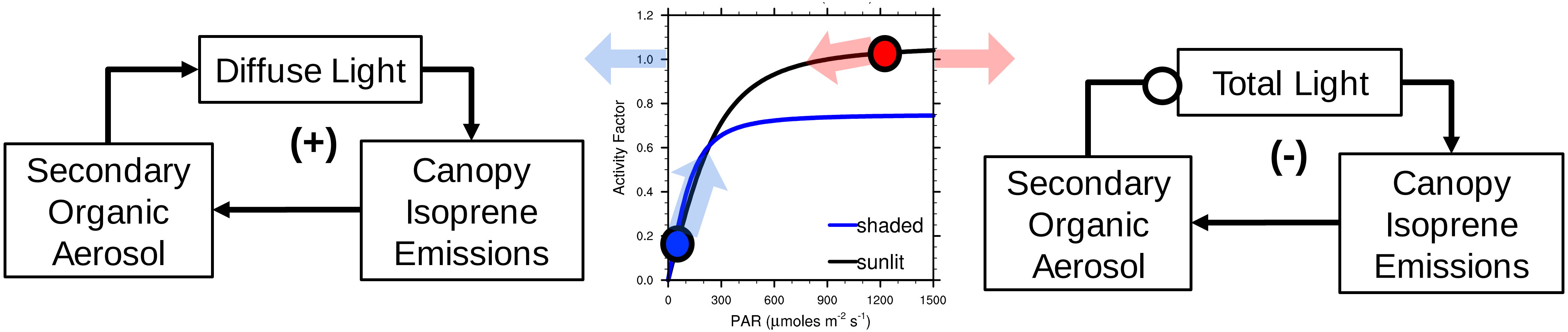


Kanniah et al., 2012



Cheng, 2016

ISOPRENE FEEDBACKS WITH DIFFUSE LIGHT

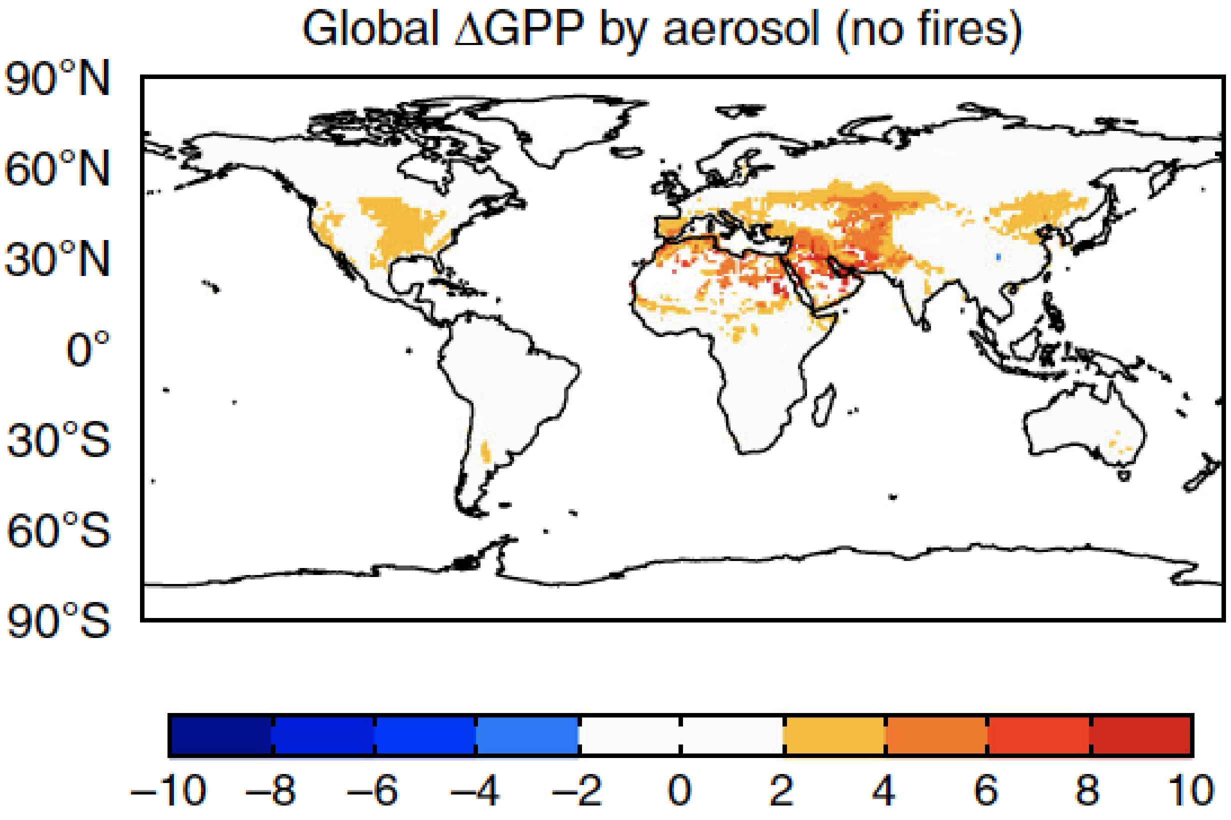


- Positive feedback loop for diffuse increases
- Negative feedback loop for total light reductions
- How does aerosol-generated diffuse light affect emissions and subsequent SOA formation?

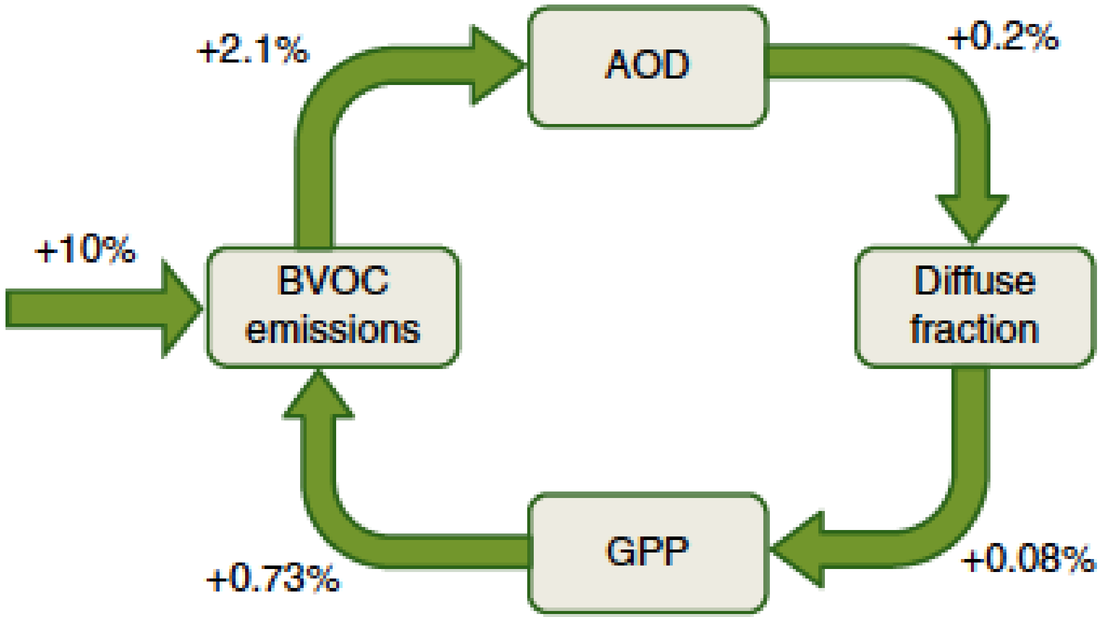
Steiner, 2020; Acc Chem Res

MODELING STUDIES PROMOTE THE IMPORTANCE OF THE DIFFUSE EFFECT

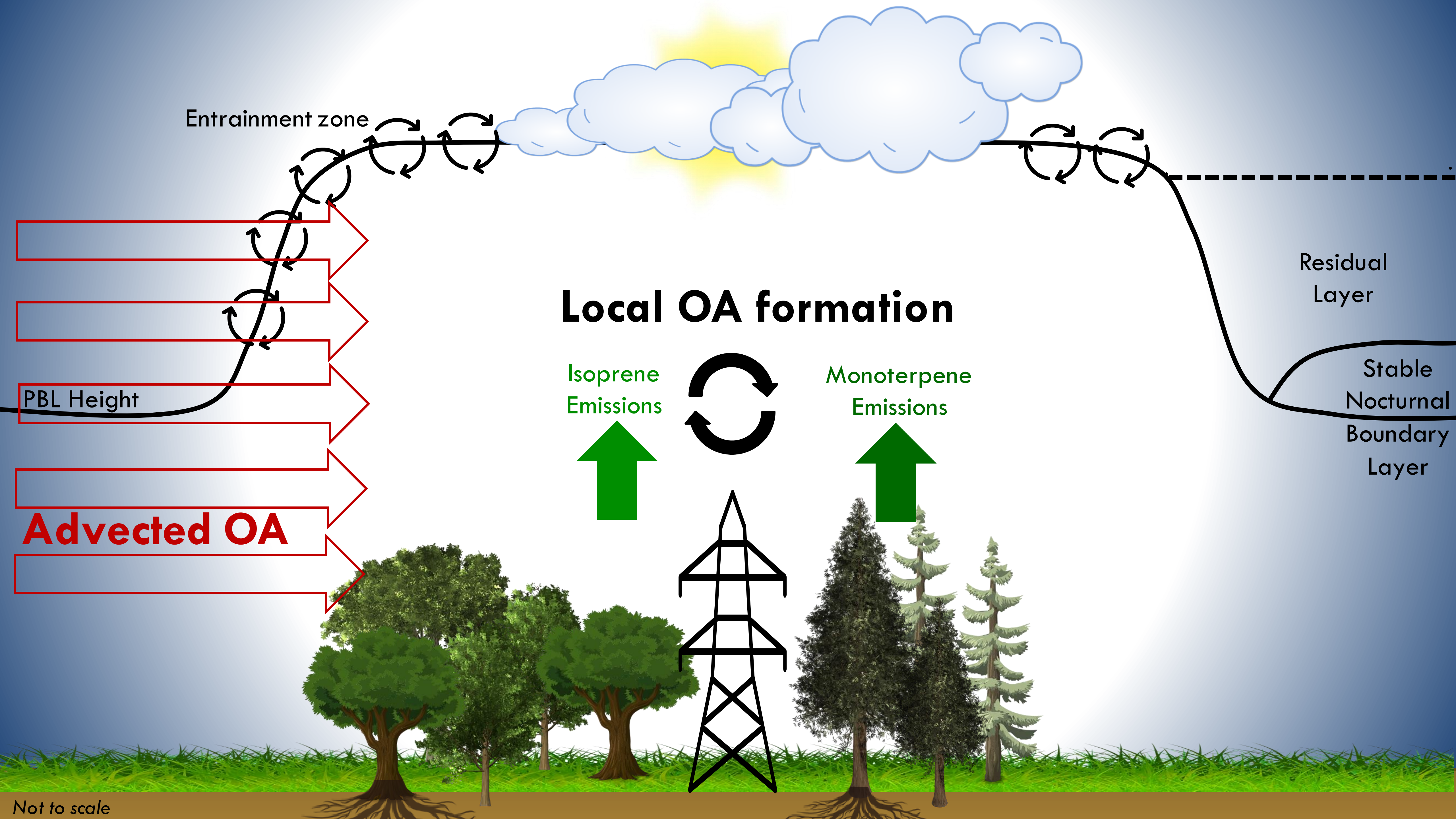
How can we verify these modeling studies with observations?



