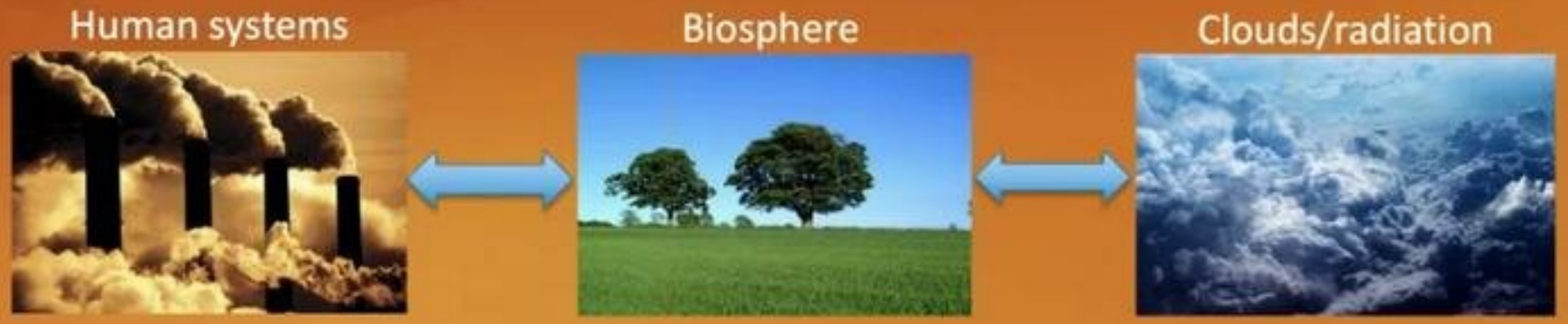
Development and evaluation of computationally efficient treatments of secondary organic aerosol (SOA) for E3SM

Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



Aerosol processes overlooked in climate models could greatly affect climate forcing

MANISH SHRIVASTAVA

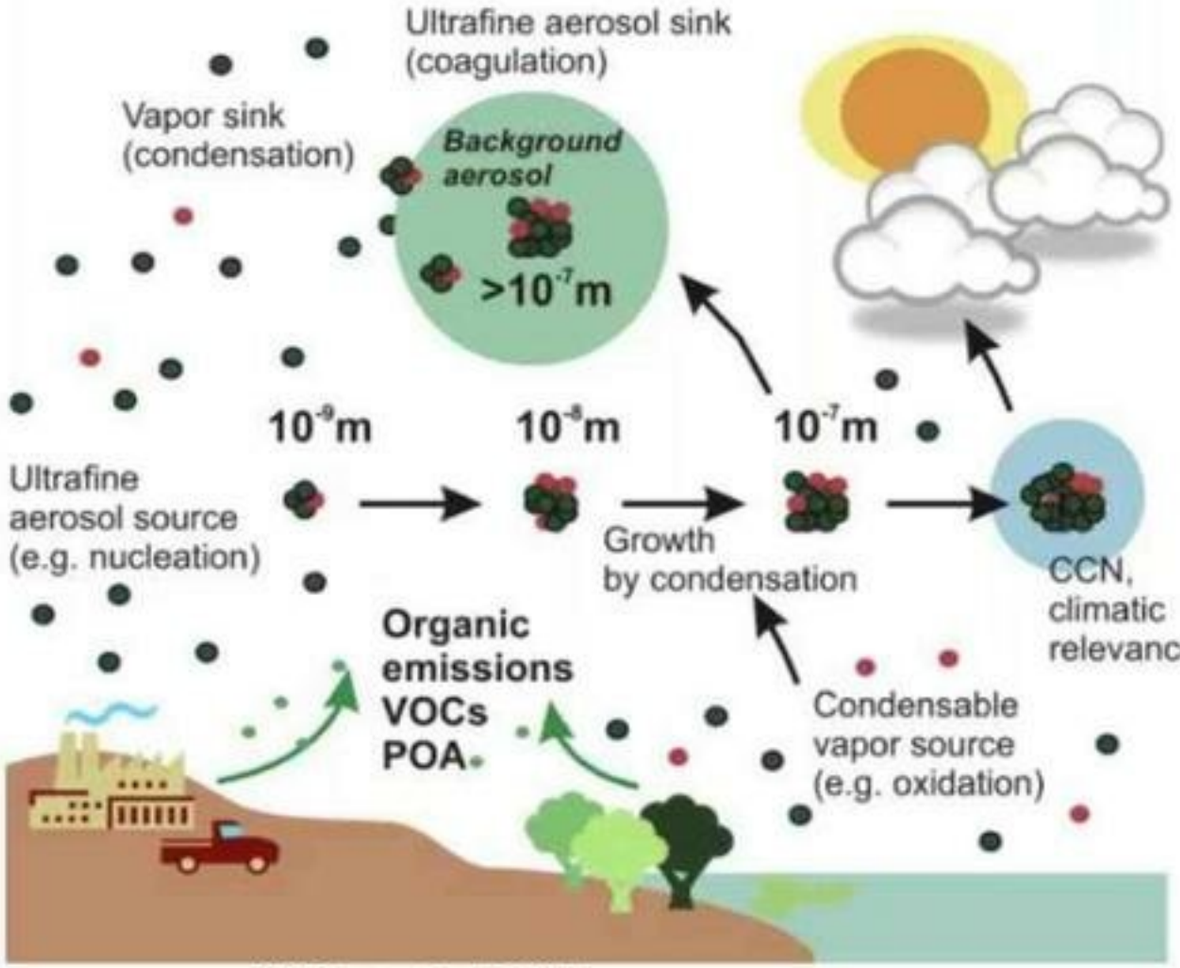
SIJIA LOU, RICHARD EASTER, HAILONG WANG, YANG YANG, PO-LUN MA, SCOT MARTIN, JOHN SHILLING, ALLA ZELENYUK, JOSE JIMENEZ, PEDRO CAMPUZANO JOST, QI ZHANG, PHILIP CAMERON SMITH, CHRISTIANE SCHULZ, JOHANNES SCHNEIDER, MANVENDRA DUBEY

LOU, SHRIVASTAVA ET AL. 2020, JOURNAL FOR ADVANCES IN MODELING EARTH SYSTEMS

Acknowledgements: U.S. Department of Energy's E3SM Atmospheric Physics NGD and my DOE Early Career Award (2018)

What are secondary organic aerosols (SOA)?

