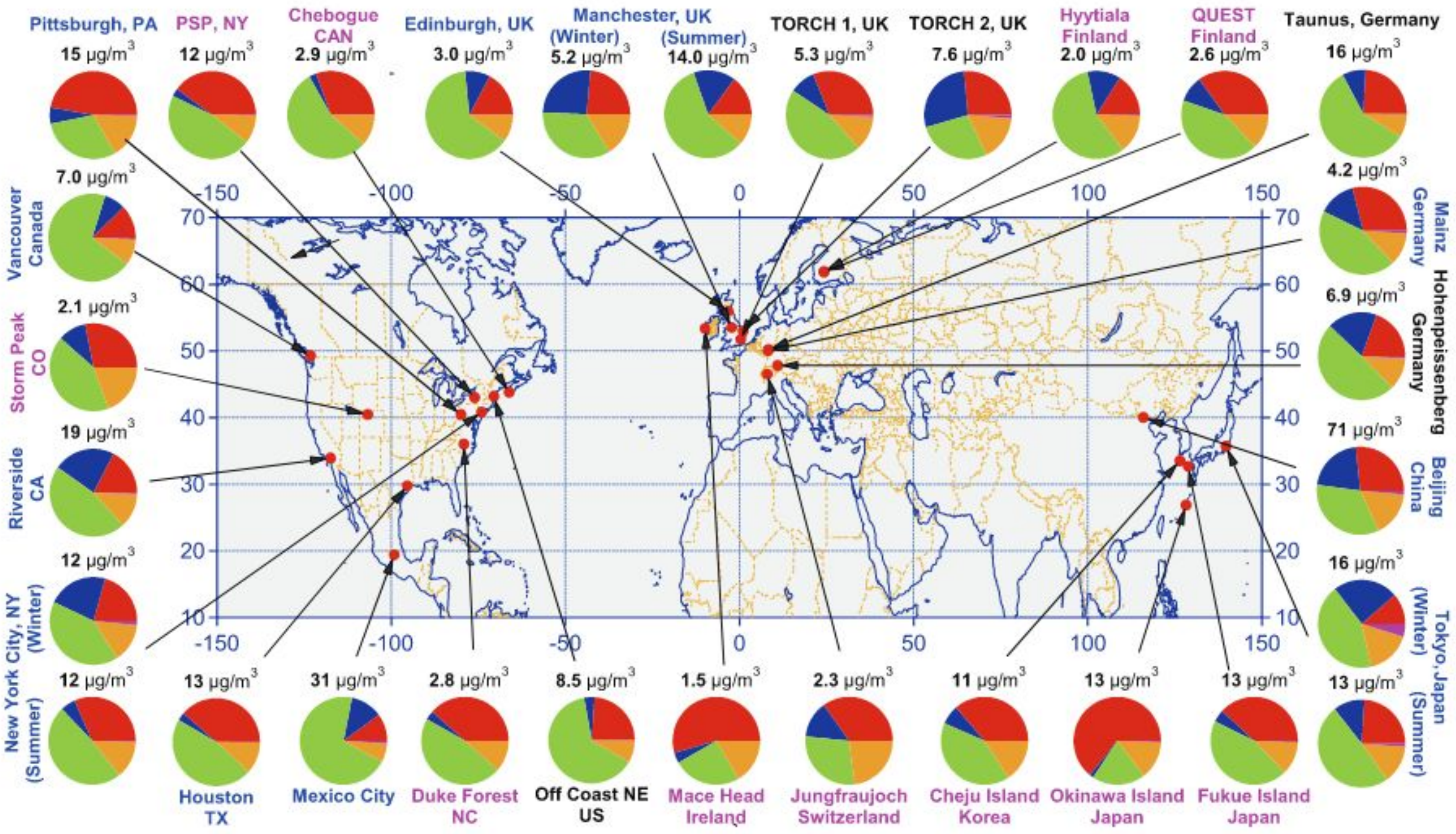
# Drivers of biogenic secondary organic aerosol from the past to the present and the future

Yang Shi, Colette L. Heald, Jesse H. Kroll

Massachusetts Institute of Technology

# Organic aerosol is a major component of atmospheric fine aerosols, and a major fraction of the total organics are **secondary organics**.




Organics
Sulfate
Nitrate
Ammonium
Chloride

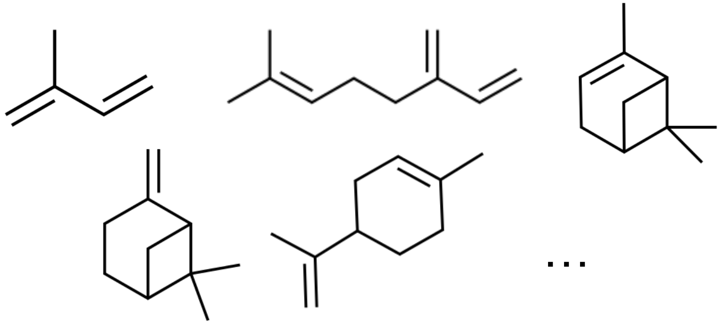
Zhang et al. (2007)

Organic contribution: 18-70%; average = 45%

# Secondary organic aerosol (SOA) formation

Vegetation (e.g., trees)

Biogenic volatile organic compounds (BVOCs)