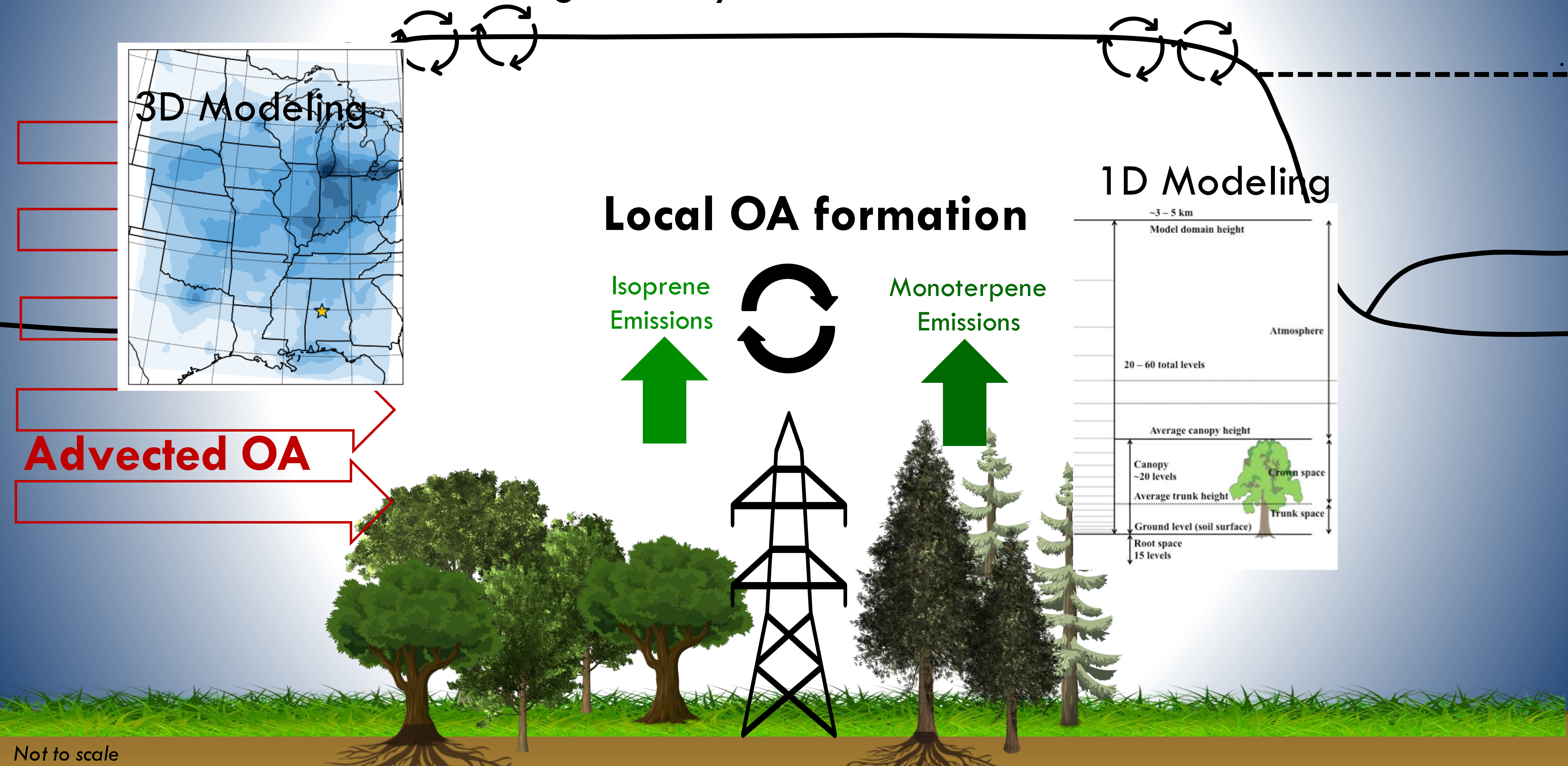
Yue and Unger, 2019: NatComms



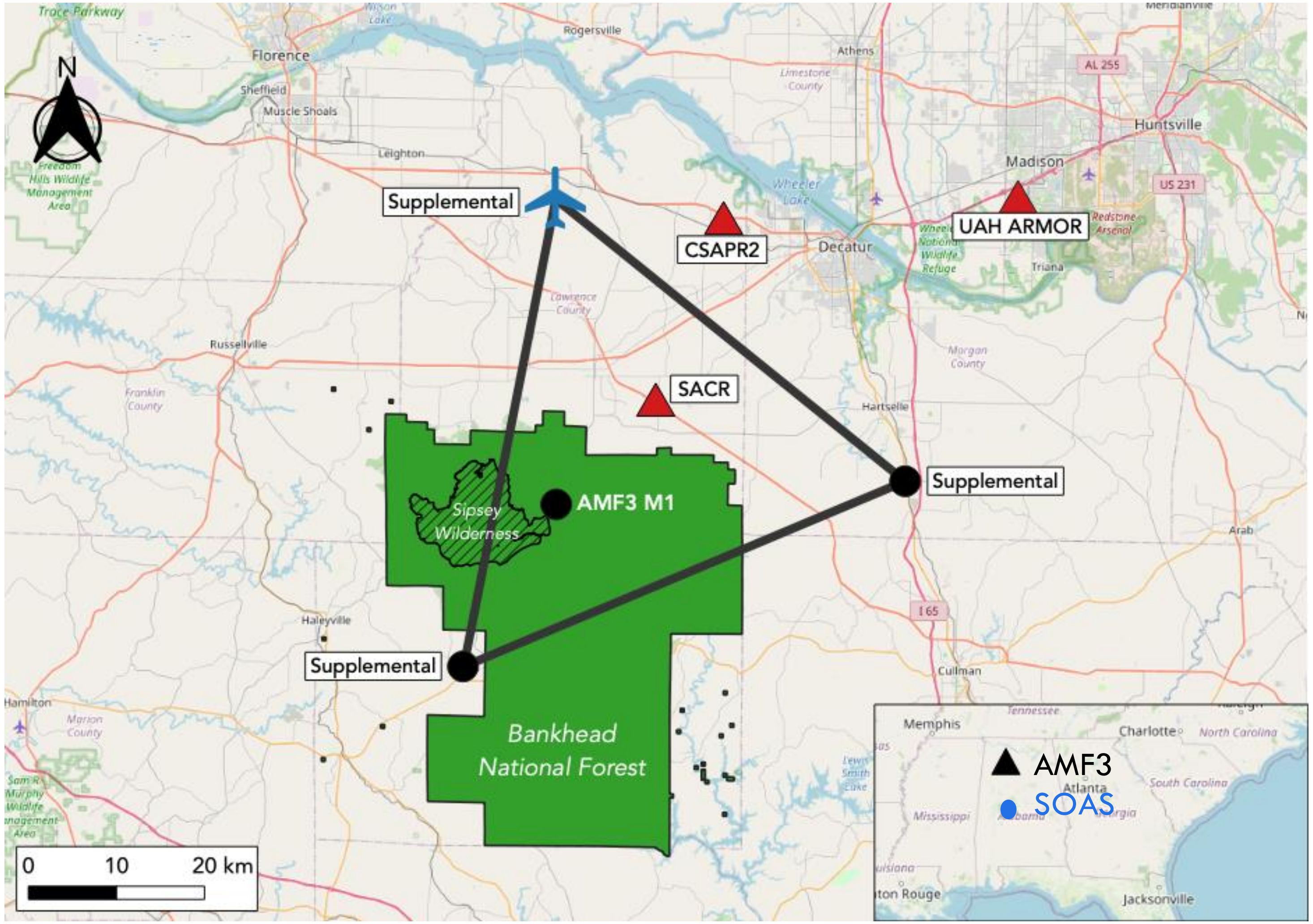
Rap et al. 2018: NatGeo


Entrainment zone
Residual Layer
Local OA formation
Isoprene Emissions
Monoterpene Emissions
PBL Height
Stable Nocturnal Boundary Layer
Advected OA
Not to scale

When is OA locally produced such that we can observe biogenically-driven feedbacks?