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

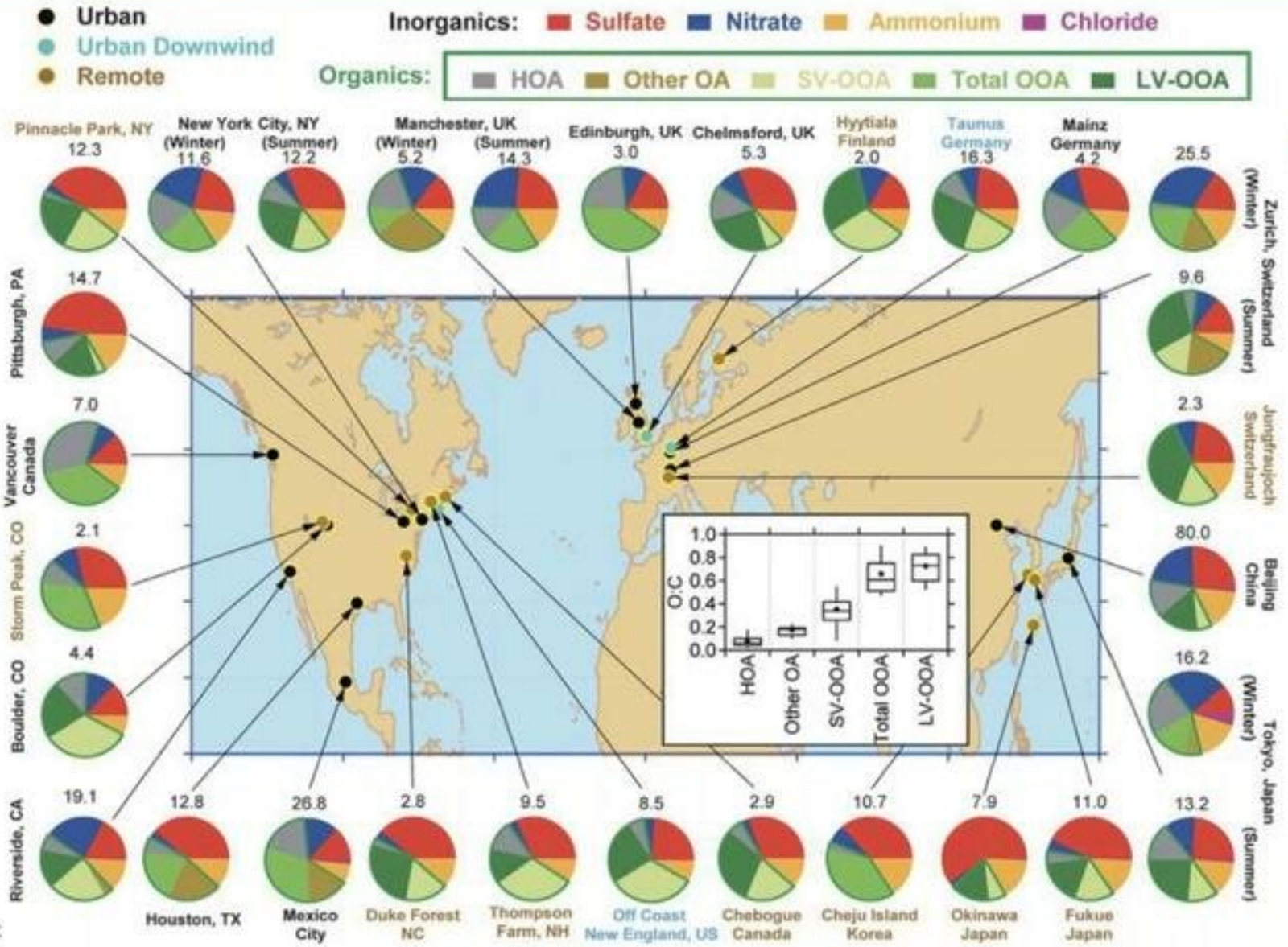


Riipinen et al. 2011

- Primary organic aerosol (POA): Directly emitted
- Secondary organic aerosol (SOA): Formed by chemical reactions of volatile organic compounds (VOCs) and gas to particle conversion of products
- Each SOA particle is made of thousands of organic compounds
- SOA is ubiquitous in the atmosphere
- SOA is a vital link connecting natural, anthropogenic and wildfire emissions to aerosols, radiation and clouds

Aerosol Mass Spectrometer (AMS) measurements around the world: SOA (green) is ubiquitous

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

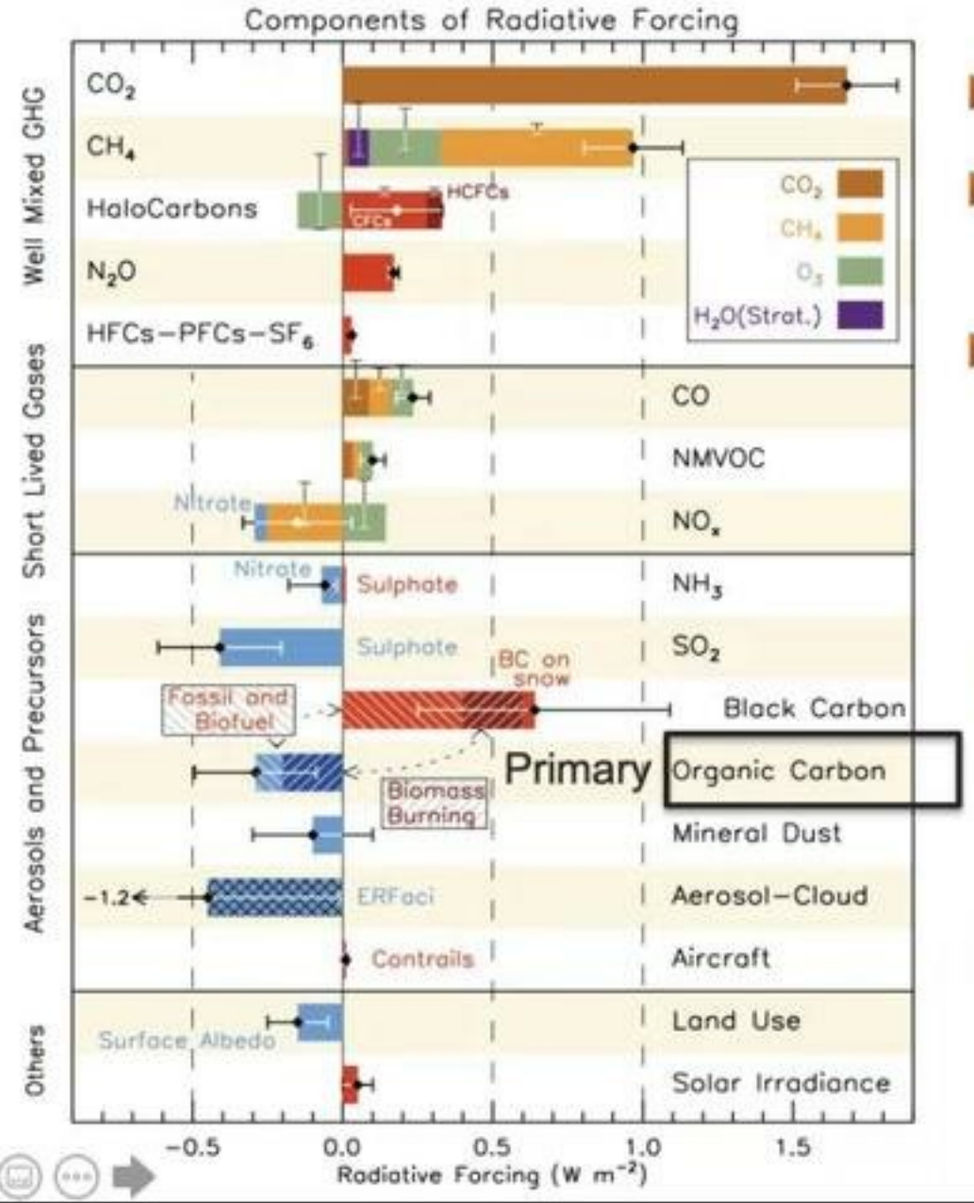


- In the Northern Hemisphere SOA concentrations are similar to or greater than sulfate