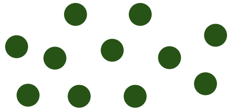



Oxidation

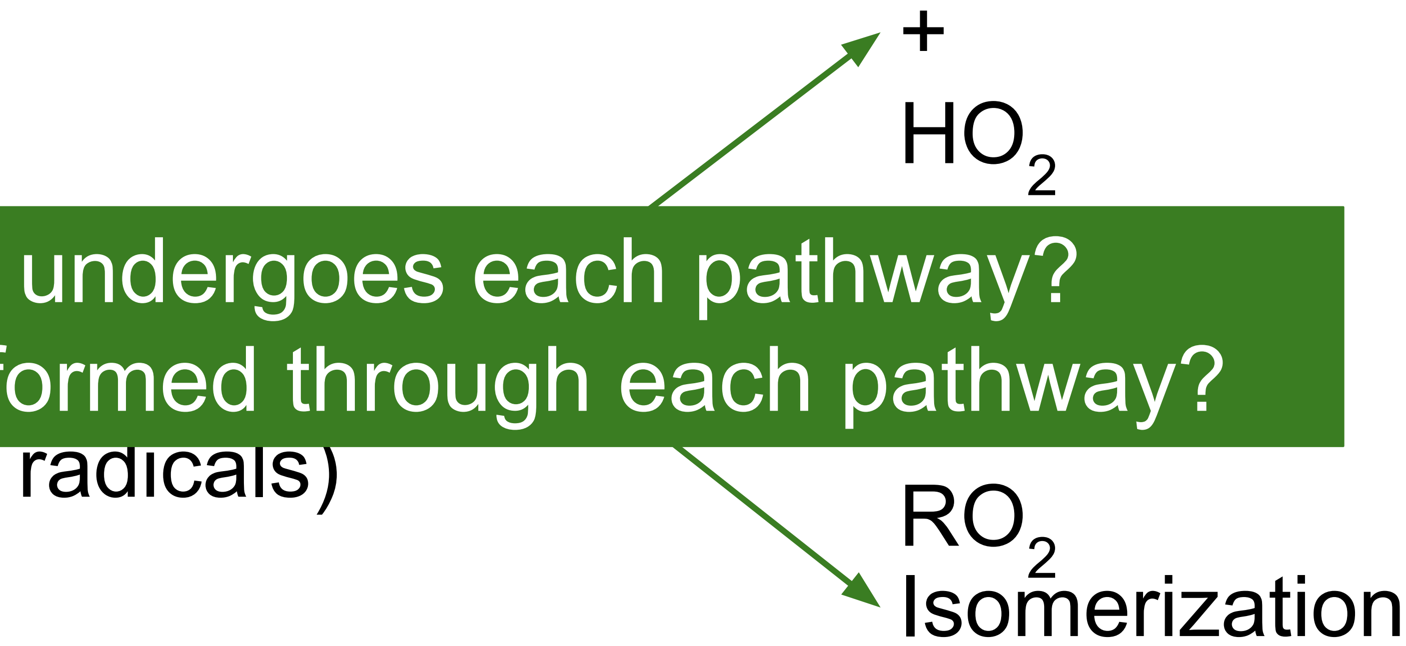
SOA precursor gases (SOAG)

Gas-aerosol partitioning

SOA



# VOC Oxidation by OH and $RO_2$ Chemistry




How much $RO_2$ undergoes each pathway?
How much SOA is formed through each pathway?

$RO_2$ fate determines the ultimate amount of SOA.

# Community Earth System Model version 2.2 (CESM2.2)

**CLM5**
(Community Land Model)

**CAM6-Chem**
(Atmosphere component with full chemistry)

MEGAN

MOZART-TS2 with Volatility Basis Set (VBS)

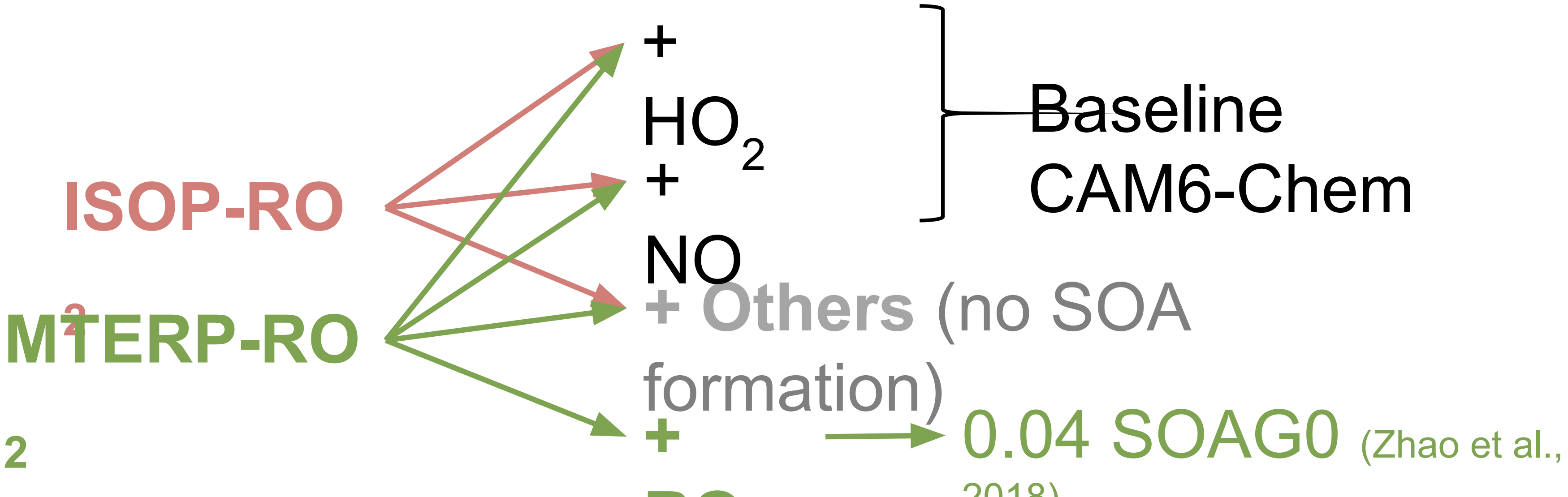
MAM4

Prognostic BVOC emissions (Isoprene, Monoterpenes, & Sesquiterpenes)

