Rethinking SOA growth and removal in 3D models based on explicit chemistry

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Objective:

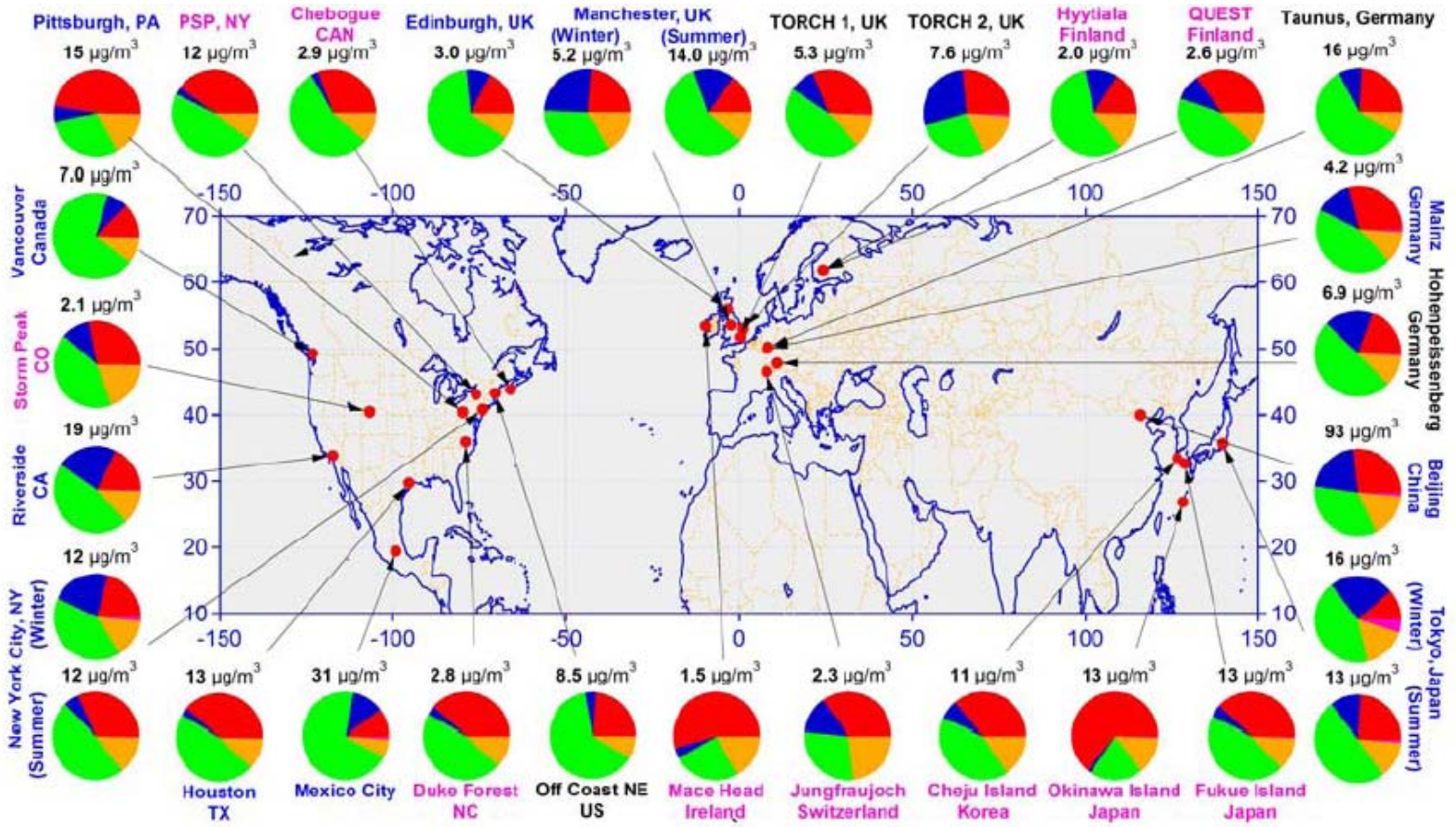
- Develop new parameterizations that can capture the SOA production and solubility predicted by an explicit chemical mechanism (GECKO-A)

CESM meeting, Boulder, 11 Feb. 2014

Secondary Organic Aerosols

Zhang et al., GRL, 2007

- Dominance of organic aerosols (30-70%)
- Scattering (and absorbing?)
- Direct & indirect forcing on climate



AMS obs
Sulfate
Organics
Nitrate

-> formation mechanisms are complex and unconstrained

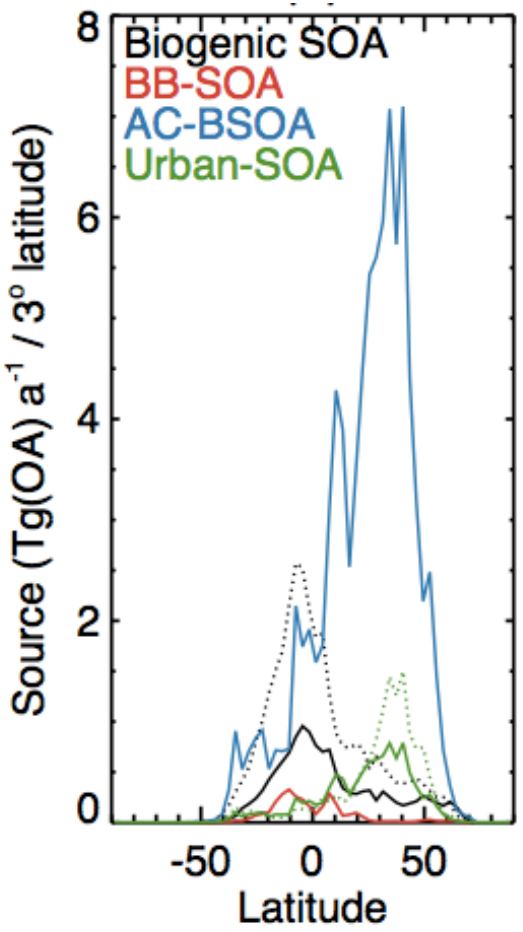
-> removal mechanisms rely mainly on aerosol wet deposition, others not quantified

How important is anthropogenically controlled SOA?

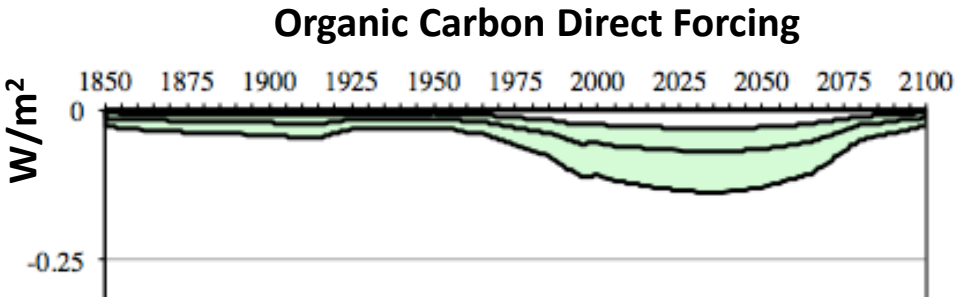
- **Spracklen et al., 2011:** Top-down using AMS & C^{14} obs
  - Production: 100 Tg yr^{-1} (anth. Controlled)
  - Direct forcing: -0.26 ± 0.15 Wm^{-2}
  - Indirect: -0.60 ± 0.24 Wm^{-2}