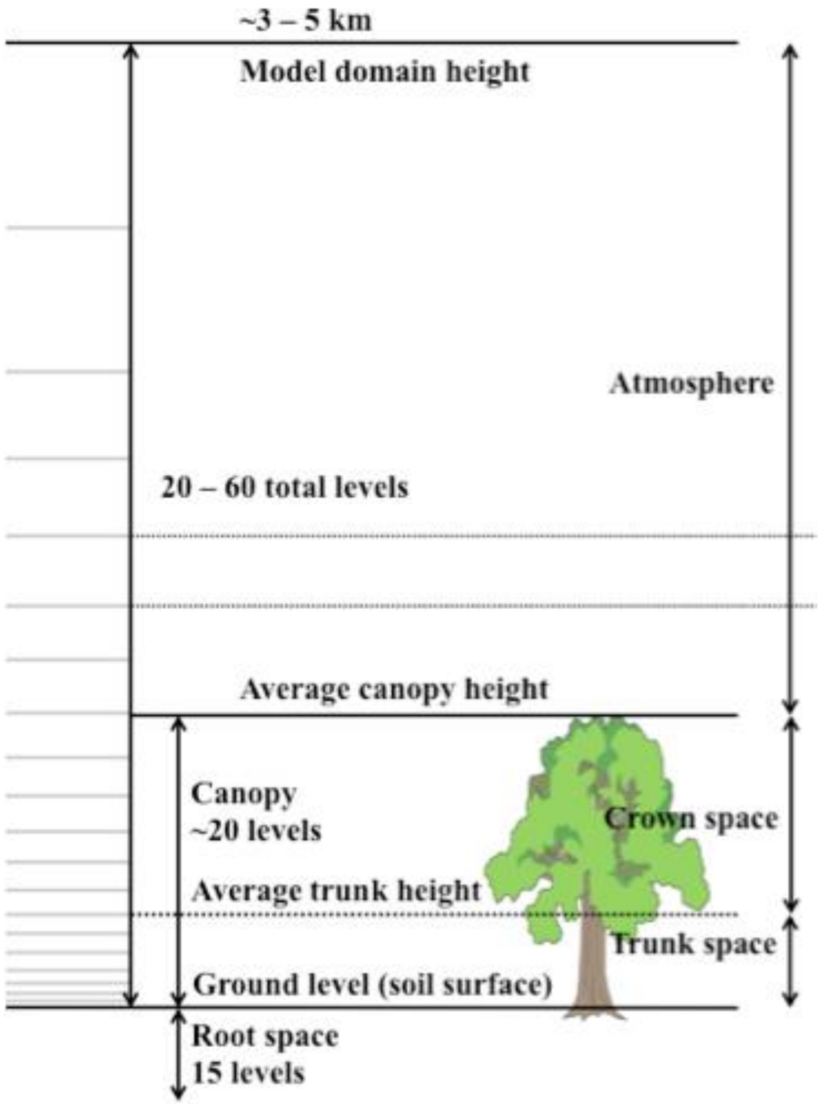
TEST CASE: SOUTHEASTERN UNITED STATES

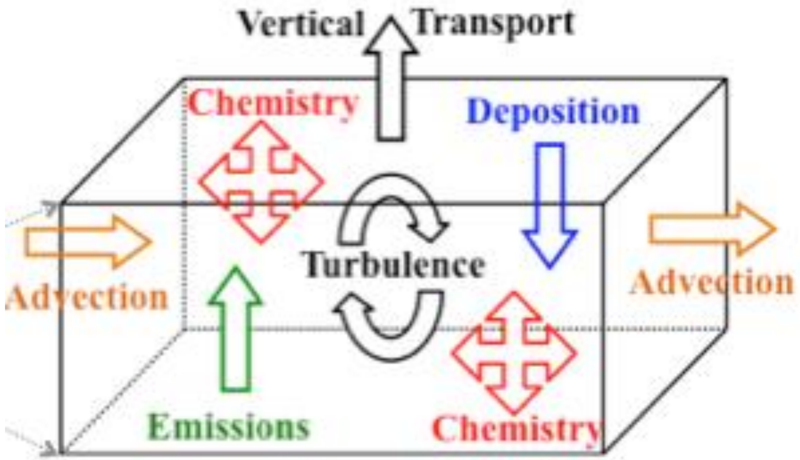


- New Department of Energy AMF3 Deployment in Southeastern United States: Bankhead National Forest to understand chemistry-climate-ecosystem feedbacks
- Also have observations from former field campaign Southern Oxidant and Aerosol Study (SOAS) in 2013 (Carlton et al. 2018)

SOAS Southern Oxidant & Aerosol Study

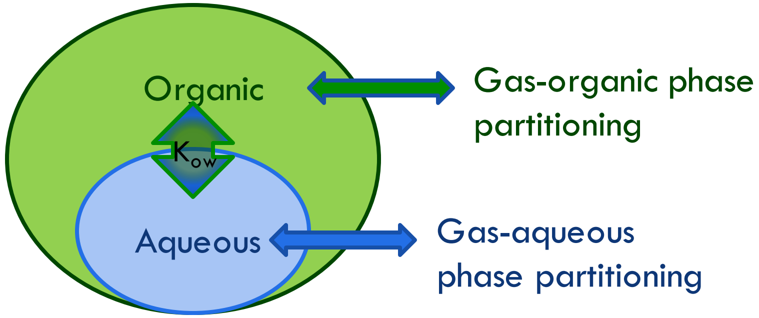
LOCAL PRODUCTION: FORCAsT: 1D CANOPY MODELING TOOL





$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial z}\left(K\frac{\partial c}{\partial z}\right) + E \pm C - D$$

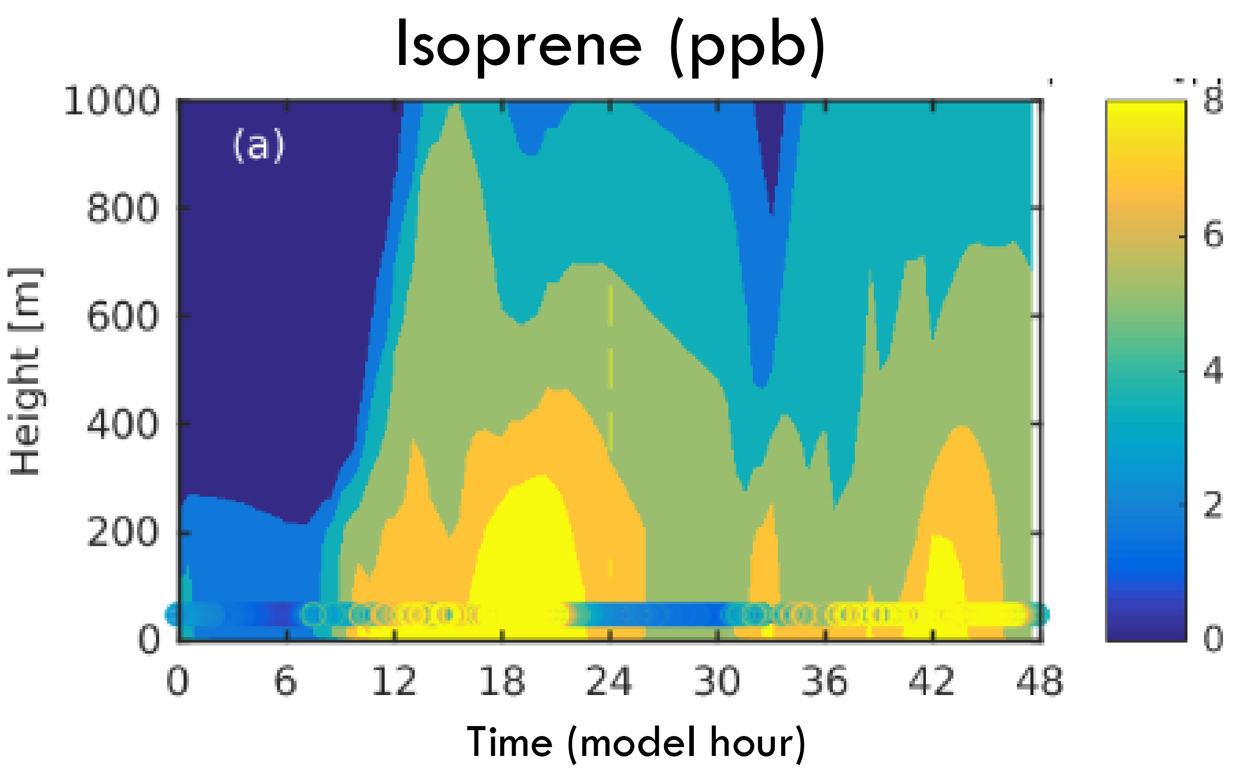
CACM + updated isoprene chemistry (RICM) and the MPMPO aerosol partitioning model