Gas-phase chemistry

Aerosol model

# Expanded $RO_2$ chemistry in VBS

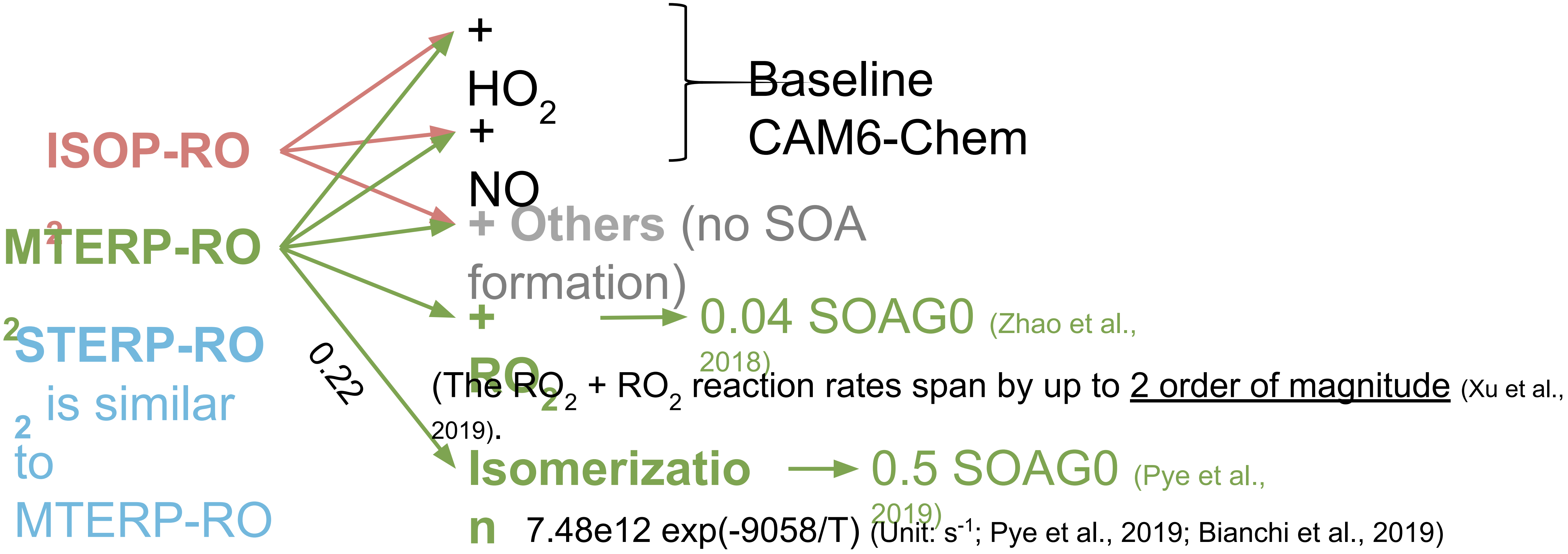



(The $RO_2$ + $RO_2$ reaction rates span by up to 2 order of magnitude (Xu et al., 2022).

| | **Fast** | **Slow** |
|---|---|---|
| + ISOP-$RO_2$ | 2e-11 (Berndt et al., 2018b) | 2e-11 (Berndt et al., 2018b) |
| + MTERP-$RO_2$ | 4e-11 (Berndt et al., 2018b) | 1e-12 (Zhao et al. 2018) |
| + Later generation Terpene $RO_2$s | 2.6e-10 (Berndt et al., 2018a) | 1e-12 (Zhao et al. 2018) |

Unit: $cm^3$ $molec^{-1}$ $s^{-1}$

# Expanded $RO_2$ chemistry in VBS




- Photolysis of BSOA in SOAG0 bin is turned off (e.g., O'Brien and Kroll, 2019; Baboomian et al., 2
- Individual reactions are tagged separately.

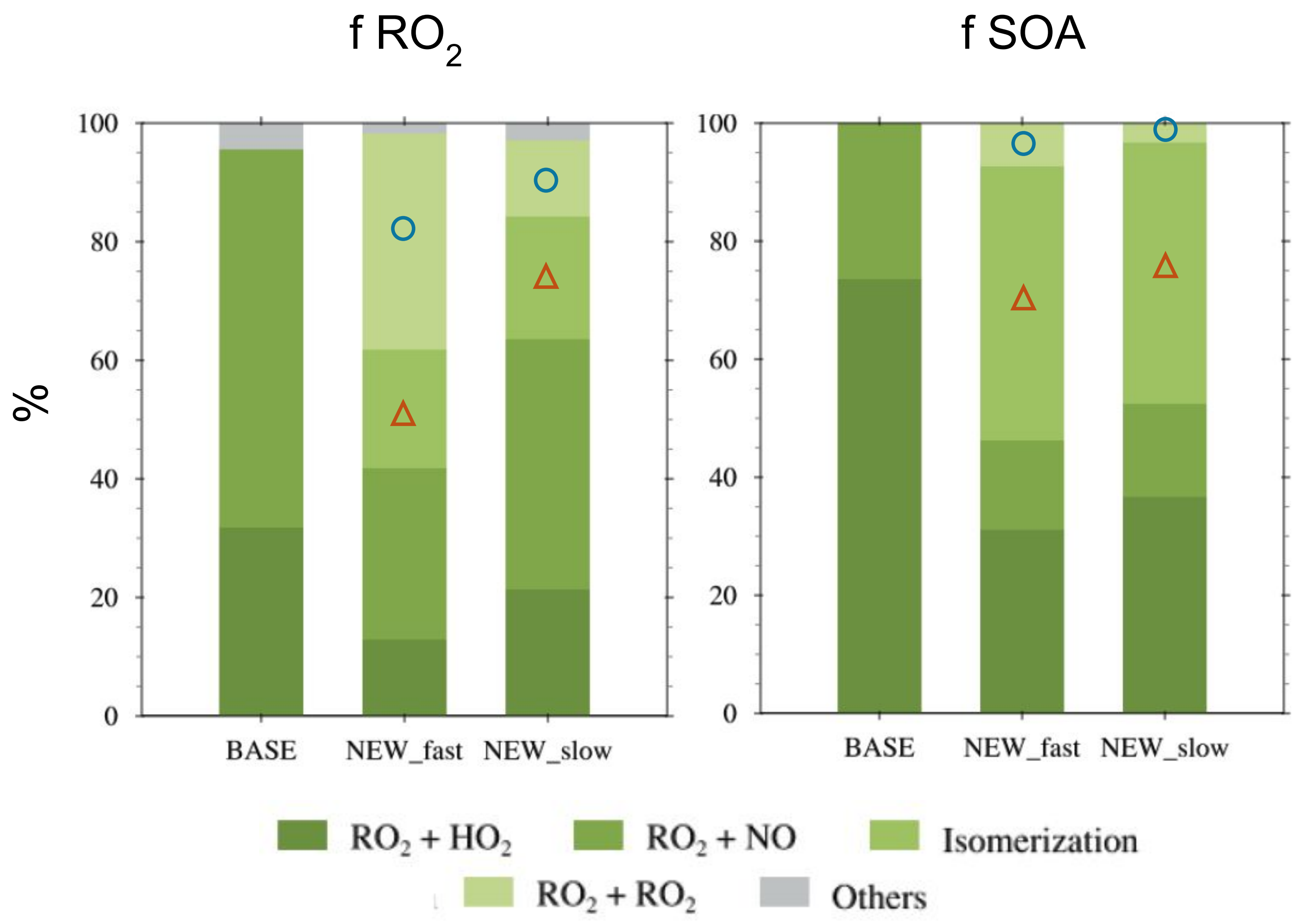
# Experiments

| | |
|---|---|
| **BASE** | Baseline chemistry |
| **NEW_fast** | Expanded chemistry (Fast and slow $RO_2 + RO_2$ reaction rates are tested) |
| **NEW_slow** | |