- **Jo et al., 2013:**
  - Production: 88 Tg yr^{-1}
  - Direct forcing: -0.28 Wm^{-2}

- Carlton et al., 2010: 50% of biogenic SOA in the USA is anthropogenically controlled



=> Larger effect than in Smith and Bond, [2013]

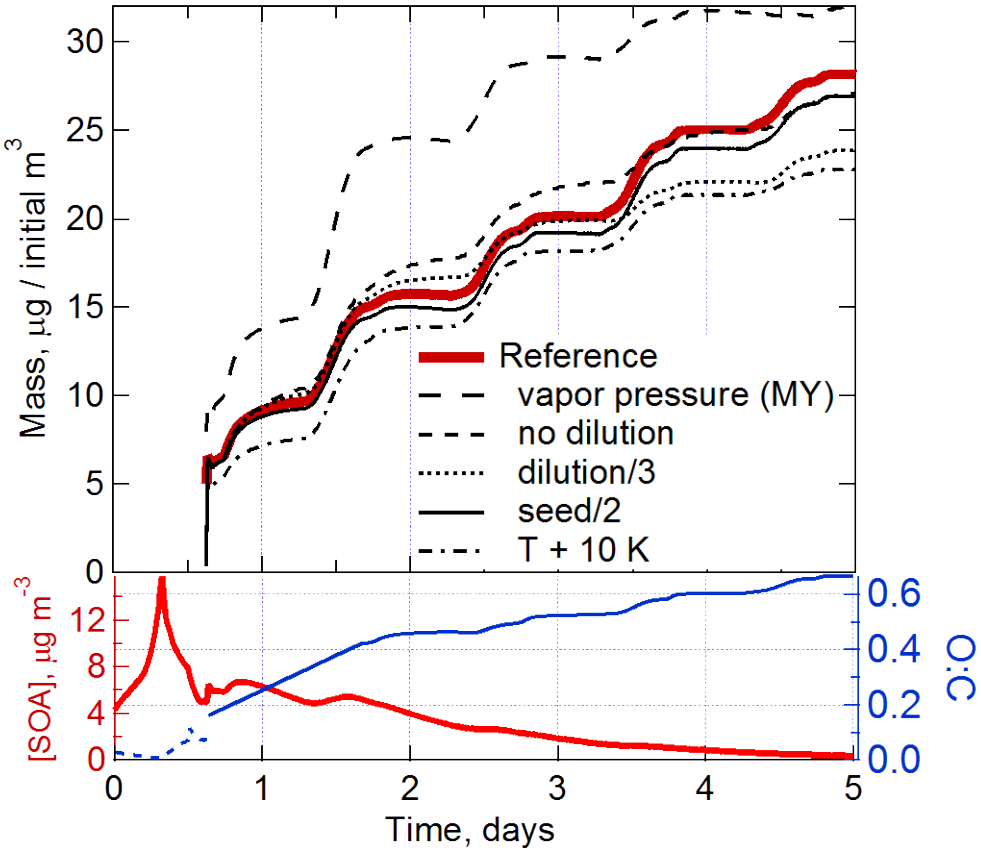


No SOA in those runs!!
Inconsistent with the abundance of SOA

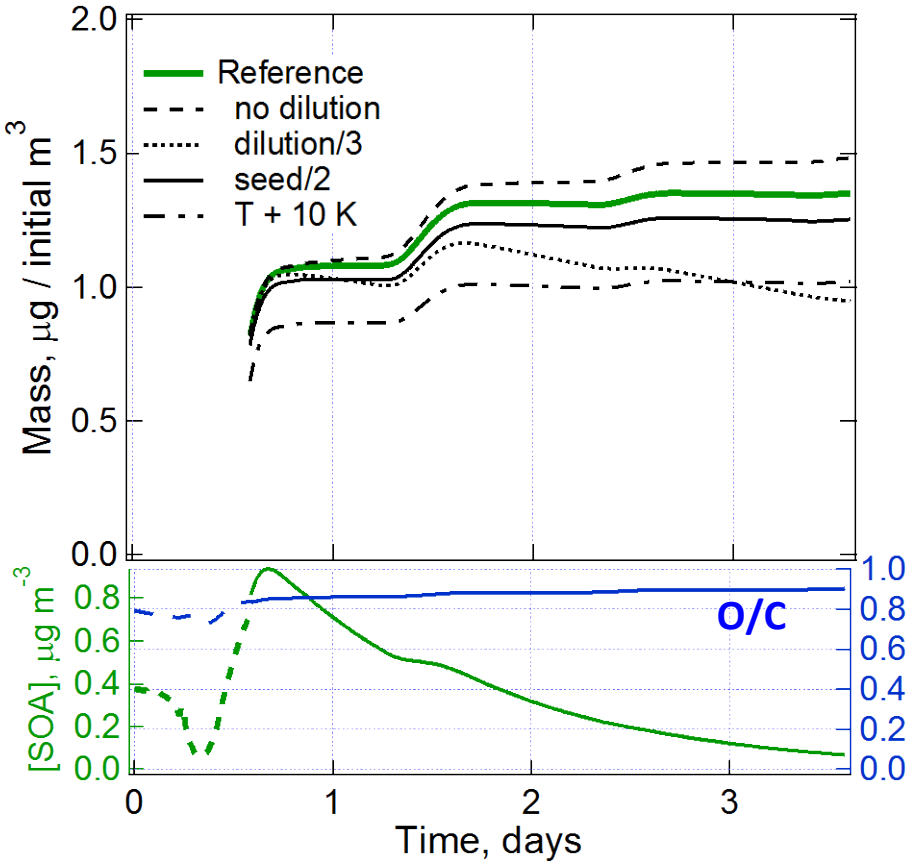
Explicit model GECKO-A suggests growth of anthropogenic SOA

[*Lee-Taylor et al. in prep.*]