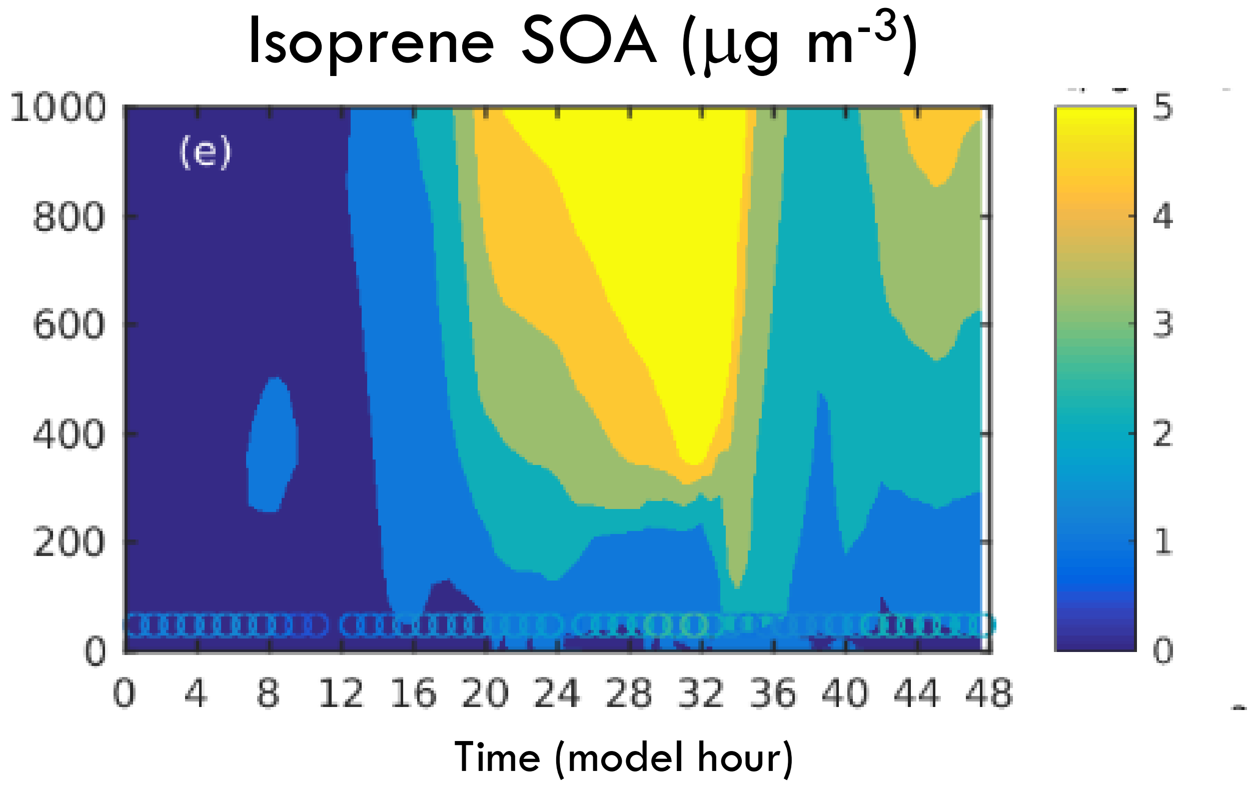


Bryan et al. 2012; ACP
Ashworth et al. 2016; GMD
Wei et al. 2022; GMD

1D SIMULATED VERTICAL GRADIENTS: ISOPRENE

- Mid-day peak in gas phase isoprene
- iSOA formation is dominantly aloft in the early AM and evening due to IEPOX and aerosol liquid water content