Configuration

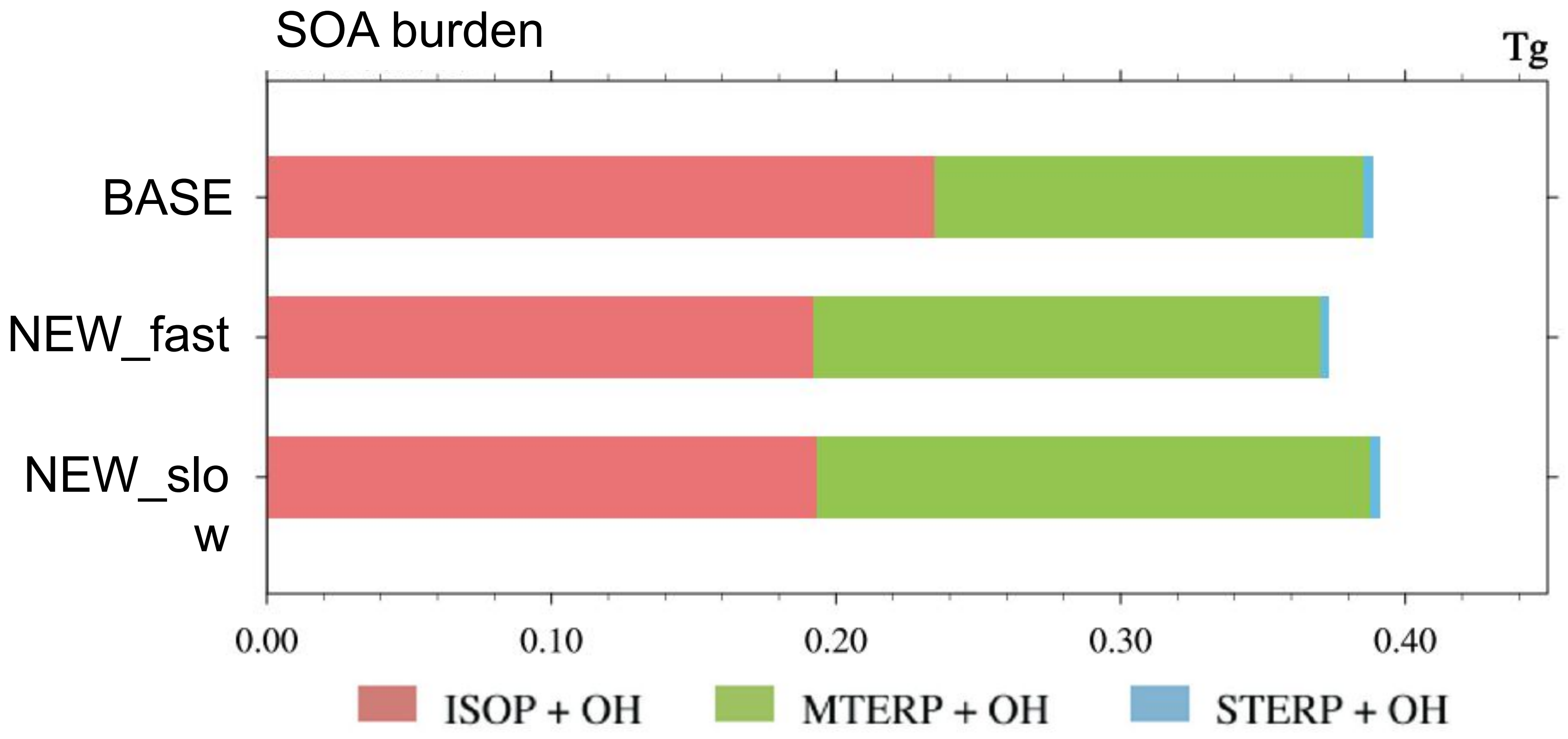
- Climate, $CO_2$ concentrations, and anthropogenic emissions in present-day (PD)

# MTERP-$RO_2$ fates and contributions to SOA burden in PD



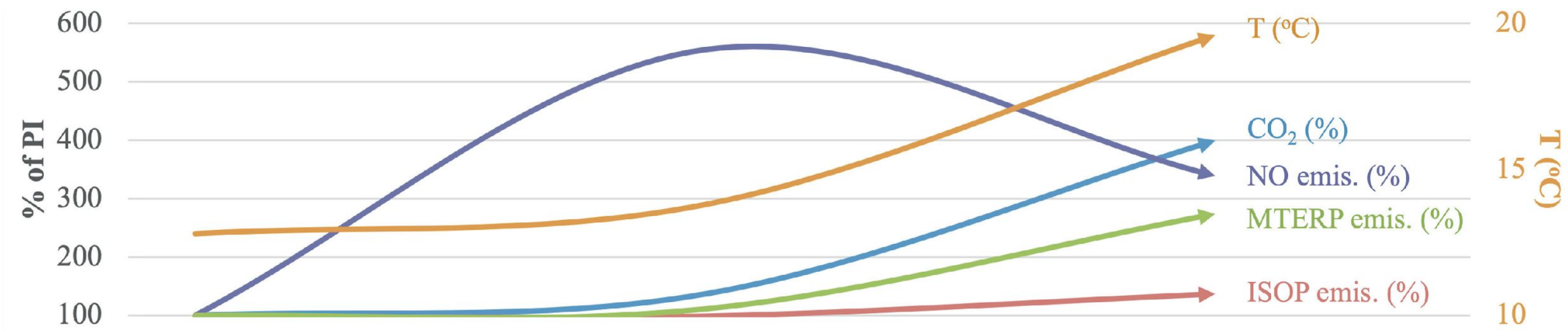

- **Isomerization** is **nearly saturated** in the MTERP-$RO_2$ fate, and it contributes to more than **40% of total SOA** formed by MTERP-$RO_2$.
- **MTERP-$RO_2$+$RO_2$** reactions contribute moderately to the $RO_2$ fate, but **minor to SOA burden**.

# Total BSOA burden is similar, despite of the large differences in chemistry mechanisms




This is due to the similarities in SOA yields in various pathways, highlighting the need to better constrain SOA related parameters in the laboratory.