Jimenez et al. 2009, Science

IPCC Radiative forcing bar chart: Does not include SOA

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



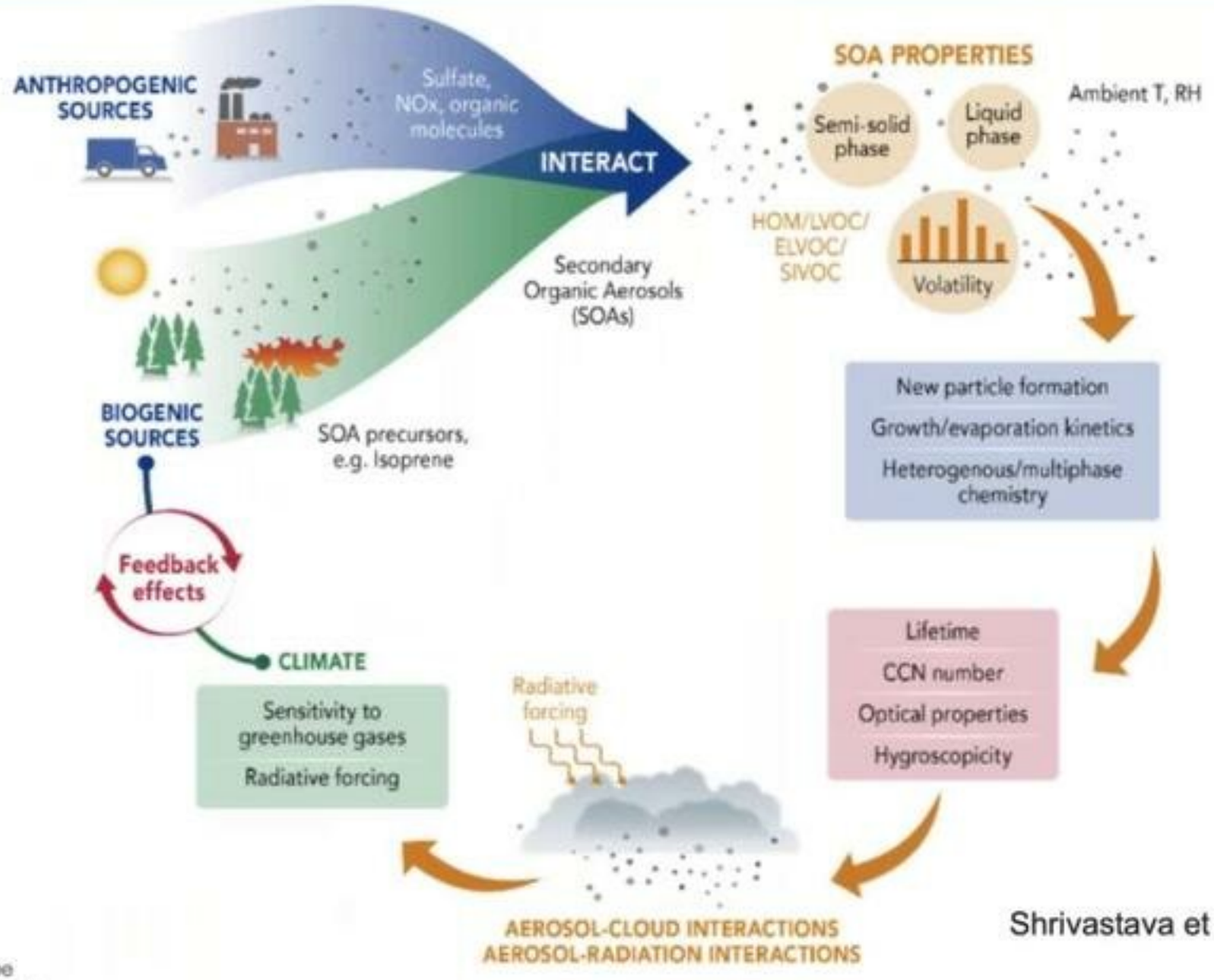
- Organic carbon (OC) radiative forcing is -0.3 Wm^{-2}
- SOA not included because formation depends on variety of uncertain factors
- There is tremendous complexity and uncertainty in the processes involved in SOA formation (IPCC, 2013)

  Myhre et al. 2013, IPCC Fifth Assessment

- SOA could change global cloud forcing by ~20% with large regional variations

Anthropogenic emissions interact with biogenic VOCs through atmospheric chemistry making SOA

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



Anthropogenic and biogenic emissions interact through a variety of pathways that affect SOA, and its impacts on clouds and radiative forcing.

Global Earth System Models do not include most of these SOA formation pathways

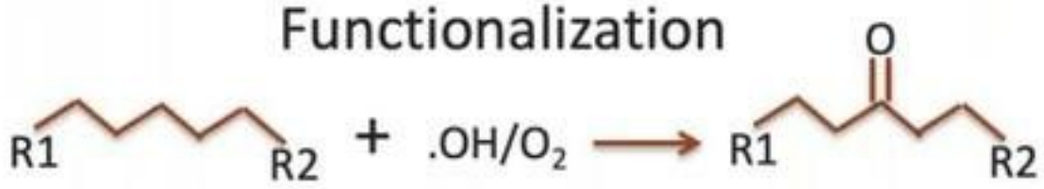
This could bias the estimated PD-PI radiative forcing

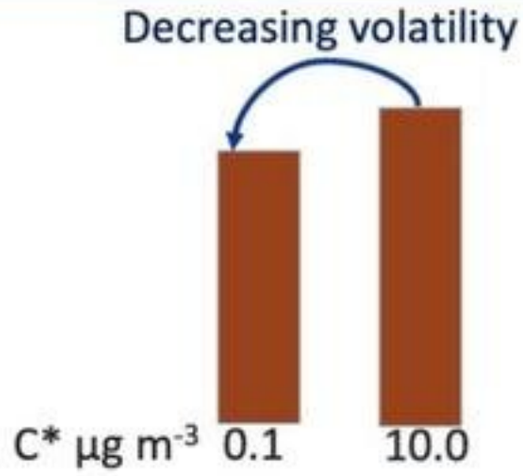
Shrivastava et al. 2017, Reviews of Geophysics