Mexico City outflow

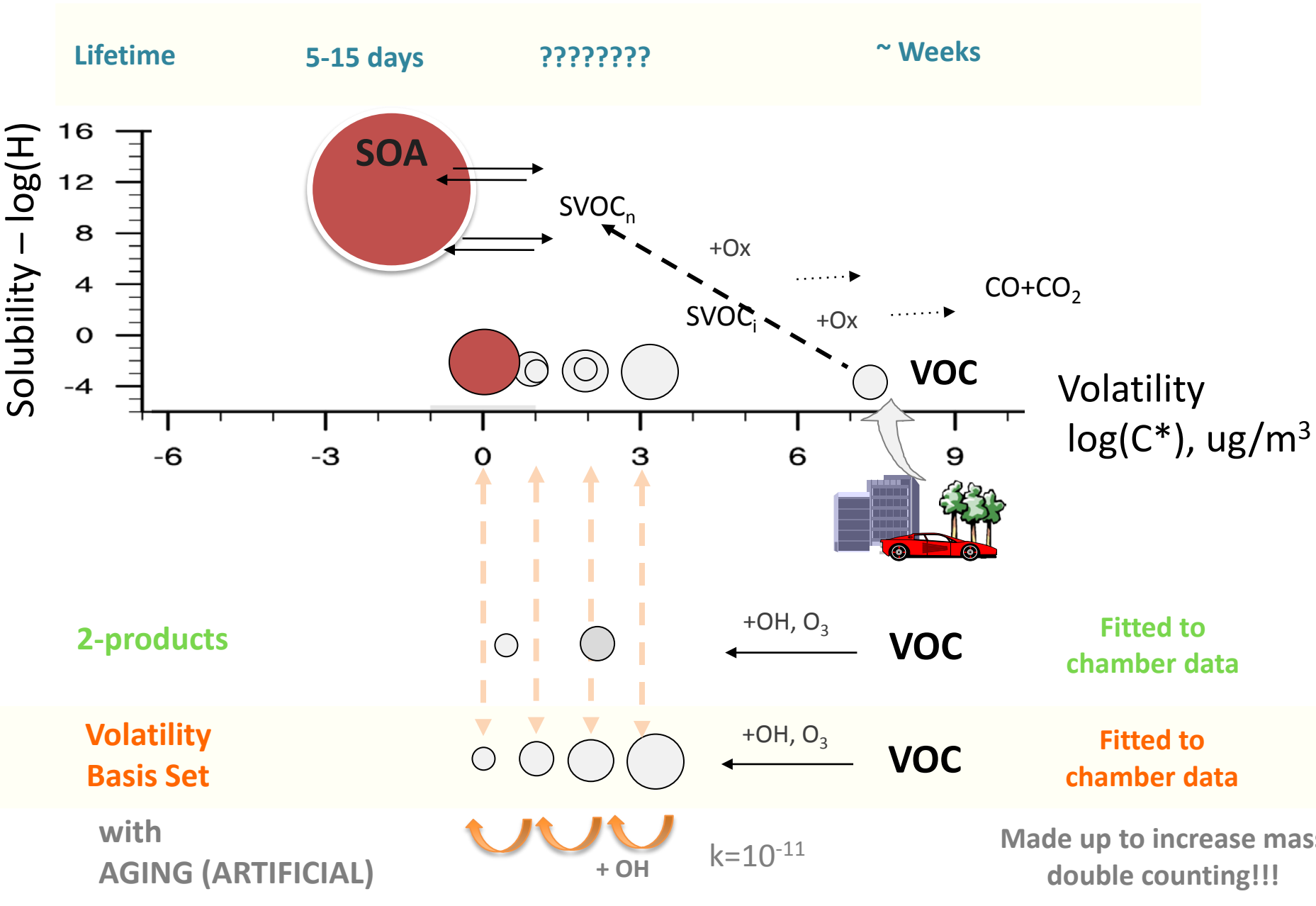


Colorado pine forest (terpenes)



- GECKO-A suggests a strong multiday growth for anthropogenics, which is not in 3D models.
- Need to re-evaluate the radiative impact of anthropogenic sources vs. preindustrial

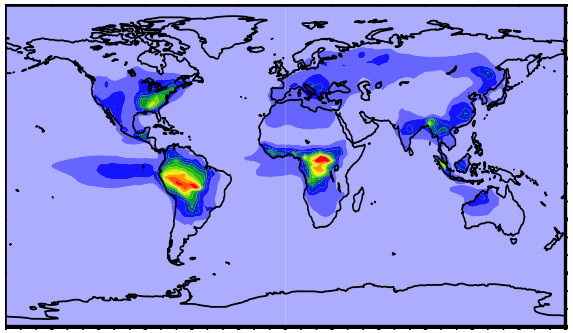
What is in current 3D models (gas-phase)?



2-product vs. VBS in CAM-chem

Boundary layer SOA predicted from various approaches in CAM4

2-product model



SOA (μg/m^3)

Burden: **0.8 Tg**
Production: **30.2 Tg/year**
Lifetime: **9.7 days**

Increase SOA VBS

SOA (μg/m^3)

0.67 Tg
30.3 Tg/year
8.1 days

Increase SOA VBS with ageing

SOA (μg/m^3)

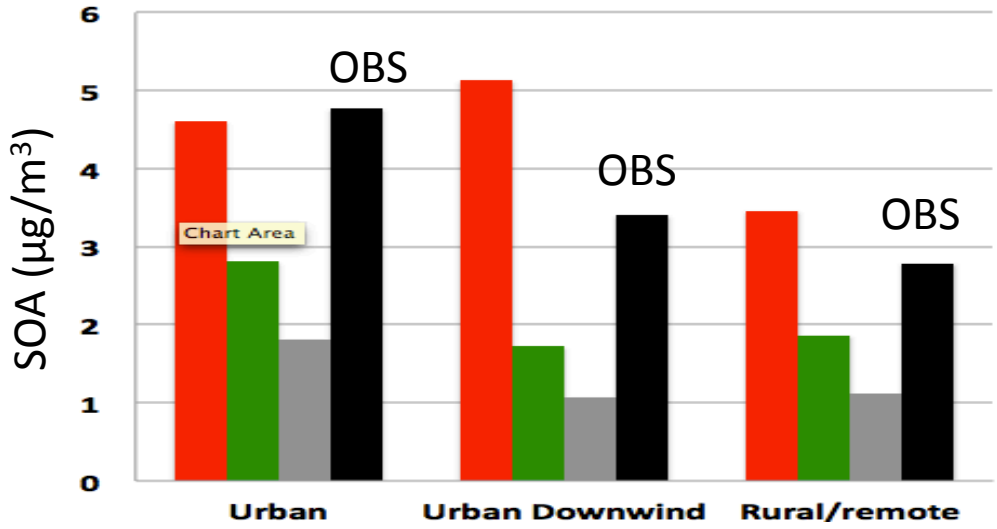
1.18 Tg
67.1 Tg/year
6.4 days

⇒ Changes in SOA lifetime when the burden is increased

(courtesy of Yiqi Zheng, Yale)

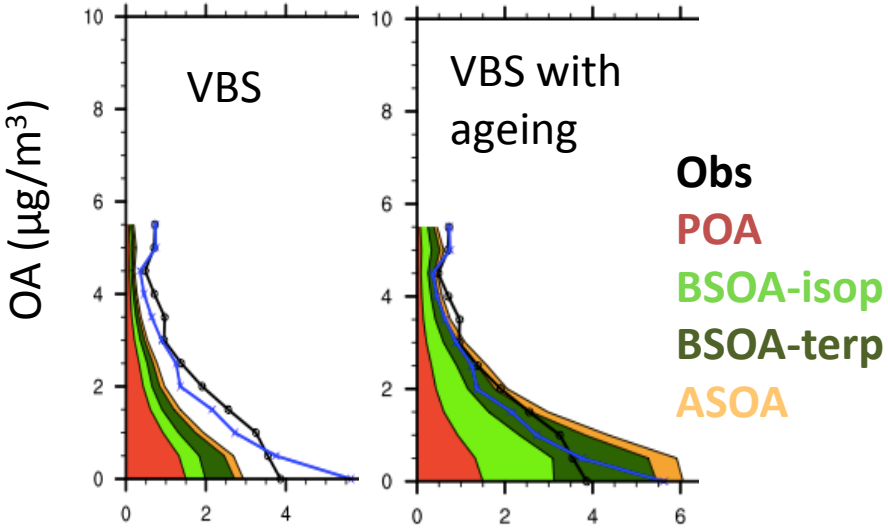
2-product vs. VBS in CAM-chem

Comparison with surface AMS global data



(courtesy of Pengfei Yu, CU)

Comparison with aircraft data (e.g. Texas-AQ)