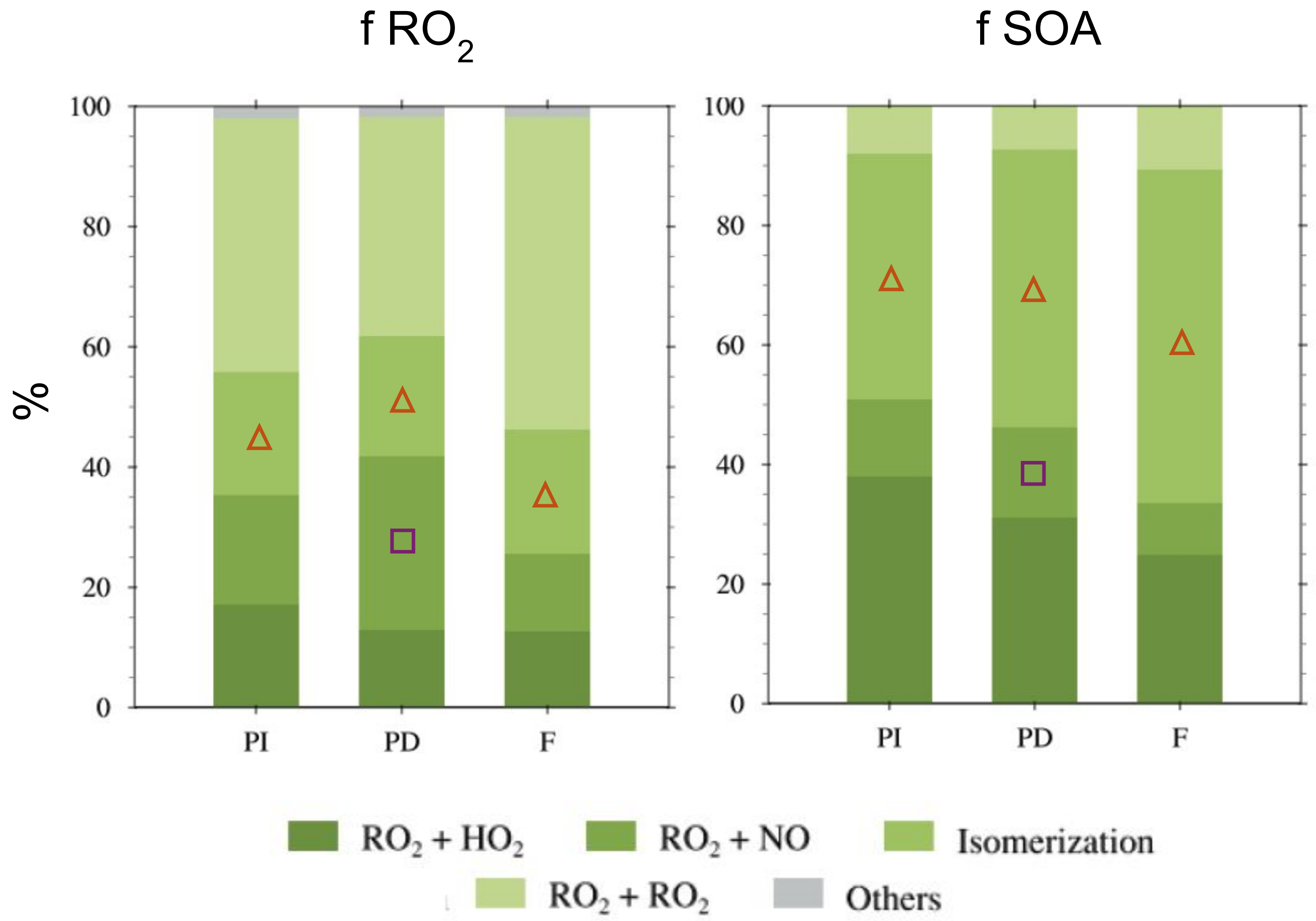
# Examine the changes in SOA formation from the past to the future




| **PI** | **PD (NEW_fast)** | **F** |
|---|---|---|
| Pre-industrial 1850 | Present-day 2010 | Future (SSP585) 2100 |

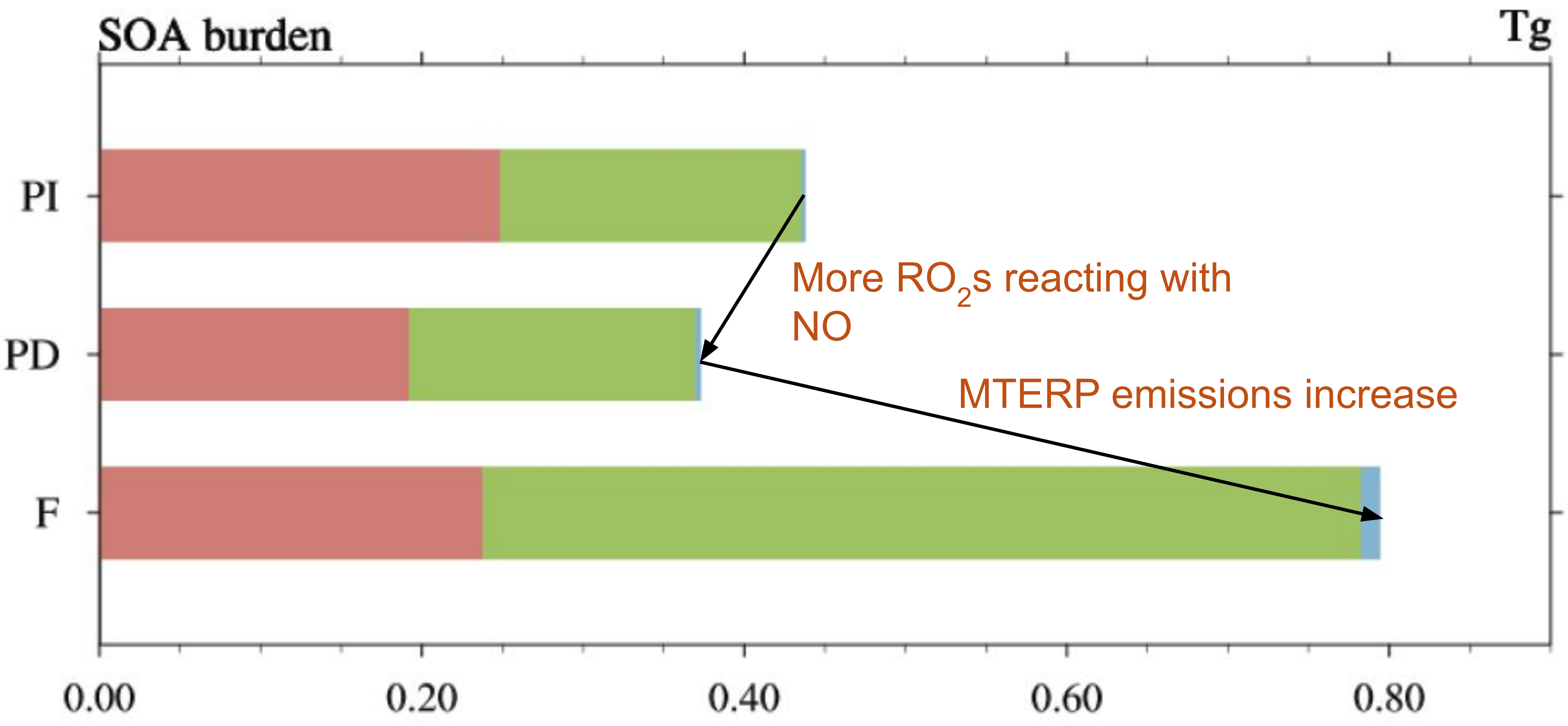
- These experiments use the same chemistry as **NEW_fast**.