Chemical oxidation reactions can be both sources and sinks : Adding functional groups, or fragmenting carbon backbone

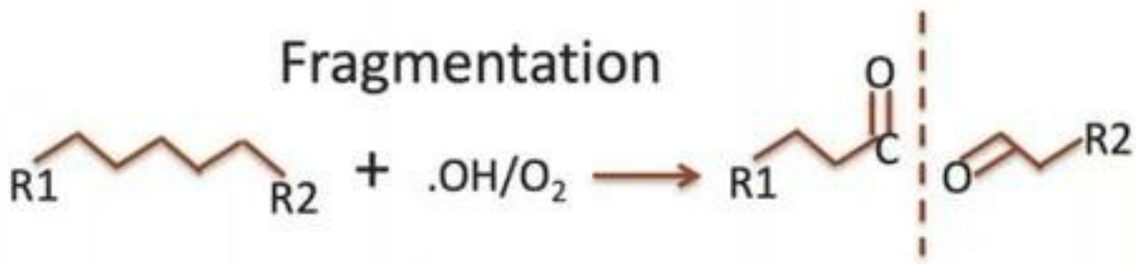
Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

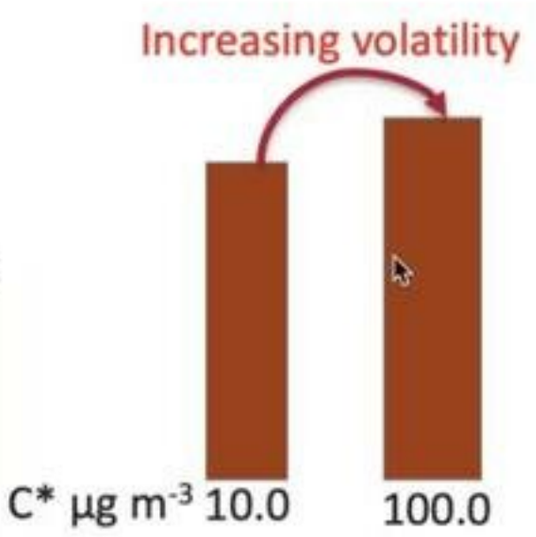
Functionalization





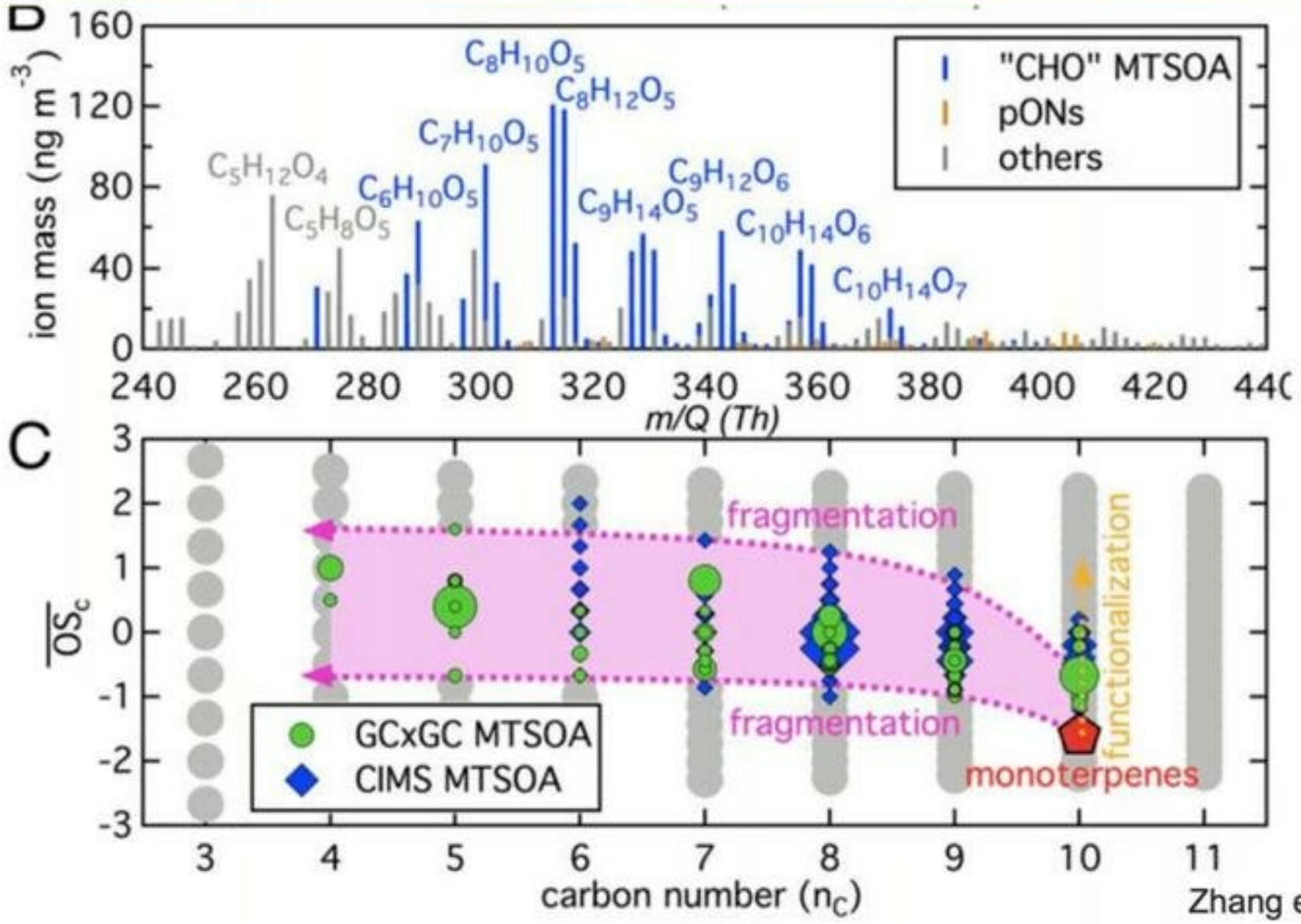
Fragmentation





Southeast USA field measurements show monoterpene fragmentation products increase with aging in presence of NOx