- Current Models can adjust the mass to match SOA observations

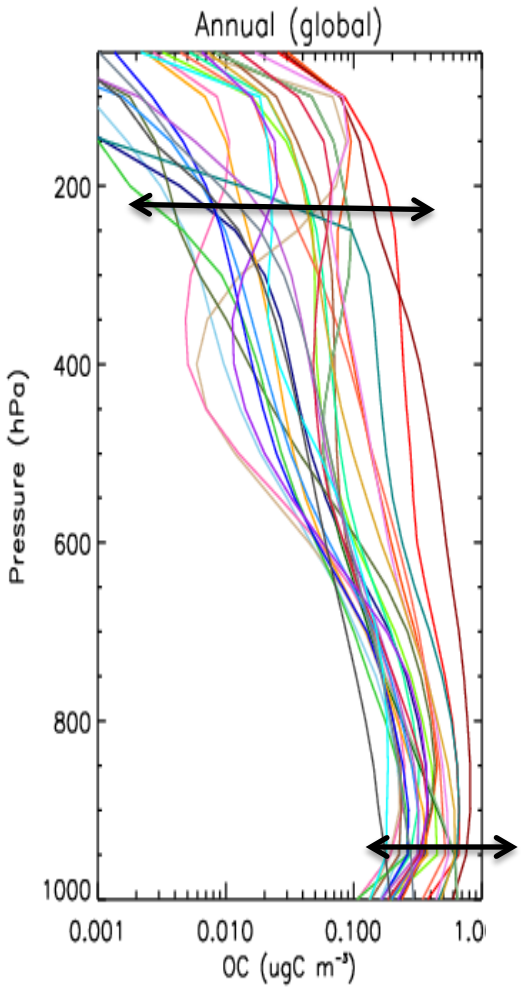
BUT

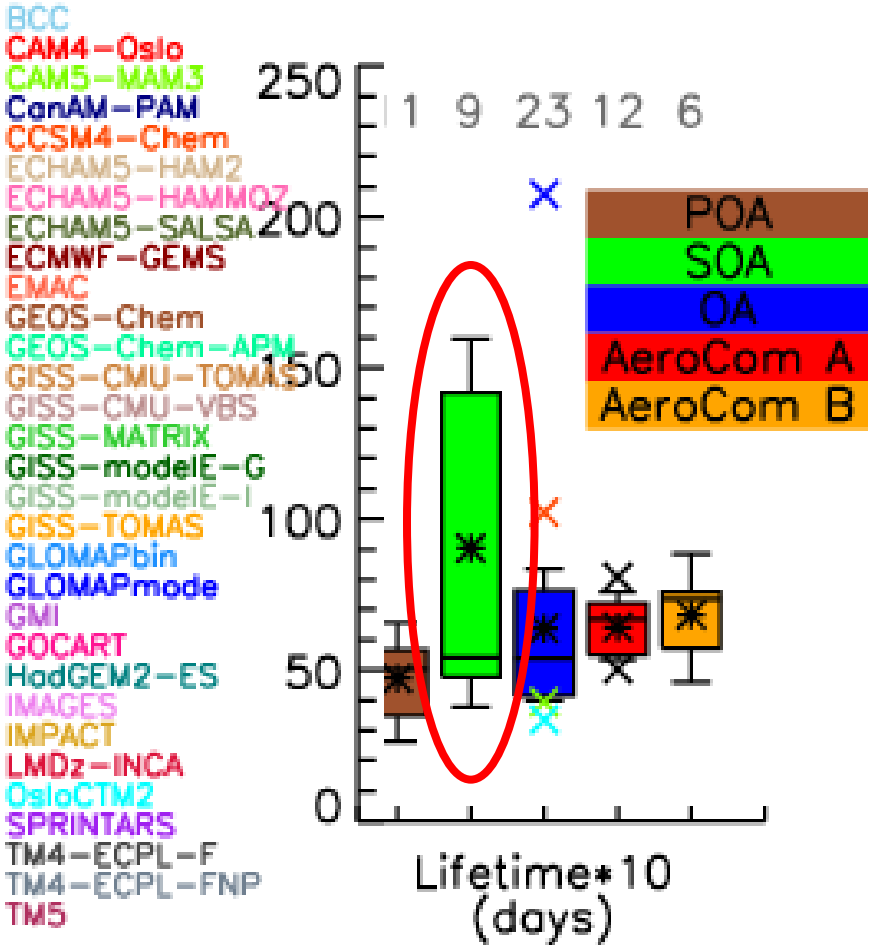
- future predictions of anthropogenically controlled SOA won't be better constrained

Global model intercomparison study for OA

[*Tsigaridis et al., ACPD*]

=> Large variability in 3D models





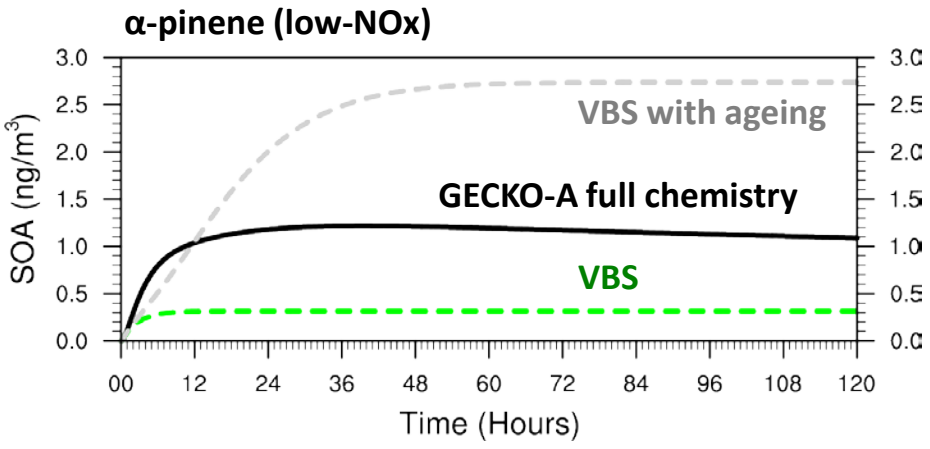
=> Need to better constrain SOA production and removals

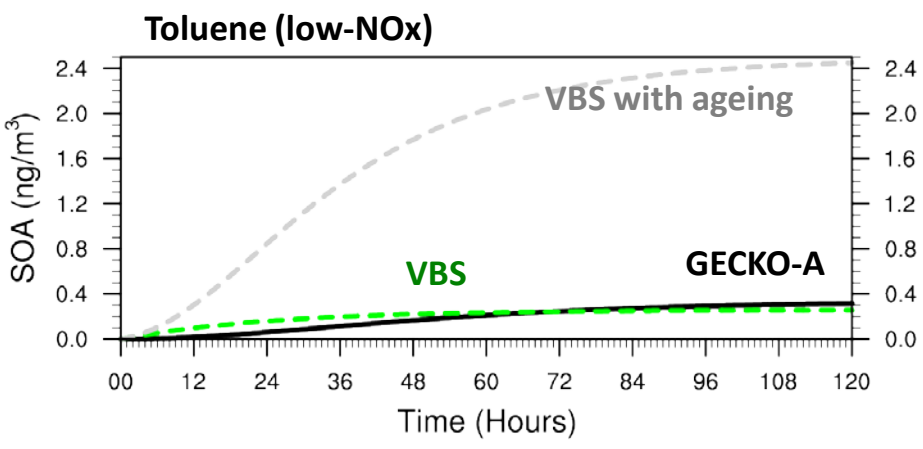
How do current parameterizations compare to explicit models?