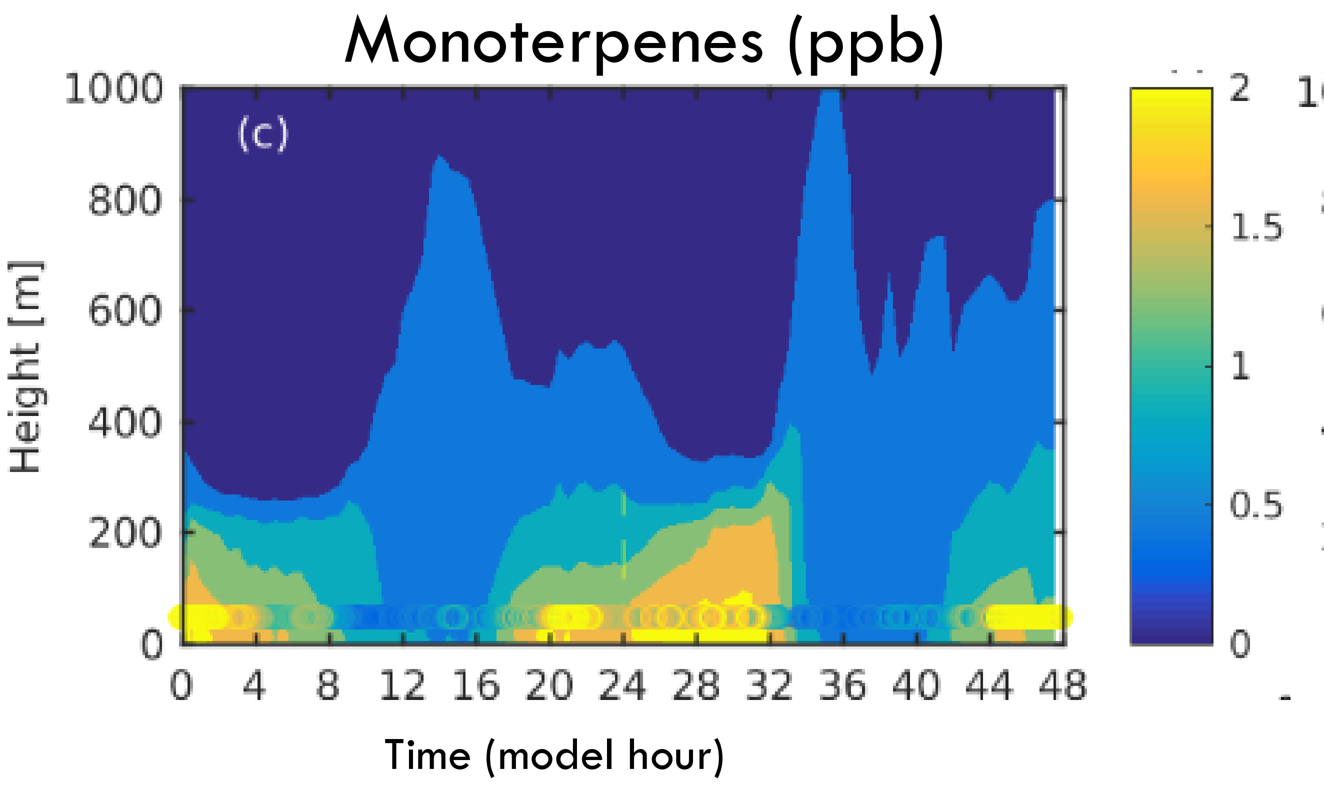


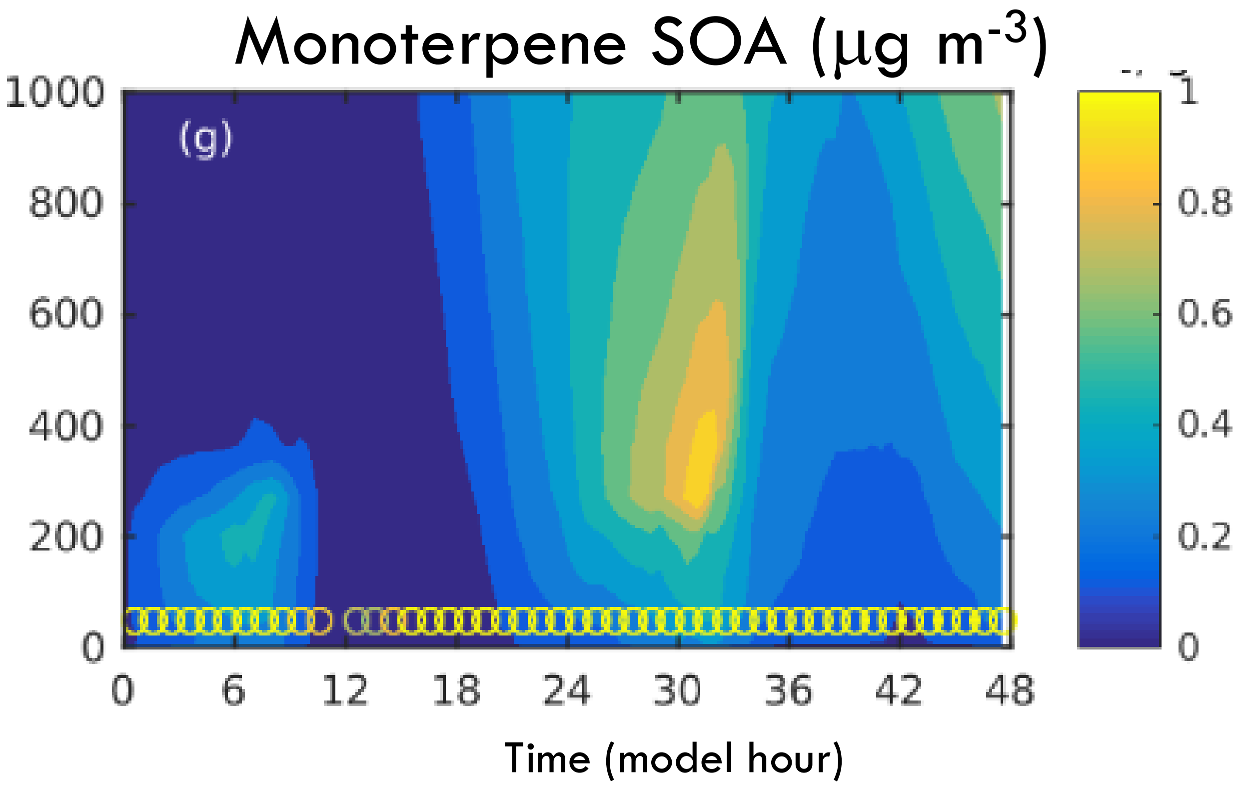


Wei et al., in preparation

1D SIMULATED VERTICAL GRADIENTS: MONOTERPENES

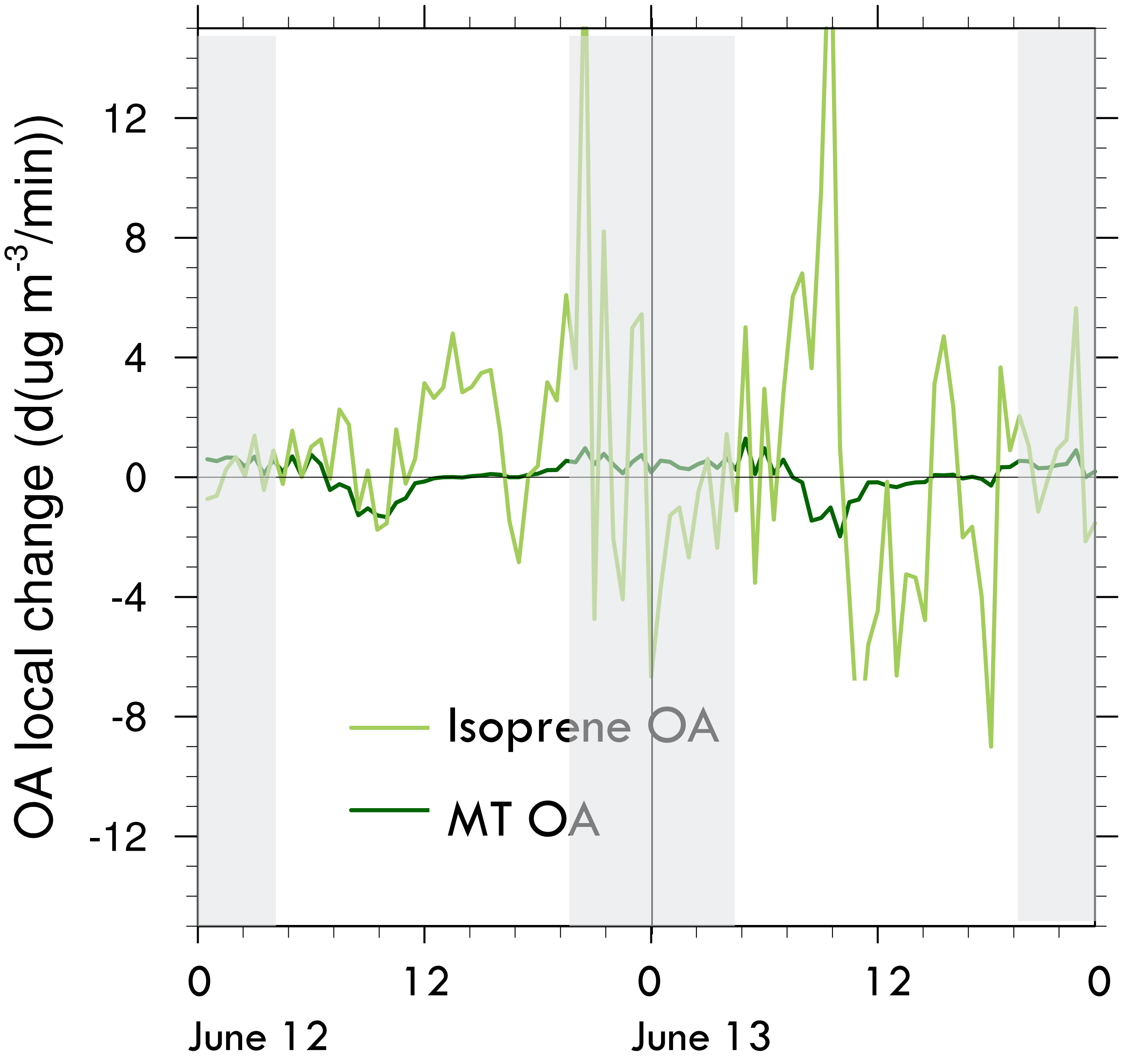
- MT nighttime concentration increases
- MT-SOA formation closer to canopy (but underestimated by model)





LOCAL PRODUCTION FROM 1D COLUMN MODEL

- Isoprene SOA local production rates reach up to 13 μg m^{-3} min^{-1}) and dominate over MT-SOA
- Local production generally peaks in the early AM/PM



Steiner et al., in preparation

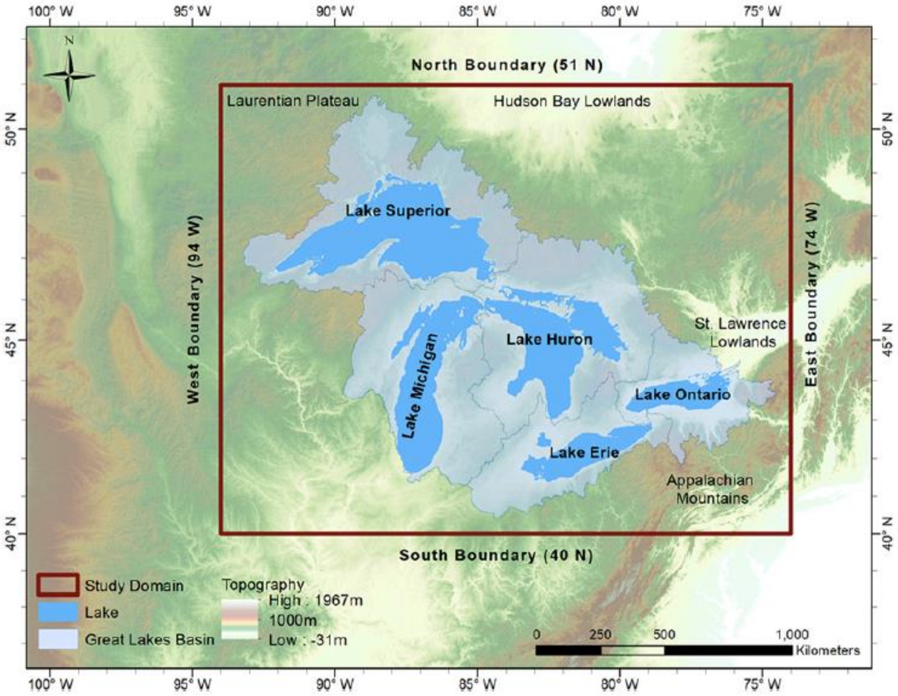
CALCULATING NON-LOCAL CONTRIBUTIONS

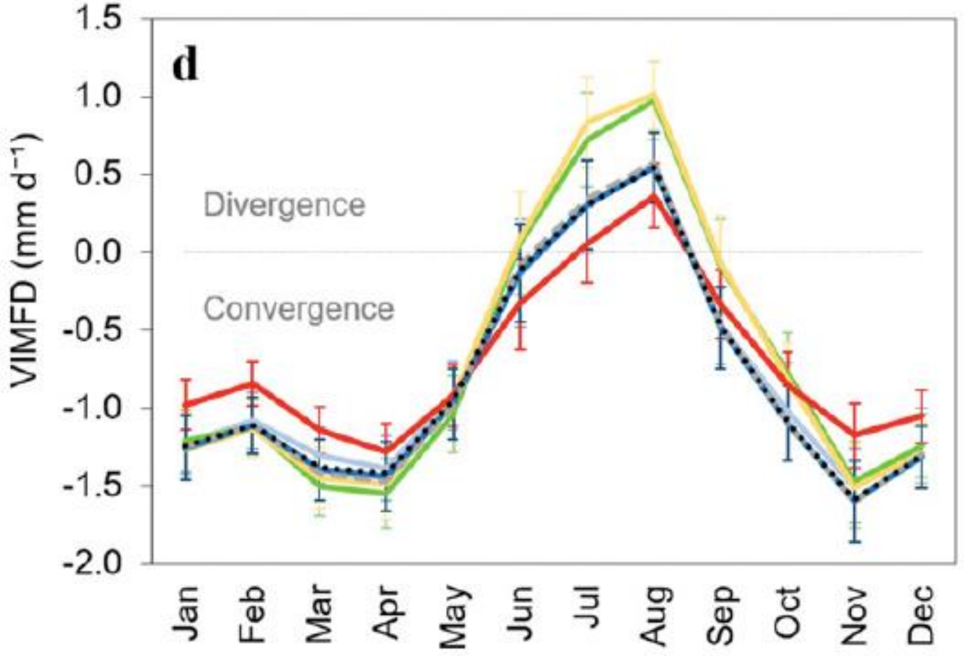
- Borrow the moisture budget analysis concept from the climate community

$$\iint_A \left[\nabla \cdot \int_{p_t}^{p_s} (q\mathbf{u})\, dp\right] dA = \int_l \left[\int_{p_t}^{p_s} (q\mathbf{u})\, dp \cdot \hat{n}\right] dl.$$

$$\text{VIMFD} = \frac{1}{\rho_w g A}\left(\text{north flux} - \text{south flux} + \text{east flux} - \text{west flux}\right)$$