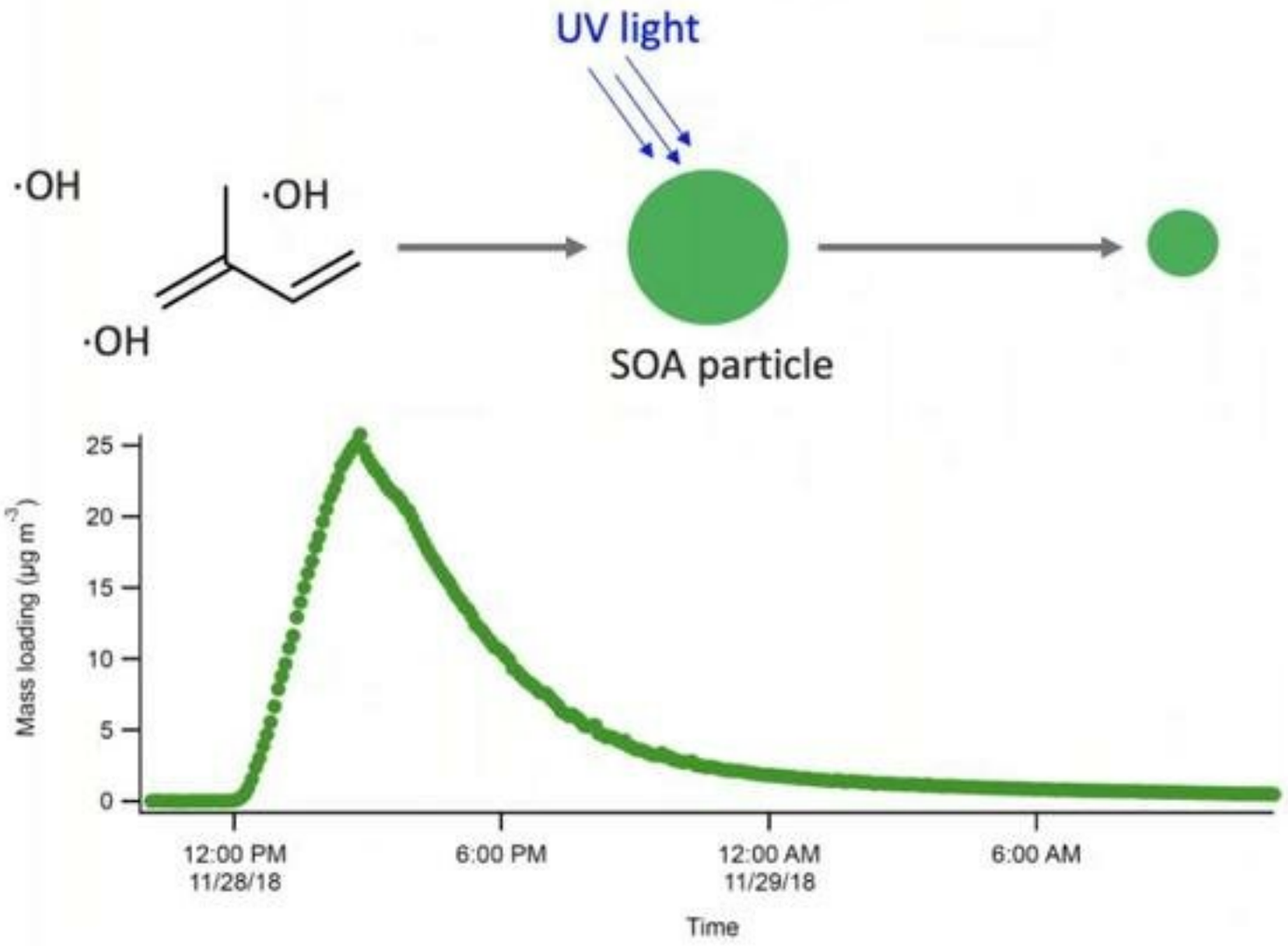
Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



Zhang et al. 2018, PNAS

Photolysis is an important sink of SOA found by laboratory measurements

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



- Isoprene SOA decreases by upto 80% within 10-12 hours upon exposure to UV light in PNNL smog chamber measurements (*Zawadowicz et al. 2020*)

New SOA model developments in E3SM

Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

- Parameterize multigenerational gas-phase chemistry of thousands of organic compounds with a few lumped species with the volatility basis-set (VBS) approach
- SOA precursors are from biogenic, biomass burning and energy-related emissions
- Included fragmentation reactions in addition to functionalization
- Included particle-phase oligomerization and photolysis of SOA

Volatilities of thousands of organic compounds represented by fixed decadal intervals (VBS)

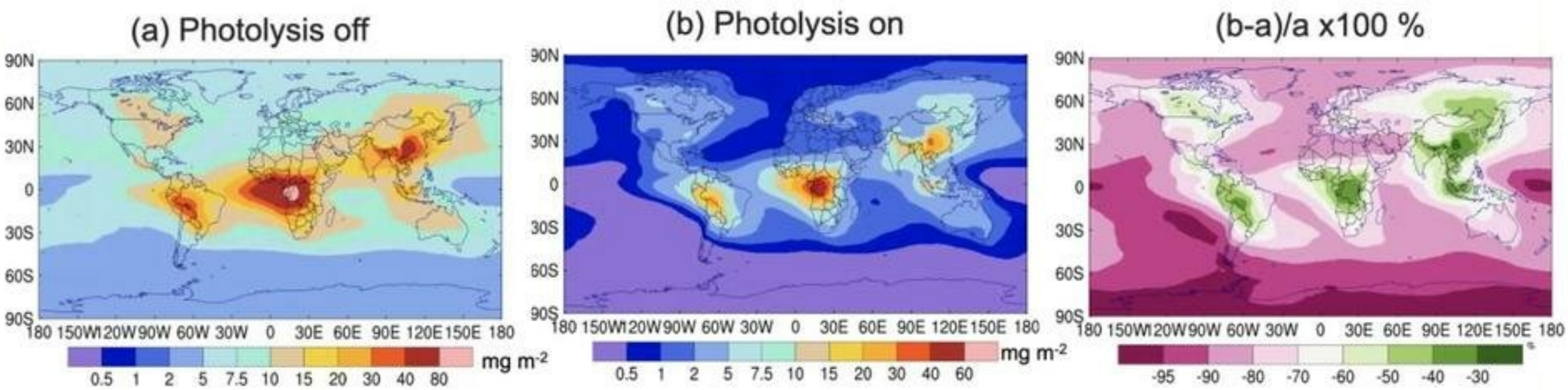
Making SOA treatments computationally efficient

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

- Added 8 gas-phase species to explicitly simulate multigenerational chemistry of SOA precursors
- At each time step gas-particle partitioning is calculated dynamically between semi-volatile gases and particle phase species within the MAM4 modes
- At the end of the time step all SOA particle-phase species are lumped together as a single non-volatile species assumed to be form oligomers within the particle
- Only a single SOA species is advected in each mode thus reducing the number of transported particle-phase species
- All SOA sources biogenic, biomass burning and fossil-related are lumped together
- This results in considerable computational savings compared to our previous implementation in CESM (Shrivastava et al. 2015)