GECKO-A (NCAR / CNRS, France) idealized runs:

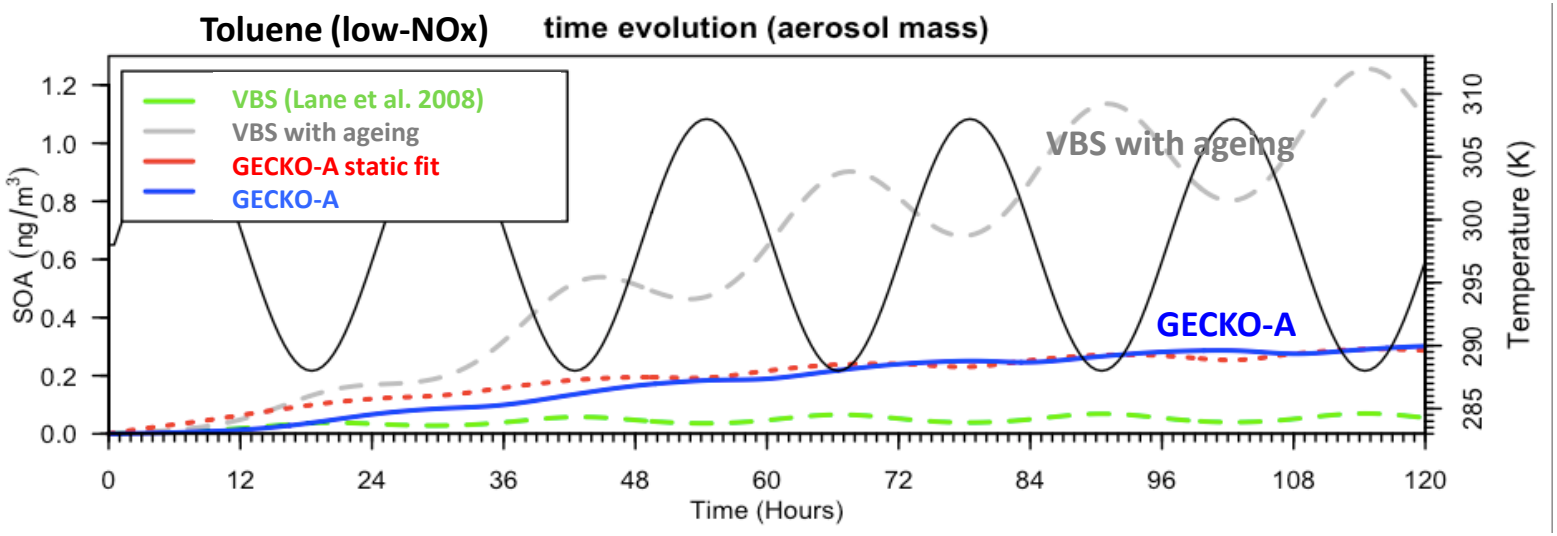
- VOC = 1ppt ; NOx (low) = 0.1 ppb
- OH = $2*10^6$ molec./cc ; seed OA : 1µg/m^3