Minallah and Steiner, 2021

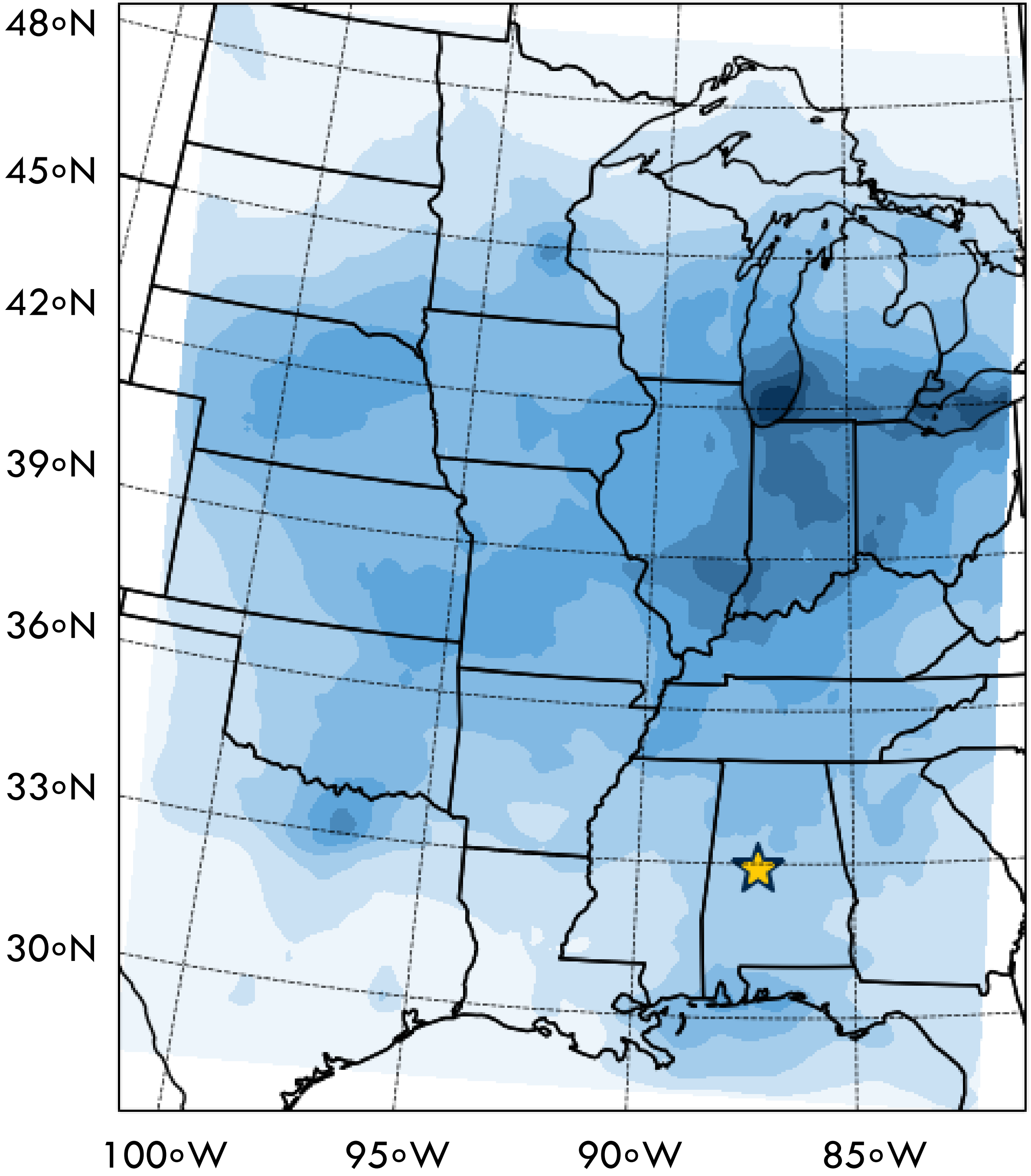




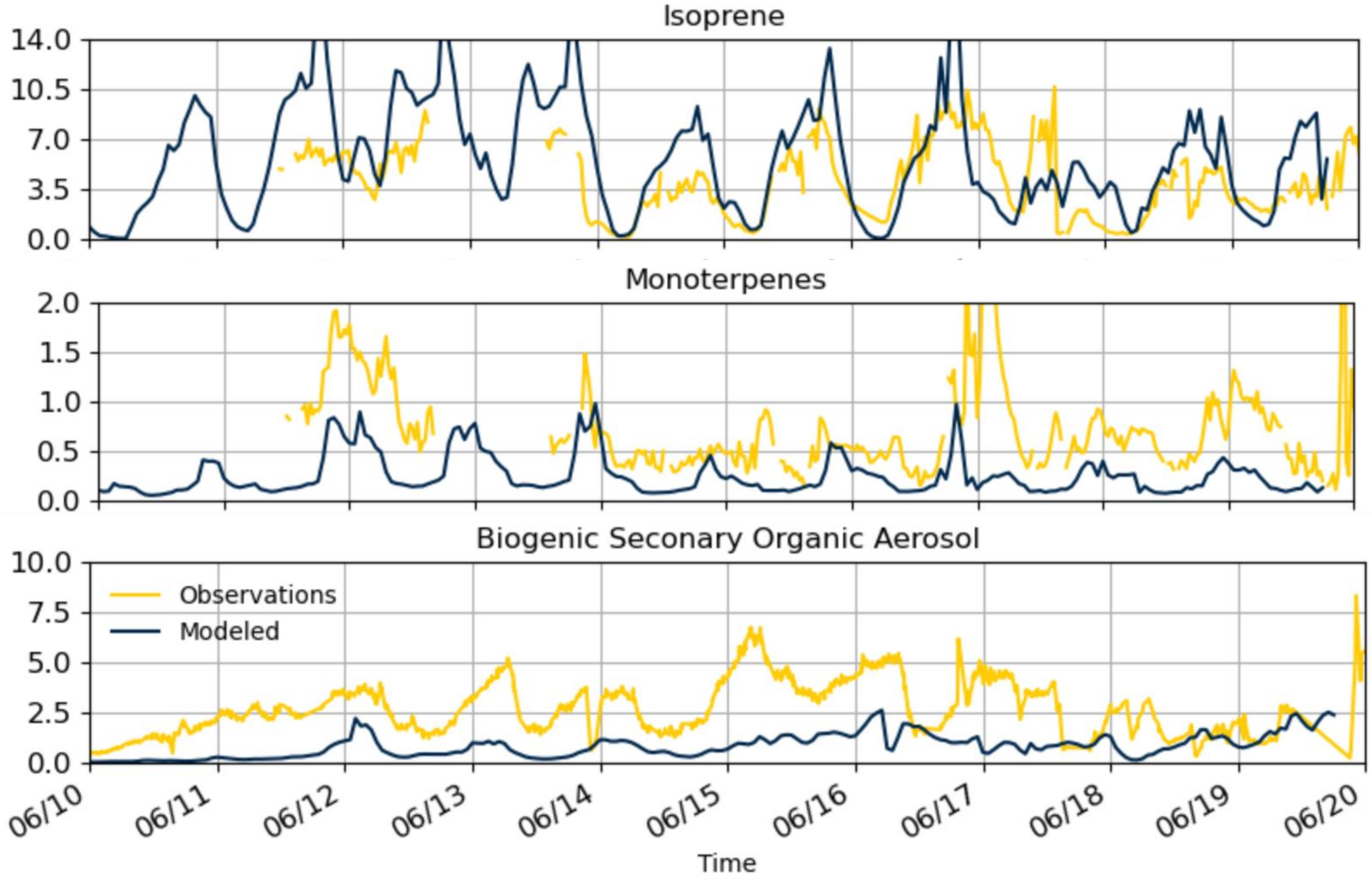
CALCULATING NON-LOCAL CONTRIBUTIONS FROM 3D MODELING

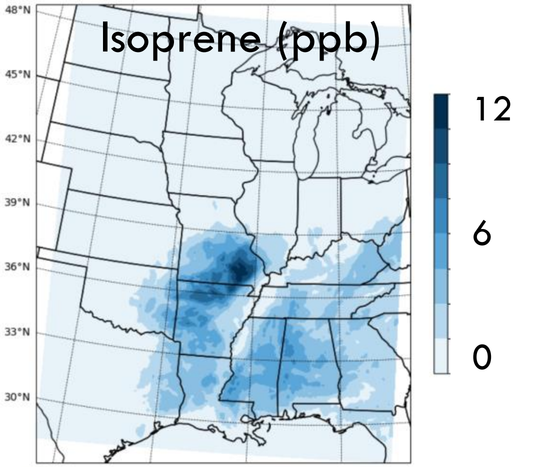
WRF-Chem simulations

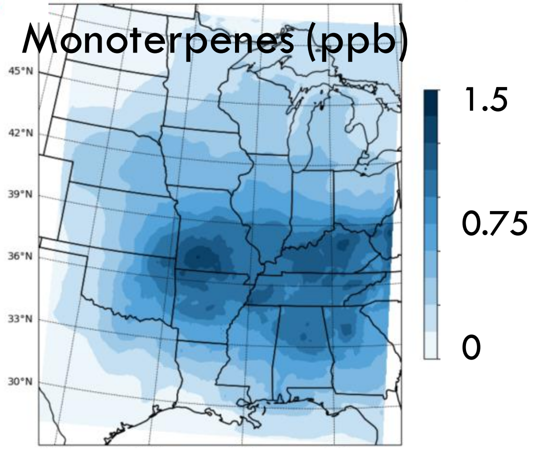
- 12 km resolution
- 1 month simulation in June 2013 (evaluate June 10-19
- MOZART gas phase mechanism with MOSAIC aerosol + VBS for OA