Particle-phase photolysis is an important sink of SOA

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

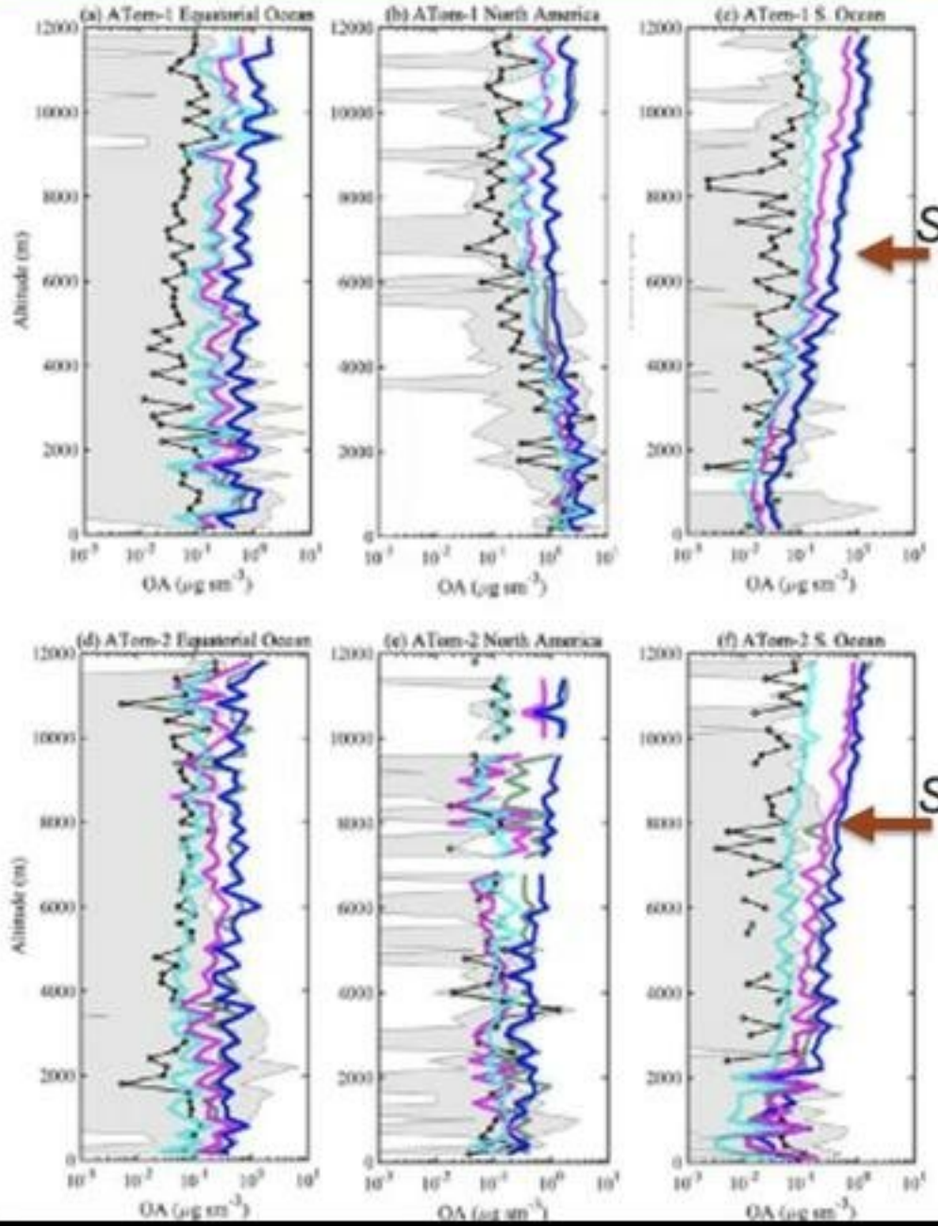


Lou, Shrivastava et al. 2020, JAMES

- Particle-phase photolysis decreases SOA by 30-50% over source regions
- Stronger (80-90%) decreases are seen over remote oceanic regions and high altitudes

Photolysis is an important sink of SOA needed to explain aircraft measurements mainly above 5km altitude

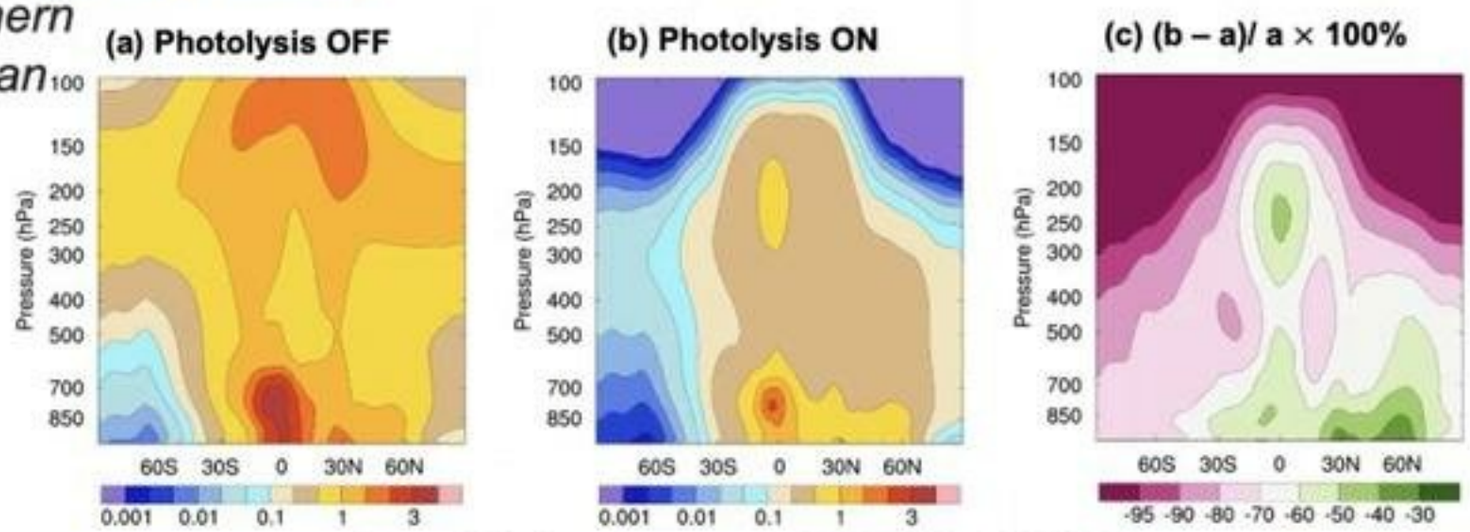
Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