1. Unconstrained SOA yields



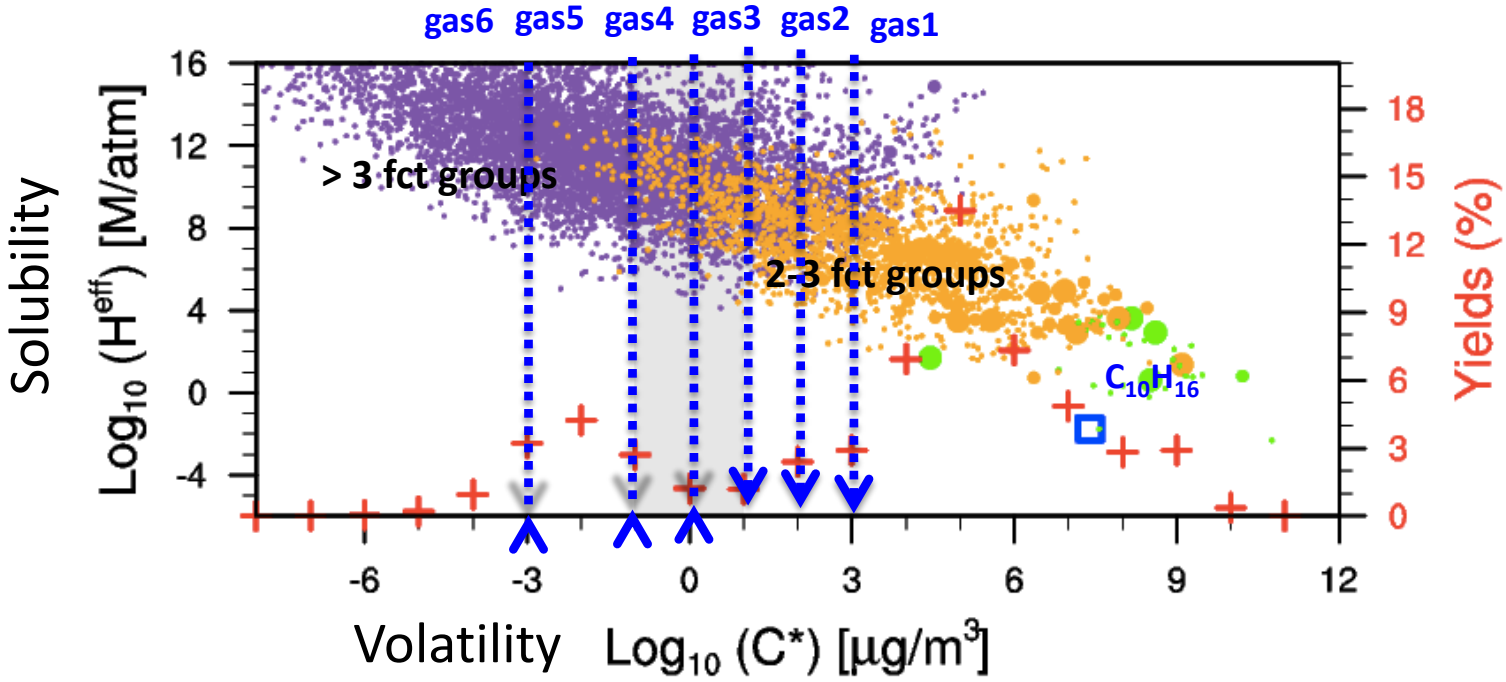


2. Too Volatile oxidation products (will evaporate with Temp. and dilution)



Fitting a reduced mechanism to GECKO-A outputs

- Idealized simulation for a-pinene +OH



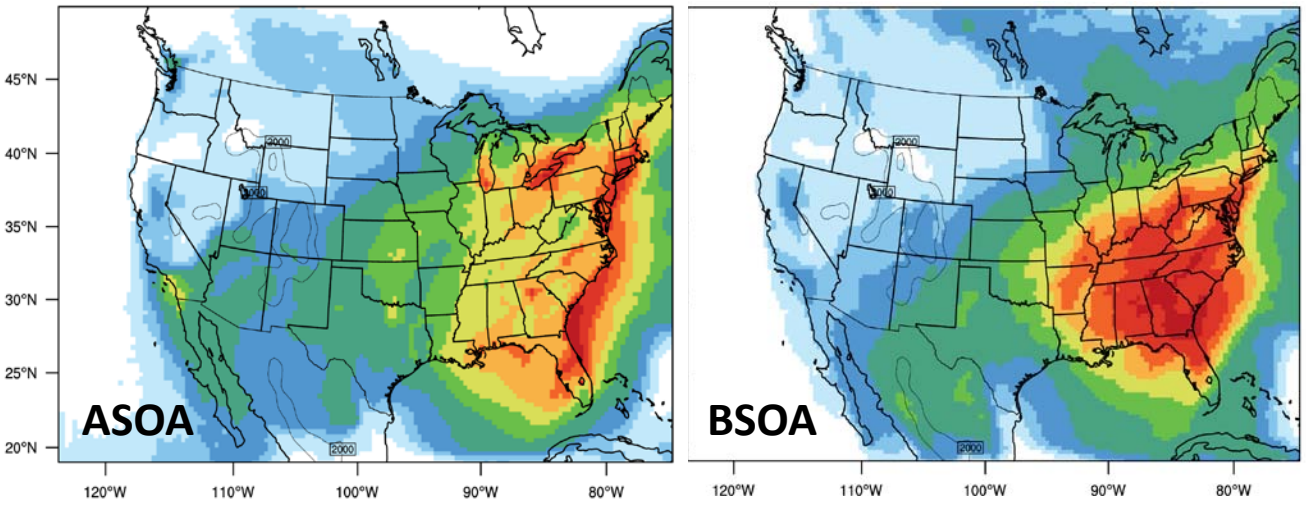
Static Yields -> parameterize GECKO overall yields for a given precursor

$C_{10}H_{16}$ + OH → 0.13 gas1 + 0.02 gas2 + 0.01 gas3 + 0.012 gas4 + 0.07 gas5 + 0.045 gas6

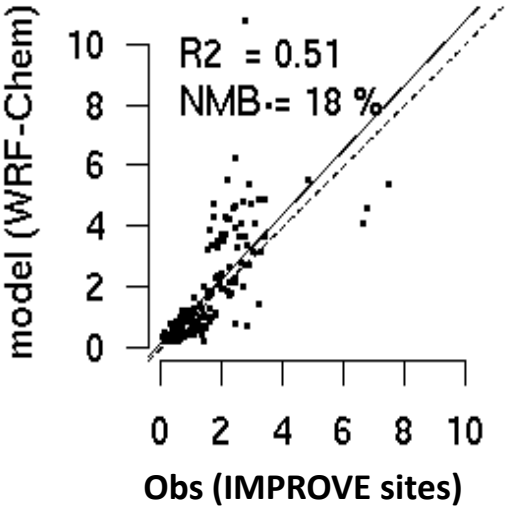
⇒ Easy to implement into 3D models, and should capture the regional SOA mass production suggested by GECKO-A

WRF-Chem simulations of surface SOA (June, 2010)

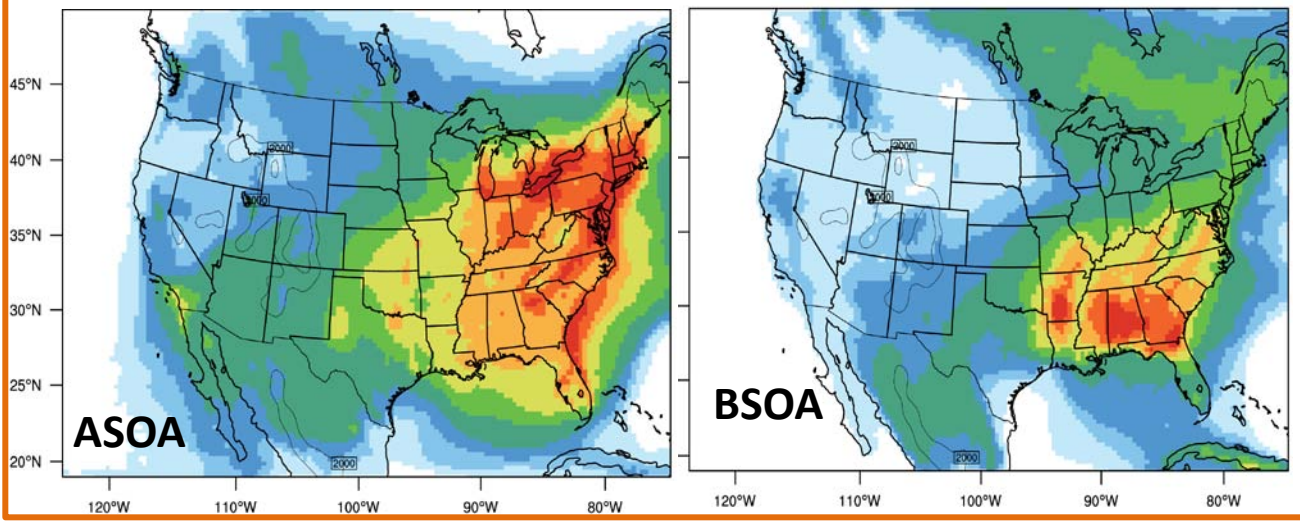
- VBS with ARTIFICIAL ageing of anth/biog

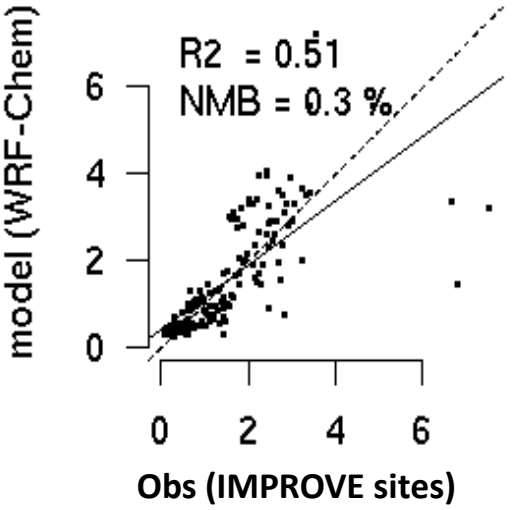


Org. Carbon (µg/m^3)