# MTERP-$RO_2$ fates and contributions to SOA burden from PI to F

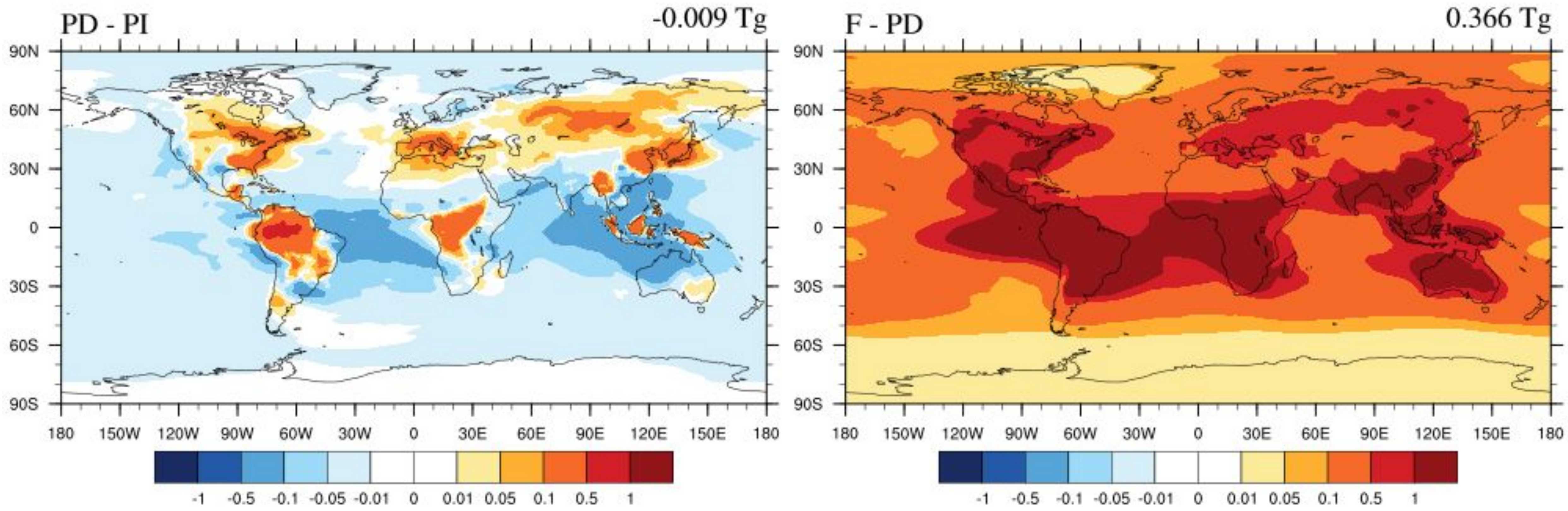



- The isomerization of MTERP-$RO_2$ remains saturated from PI to F, contributes greatly to total SOA.
- The $RO_2$ + NO pathway contributes more in PD than in PI and F.

# SOA burden decreases from PI to PD, but increases from PD to F.



# Global MTERP+OH SOA distribution change



# Summary

- We expanded the $RO_2$ chemical mechanism in CAM6-Chem VBS to include the isomerization and $RO_2$ + $RO_2$ pathways for MTERP-$RO_2$.
- The **isomerization** pathway saturates its branch and contributes significantly to MTERP SOA burden in PI, PD, and F.
- The **$RO_2$ + $RO_2$** pathway contributes moderately to $RO_2$ fate but minor to SOA burden.
- Large uncertainties exist in the SOA-related parameters. This highlights the need to better constrain them in the lab experiments.

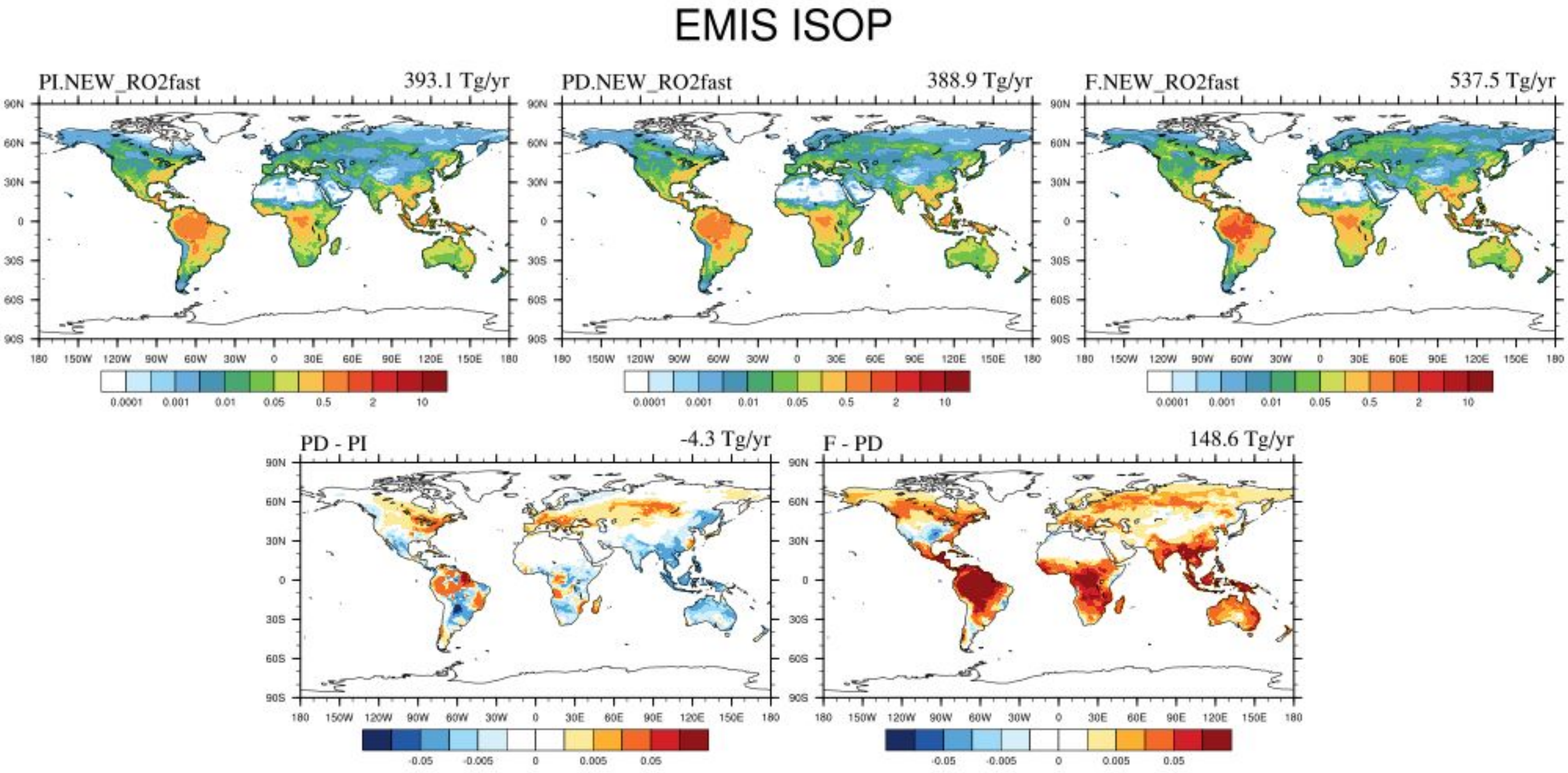
Contact: yangshi@mit.edu;
colette.heald@env.ethz.ch