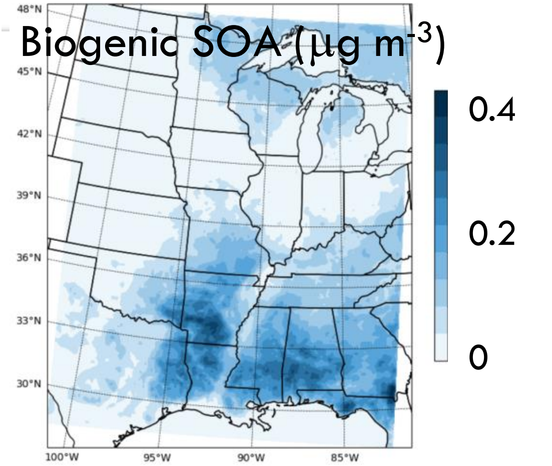


WRF-CHEM SIMULATIONS EVALUATION WITH SOAS OBSERVATIONS

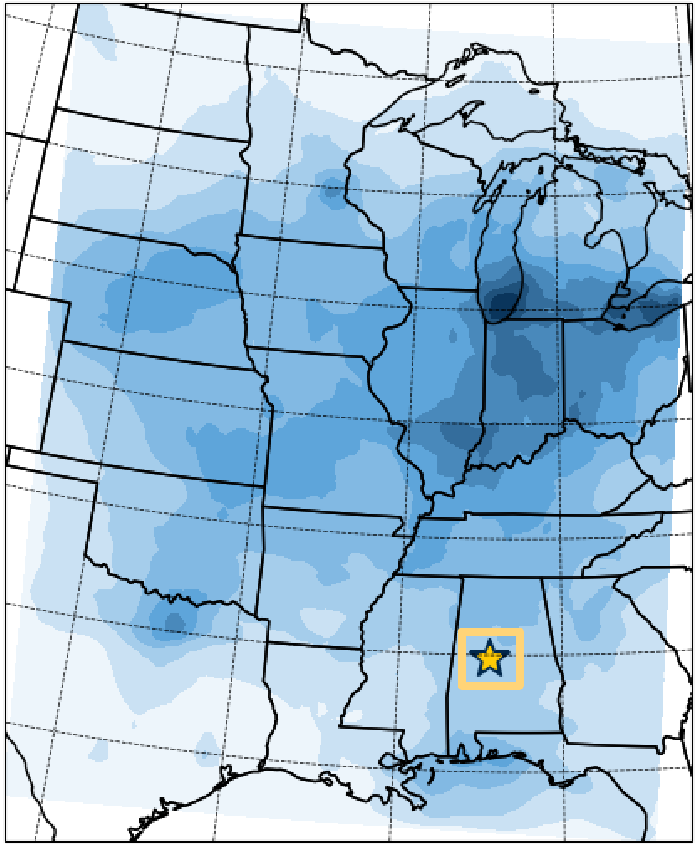




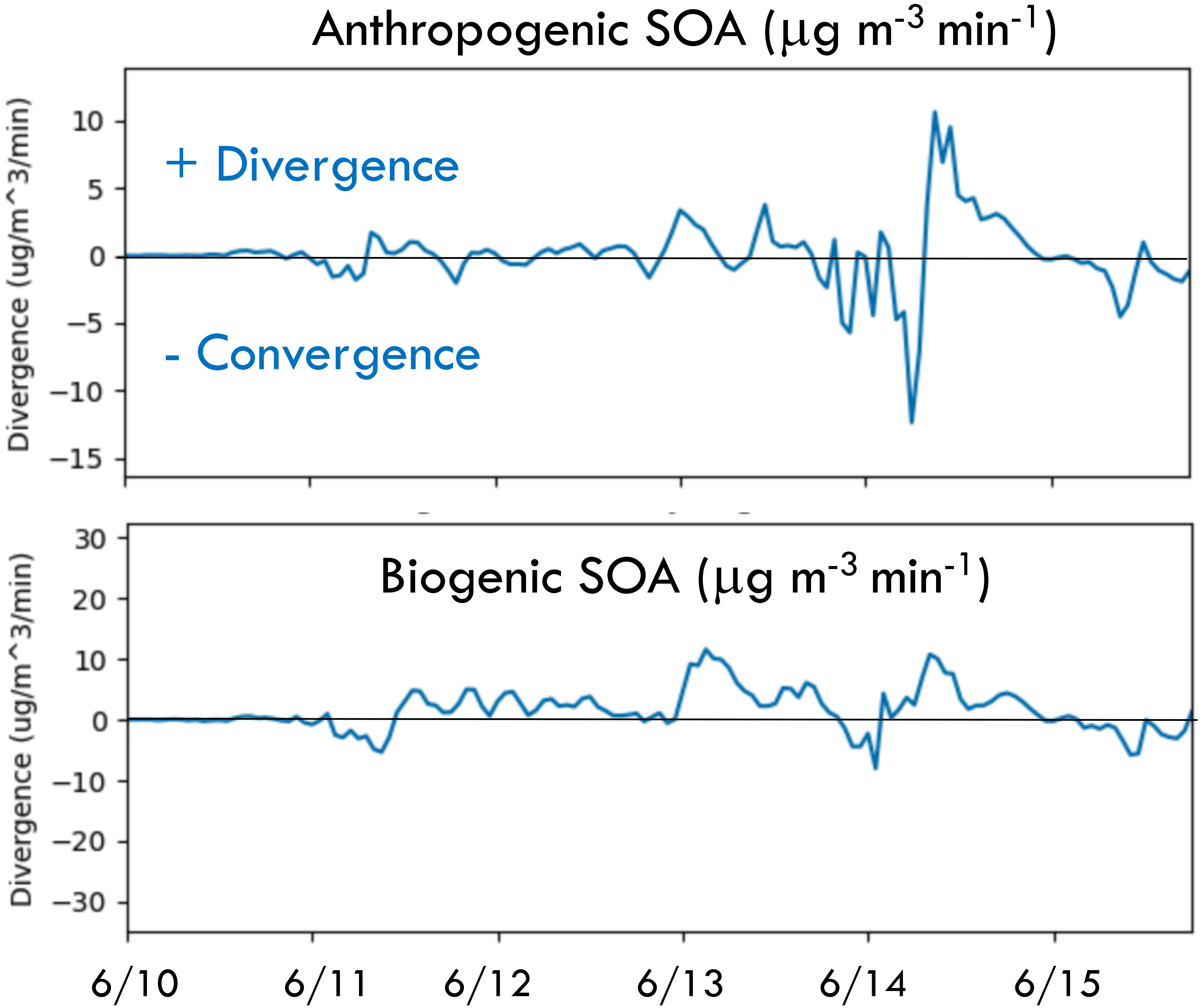




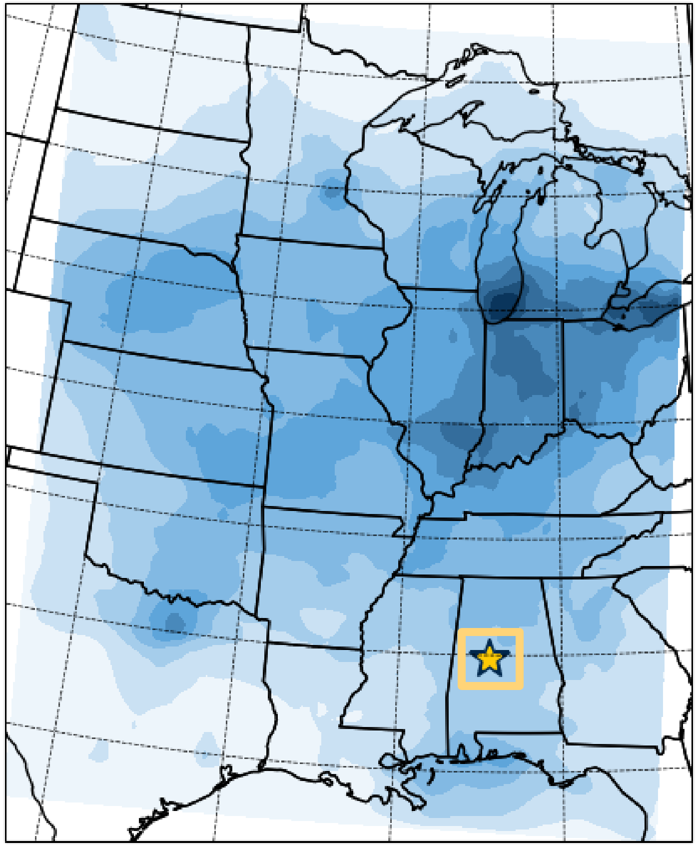
ADVECTIVE CONTRIBUTIONS TO SOA



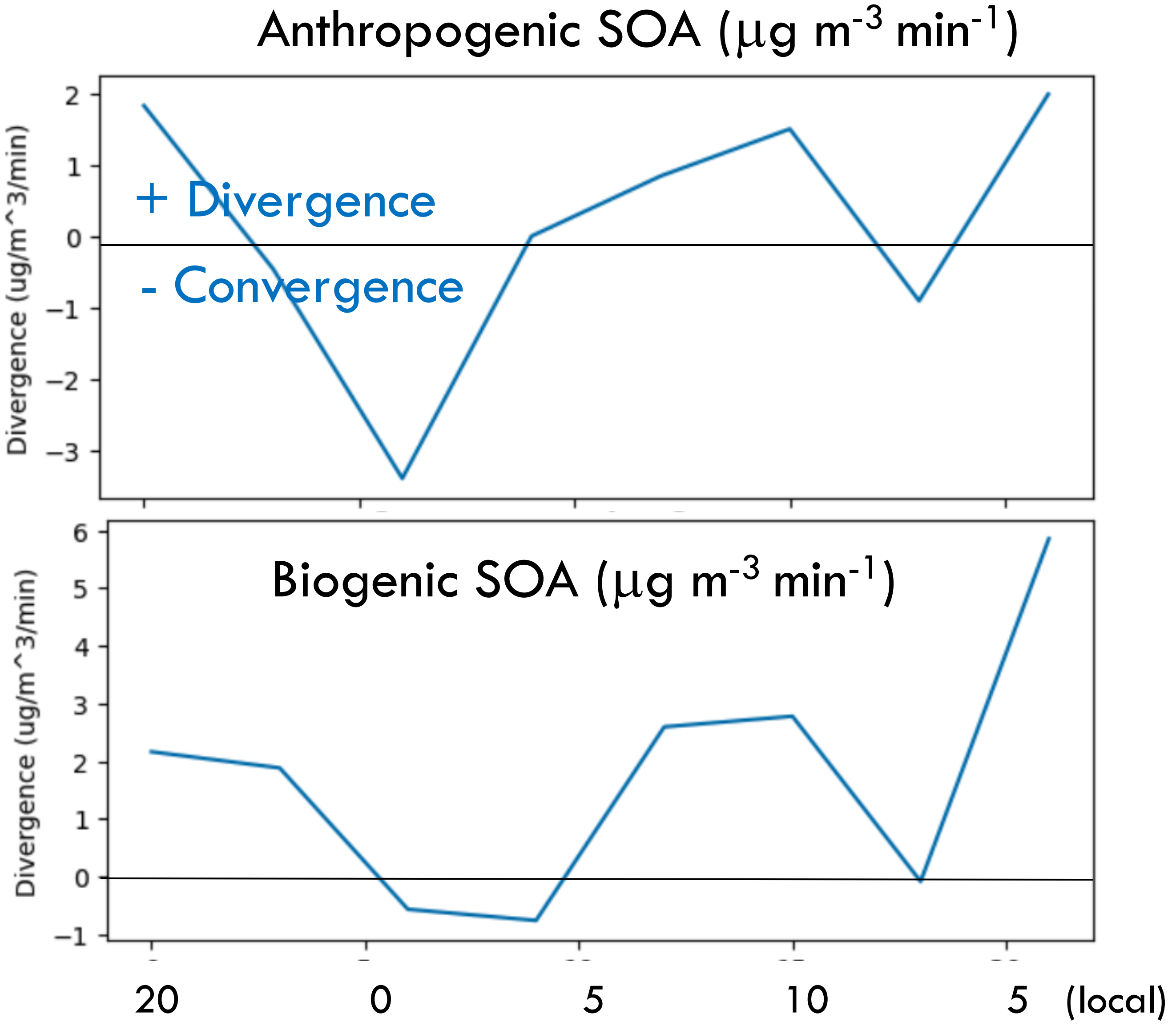
$$\iint_A \left[\nabla \cdot \int_{p_l}^{p_s} (c\mathbf{u})dp\right] dA = \int_l \left[\int_{p_t}^{p_s} (c\mathbf{u})dp \cdot \hat{n}\right] dl$$