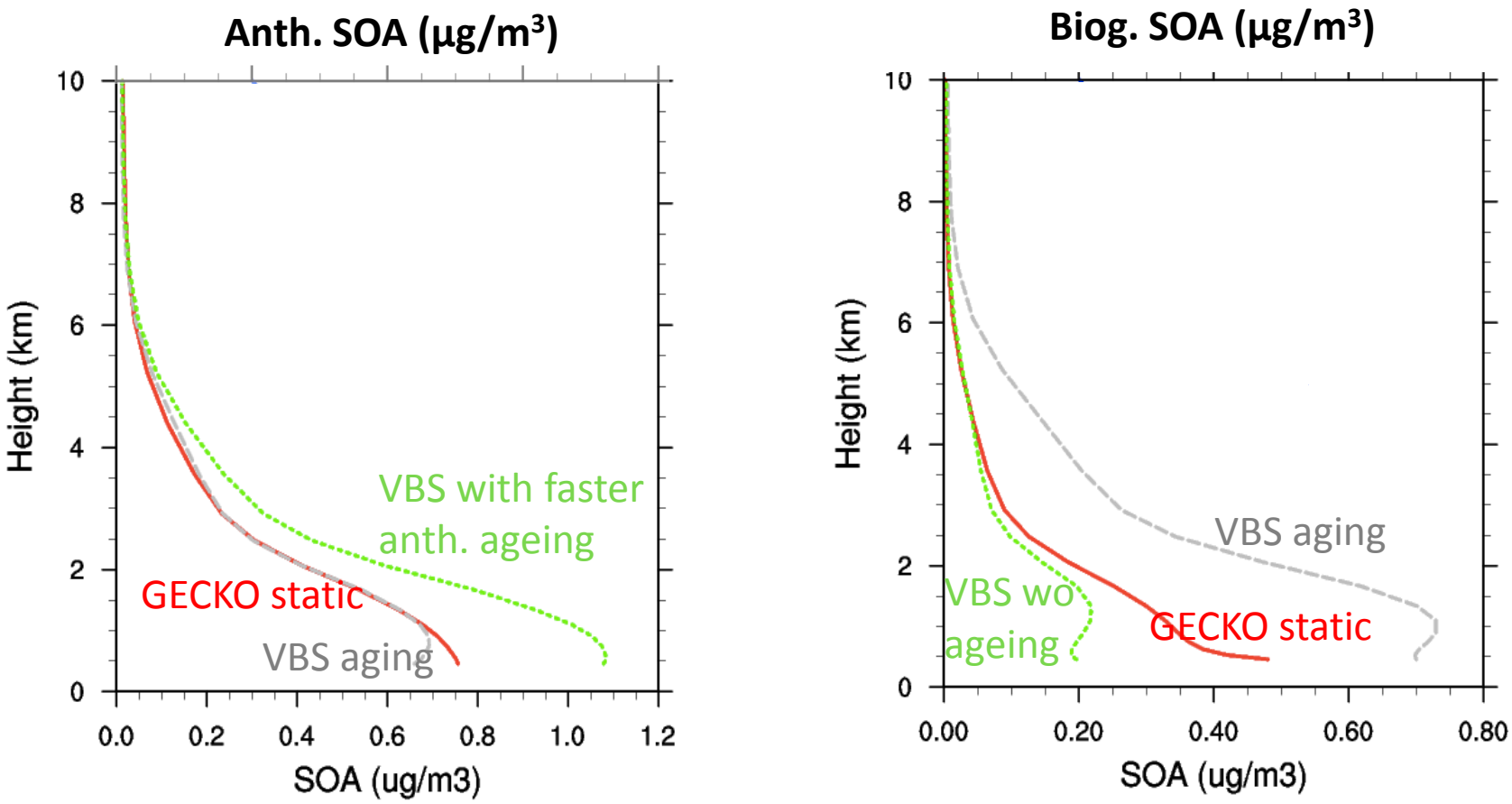
- GECKO – static fit





GECKO-A vs. current parameterizations

WRF-Chem : SOA domain-averaged vertical profiles (June, 2010)



GECKO-A and VBS with ageing

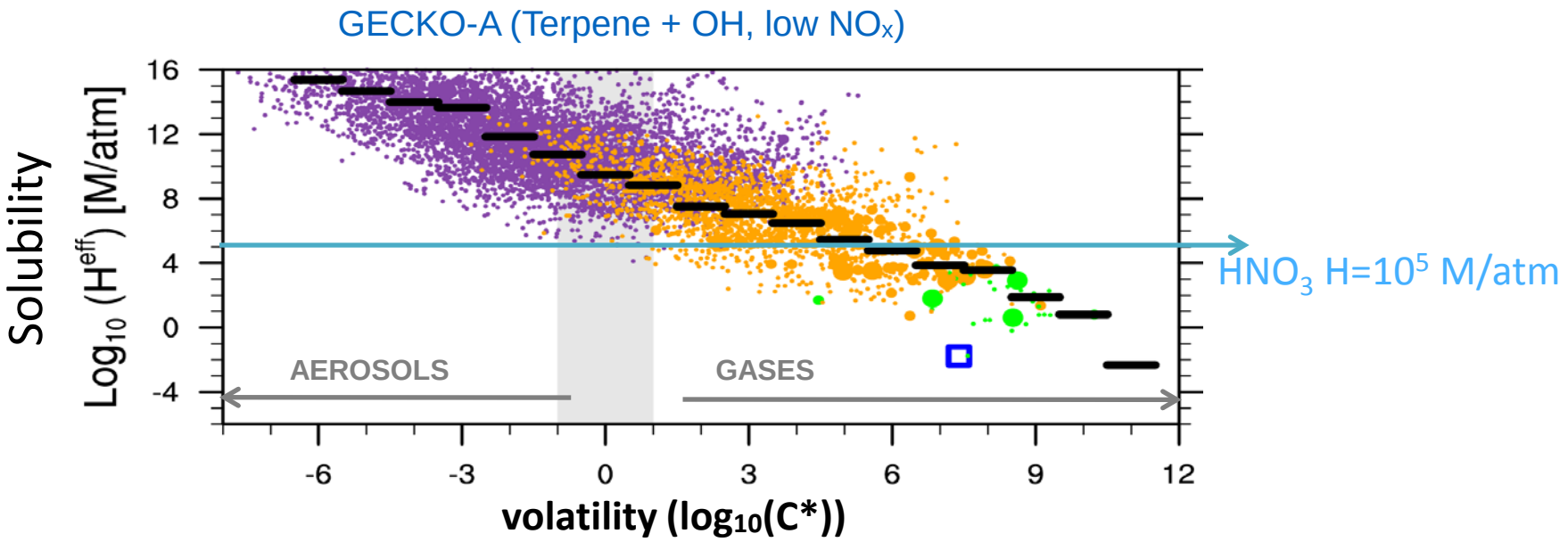
- can provide similar mass,
- but proportions of anthropogenic and biogenic species are different

⇒ **Will predict different future!**

Condensable organic compounds are highly water soluble

[*Hodzic et al. GRL submitted*]