# New tracers/reactions

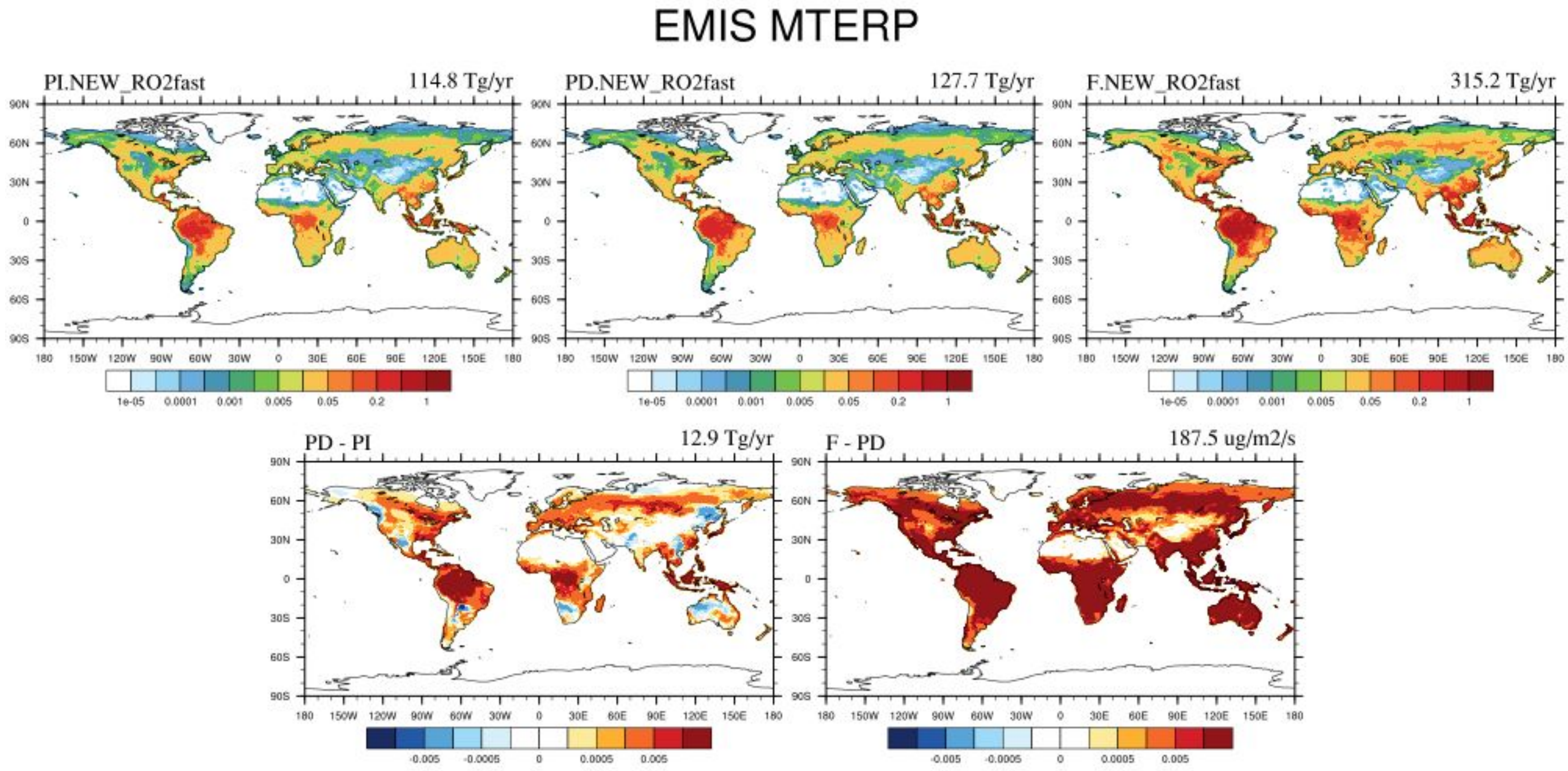
- 41 * 3 (SOAG, soa_a1, soa_s2) = 123 advected tracers
- 41 * 2 (soa_c1, soa_c2) + 4 RO2s = 86 non-advected tracers
- >170 new reactions