Southern Ocean

Southern Ocean

Observed
Photolysis ON
Photolysis OFF

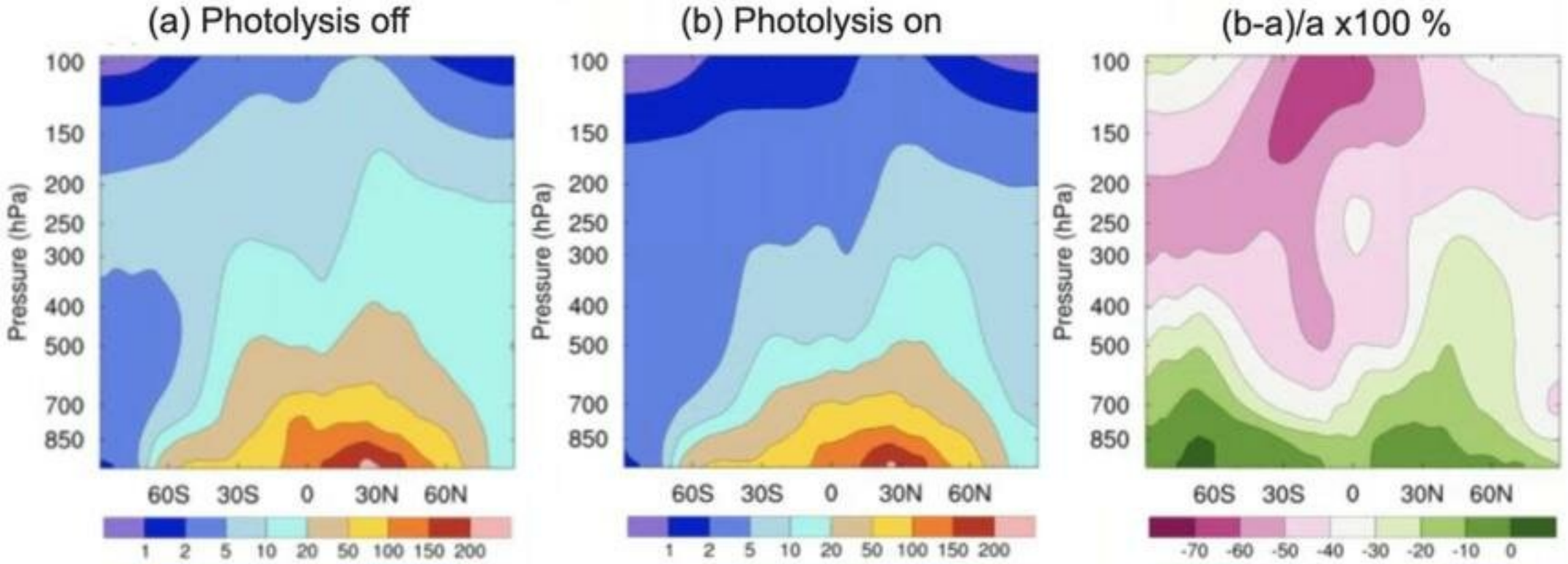


Lou, Shrivastava et al. 2020, JAMES

- Including photolysis improves simulated SOA vertical profiles significantly compared to Atom2016 aircraft measurements
- At high altitudes (above 5 km), wet removal is not efficient
- Photolysis is needed to explain observations at high altitudes

Photolysis decreases CCN more strongly at high altitudes

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

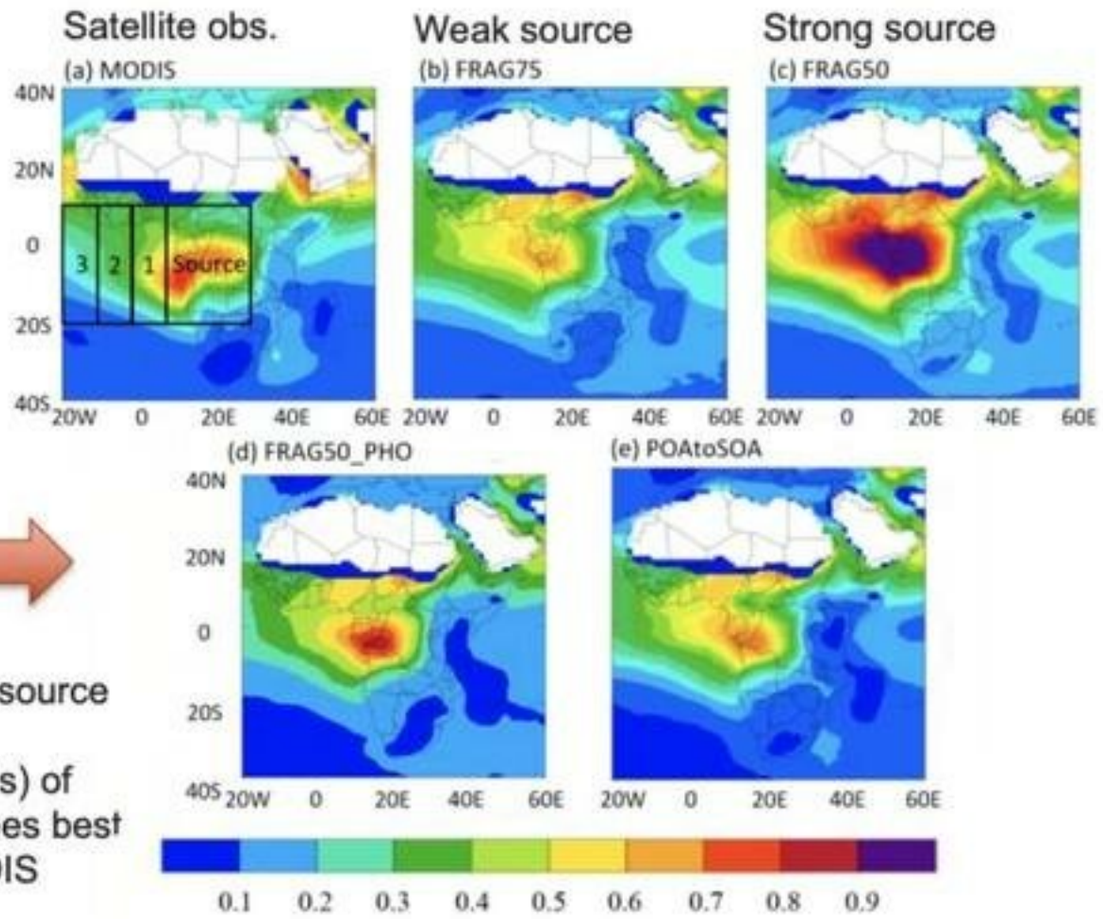


Lou, Shrivastava et al. 2020, JAMES

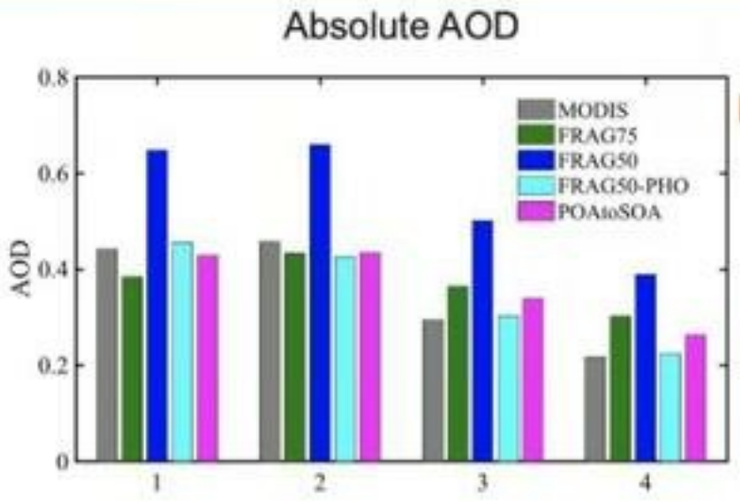
- Photolysis decreases CCN concentrations by 10-30% near surface and 50-70% at high altitudes