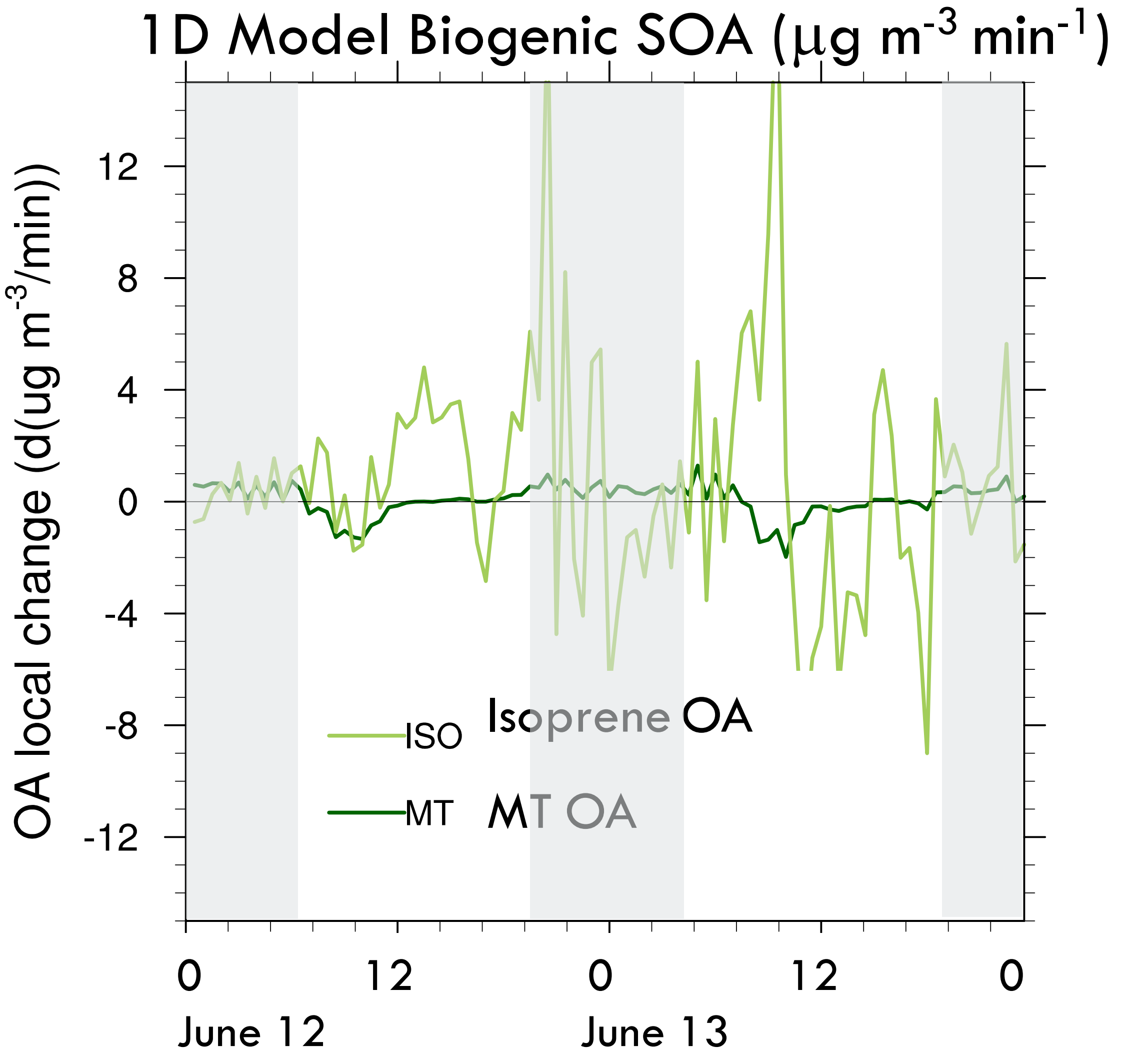
ADVECTIVE CONTRIBUTIONS TO SOA

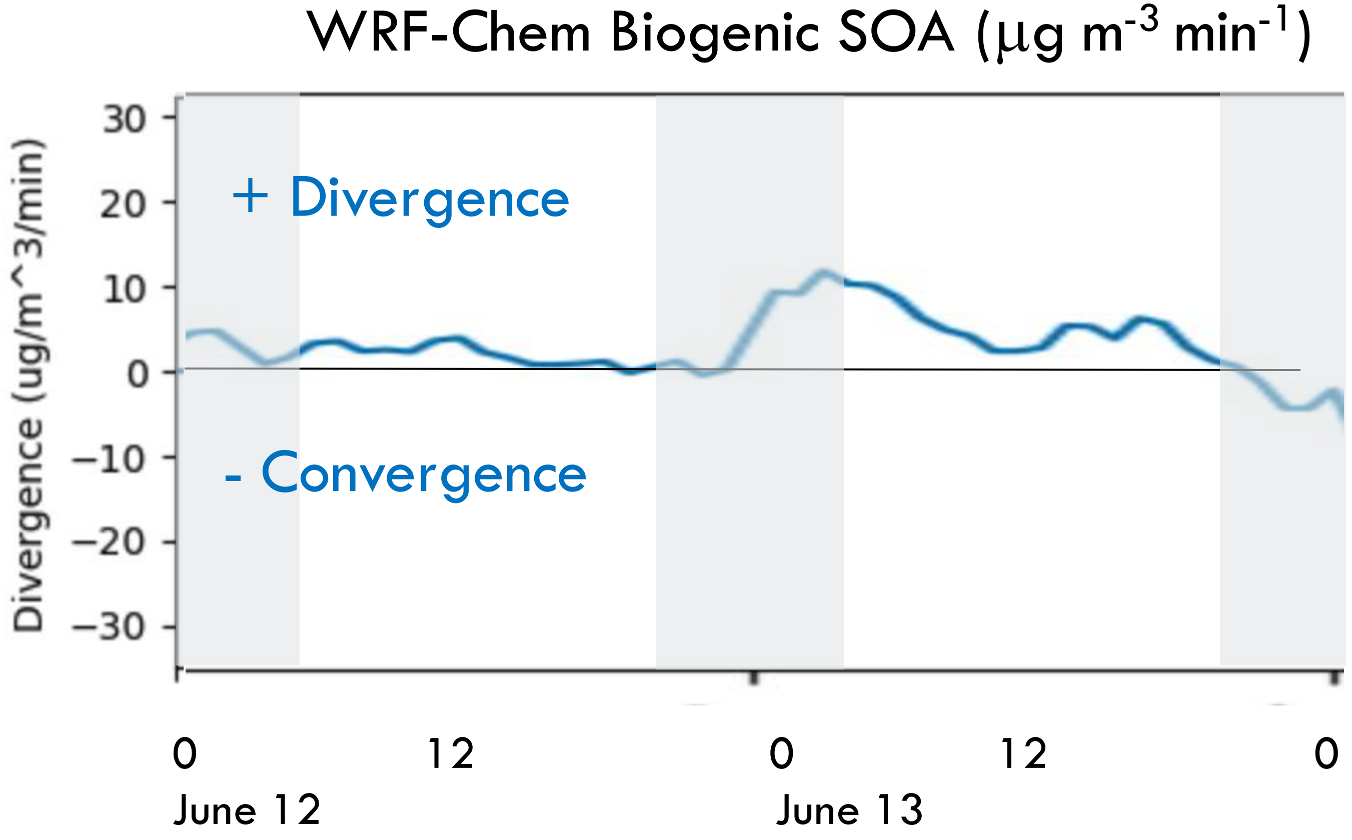


$$\iint_A \left[\nabla \cdot \int_{p_l}^{p_s} (\mathbf{cu})dp\right] dA = \int_l \left[\int_{p_t}^{p_s} (\mathbf{cu})dp \cdot \hat{n}\right] dl$$



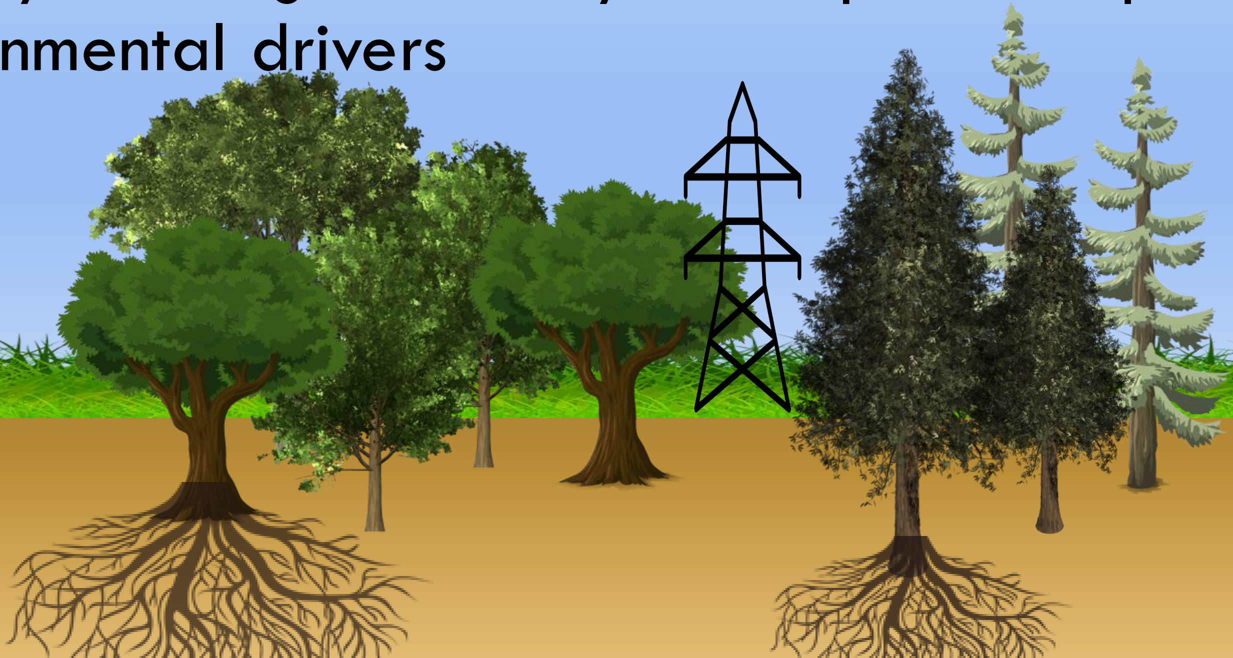
LOCAL VS. ADVECTIVE CONTRIBUTIONS TO BIOGENIC SOA





WHEN IS OA LOCALLY PRODUCED SUCH THAT WE CAN OBSERVE LOCAL BIOGENICALLY-DRIVEN FEEDBACKS?

- Combine modeling methods to highlight when local OA dominates
  - 1D model: local production only (highest for isoprene in early AM)
  - 3D model: encapsulates local + advective sources (generally divergence)
- At SOAS: early morning to midday for isoprene responses to fast time response environmental drivers



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