- Computational cost increase (only) by ~ 30%

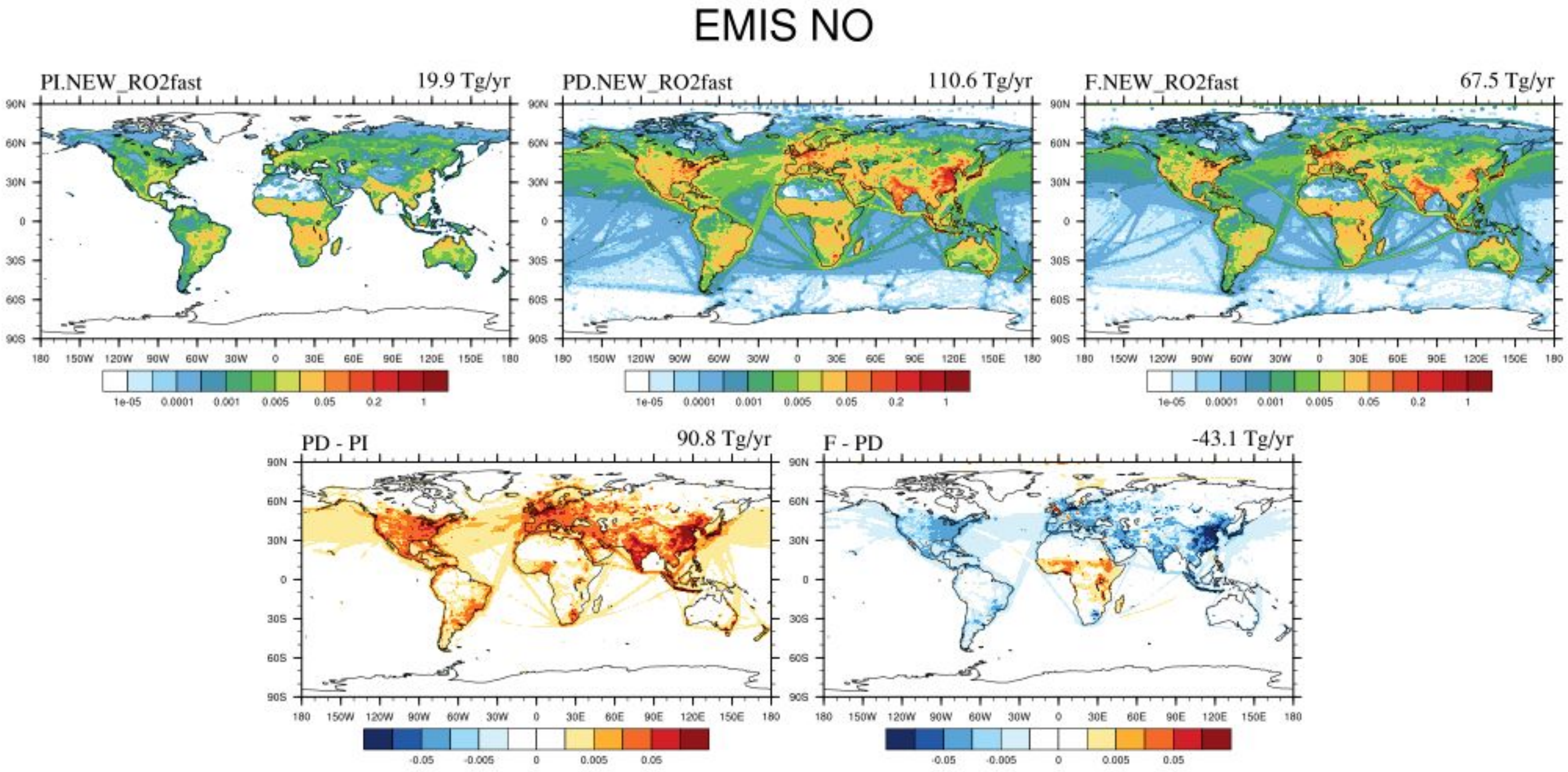
# Isoprene emission changes (PI to F)



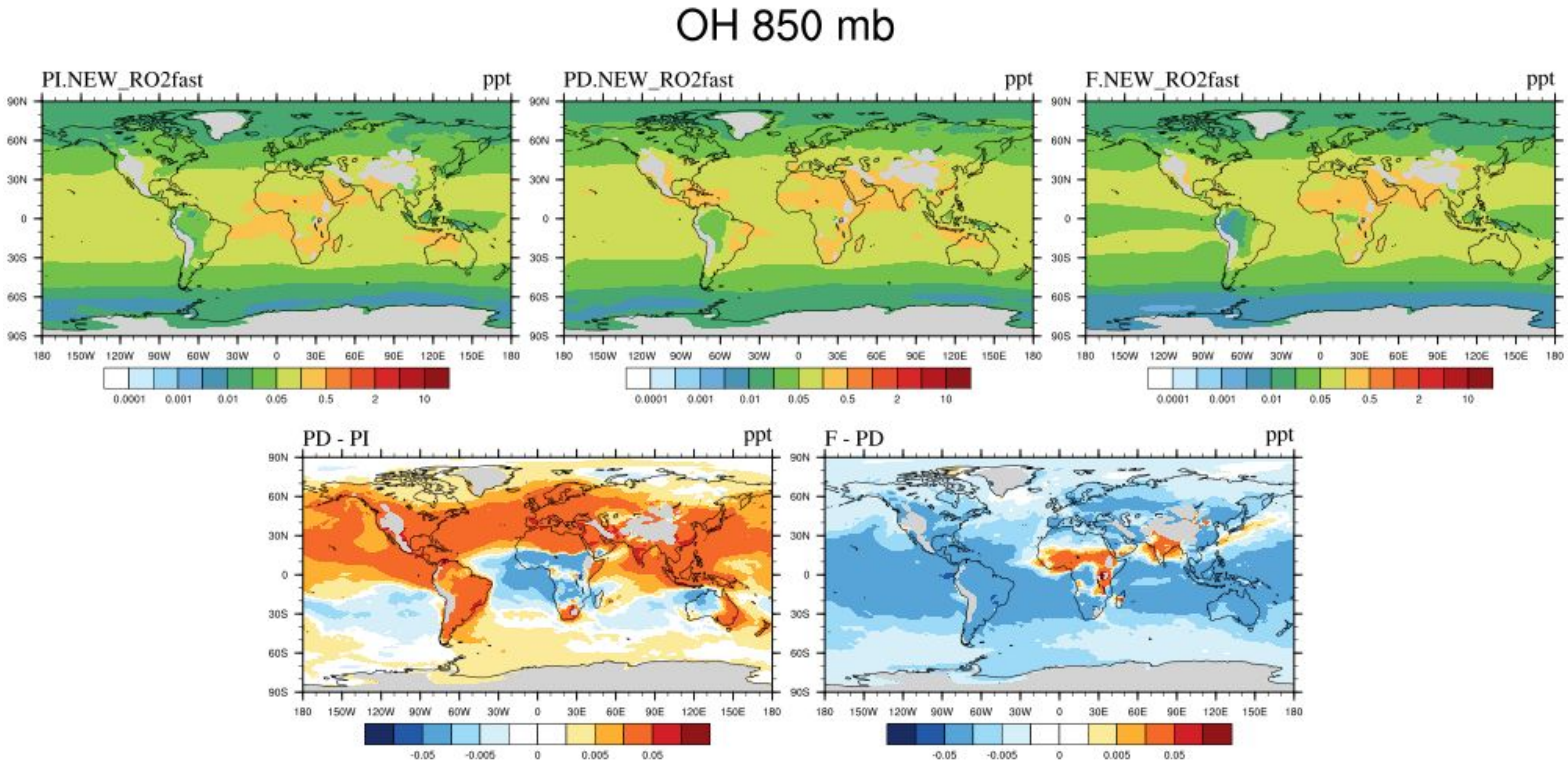
# Monoterpene emission changes (PI to F)



# NO emission changes (PI to F)



# OH changes (PI to F)



# $O_3$ changes (PI to F)