Satellite AOD over African biomass burning outflow dominated by organic aerosols: SOA model evaluation

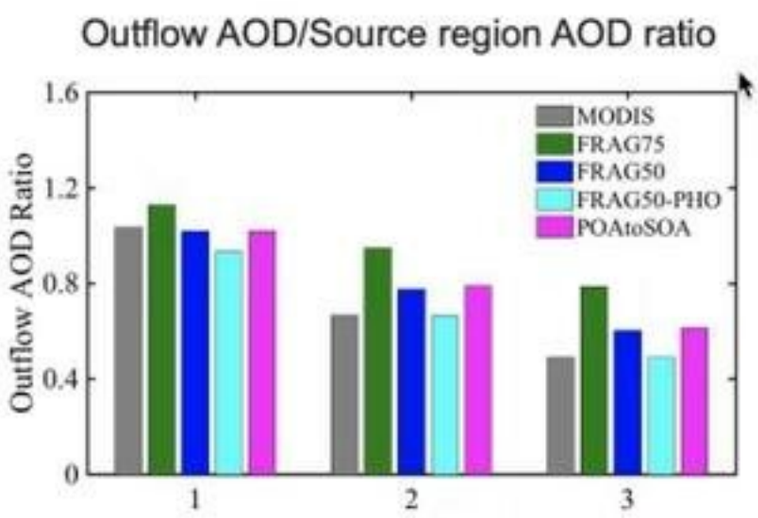
Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965



Stronger chemical source and sink (photolysis) of SOA agrees best with MODIS



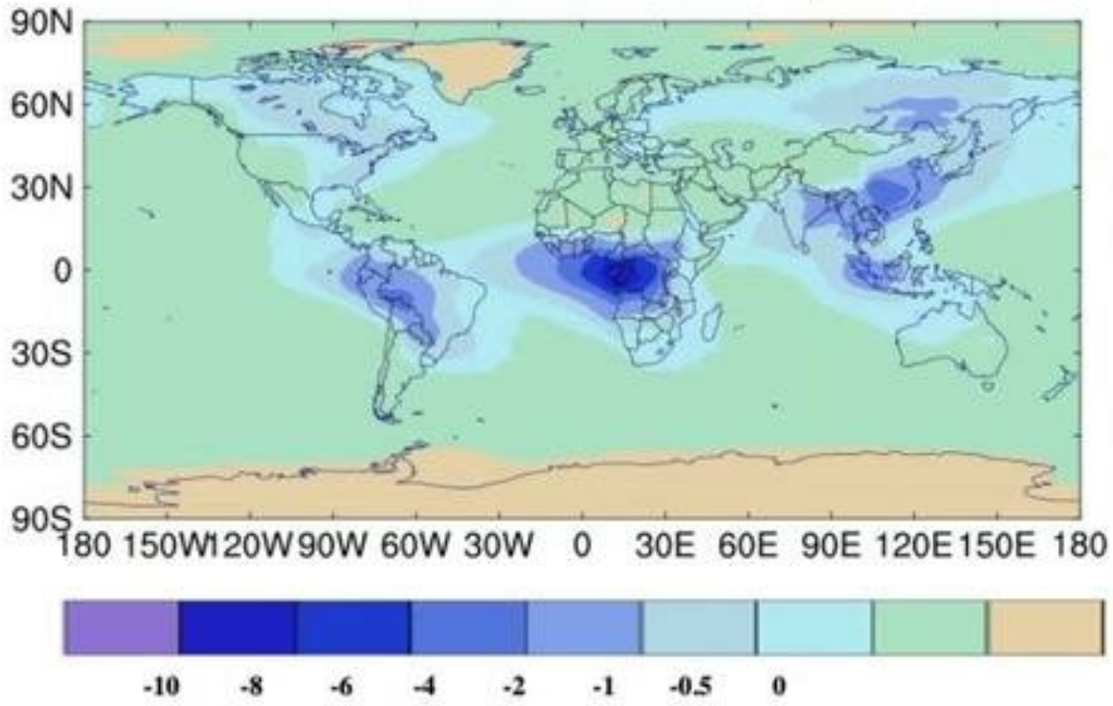
- Stronger SOA source (low fragmentation) and a strong sink (photolysis) needed to explain both absolute AOD and Outflow/Source AOD ratios



Simulated SOA direct radiative forcing at the top of atmosphere

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

DRE= -0.67 W m-2)



Lou, Shrivastava et al. 2020, JAMES

- IPCC radiative forcing of organic carbon is (-0.3 W m^{-2})
- IPCC estimates do not include SOA
- This study: DRE (radiation diagnostics in PD) for SOA alone is -0.67 W m^{-2}
- DRF of SOA (PD-PI) with photolysis is -0.1 W m^{-2}
- DRF of SOA when photolysis is turned off is -0.4 W m^{-2}
- Photolysis decreases DRF of SOA by a factor of 4

Key Takeaways and Modeling Implications

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

- We developed a computationally efficient framework that enables explicit coupling of SOA processes with energy and land use changes
- Both stronger SOA sources (functionalization) and stronger sinks (photolysis) needed to explain SOA distributions
- Vertical profiles of SOA measured by aircraft are key to constraining SOA processes in models
- Satellite AOD in regions dominated by biomass burning provides valuable constraints
- PD-PI effective radiative forcing of SOA decreases by a factor of four from -0.4 to -0.1 W m^{-2} when we include the photolysis of SOA
- Accurately representing key SOA chemical processes in global models is essential for understanding their interactions with clouds and radiative forcing

Ongoing and Potential Future Developments

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

- Currently ongoing as part of NGD atmospheric physics: Coupling new SOA treatments to E3SM-MOSAIC that includes nitrate aerosols in addition to sulfate

Other Potential developments (depending on support/approval):

- Include the treatment of phase state of SOA (solid, liquid, glassy)
- Phase state affects both SOA formation and cloud microphysical properties, not included in current models
- Represent interactions between sulfate and SOA chemistry in aqueous aerosols and clouds
- Treat light absorbing components of SOA (brown carbon) and evaluate their role in aerosol-radiation and aerosol-cloud interactions