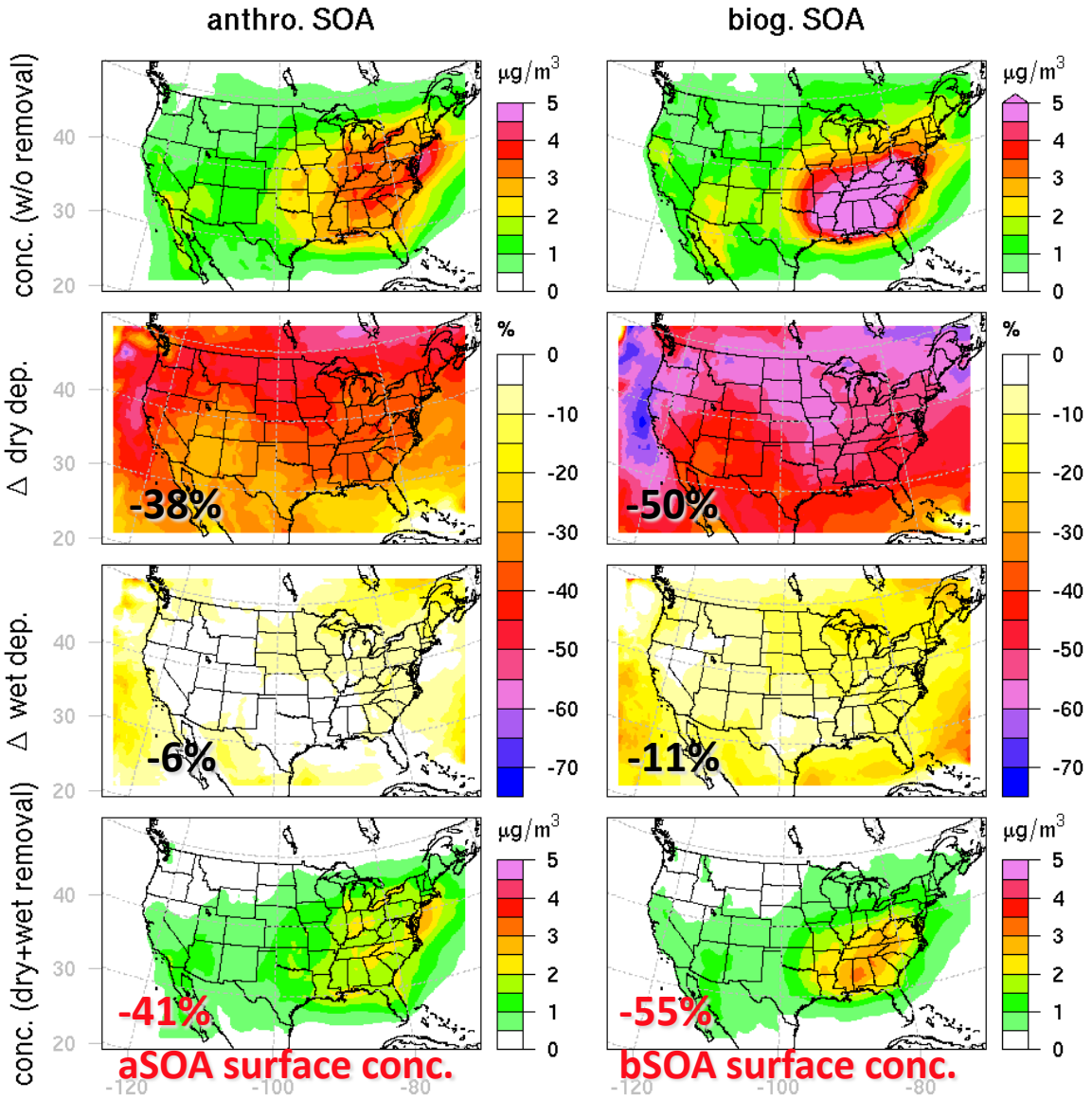
- Organic vapors dry and wet deposit depending on their solubility (Henry's law coef.)
- Solubility is unknown for condensable organic vapors
- Crudely represented in 3D models (as HNO_3 $H=10^5$ M/atm)



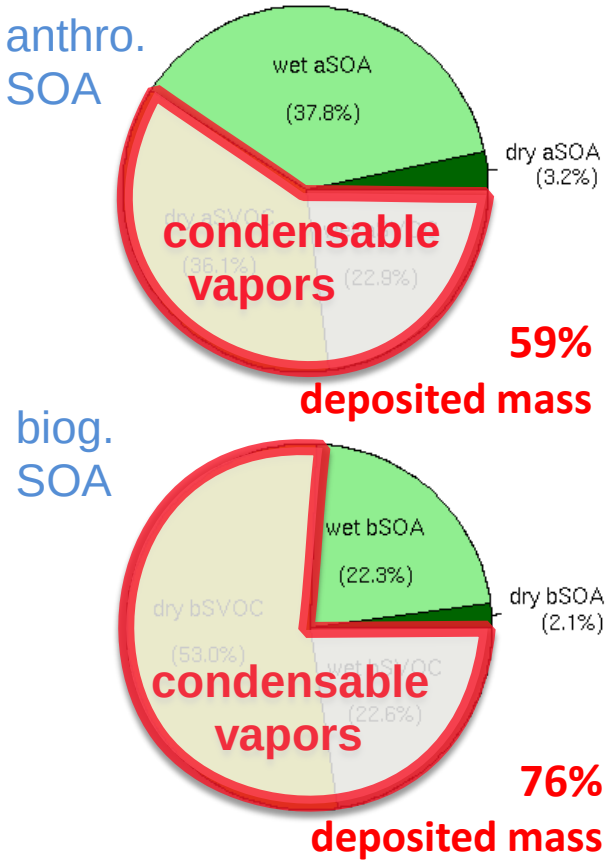
SOA removal mediated by dry & wet deposition of condensable vapors

Summer surface concentrations (VBS with ageing)

[*Hodzic et al. GRL submitted*]



Accumulated deposition



Summary and ongoing work

Regional SOA growth & removal

- strong growth for anthropogenic precursors
- highly soluble oxidation products sensitive to dry and wet deposition

=> need to re-estimate the radiative impact of anthropogenic sources

Current parameterizations VBS vs. explicit modeling

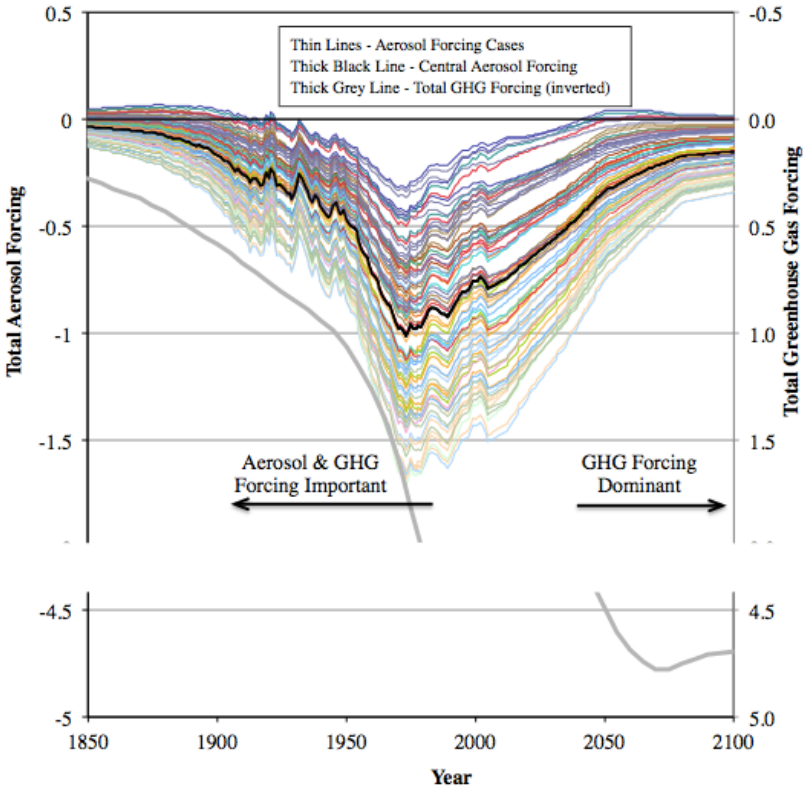
- VBS with aging can be tuned to represent SOA observations however, ageing ARTIFICAL not constrained by chamber experiments,
- GECKO-A predicts less volatile, and more soluble species than VBS

Fitting parameterizations to GECKO-A

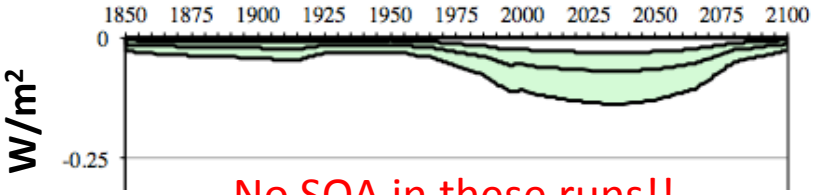
- simple static fit for yields compares well with data, but needs more testing at both regional (WRF-chem) and global (CESM) scales.
- parameterization of the water solubility provides a constraint on the removal

Should we still worry about Organic Aerosols impact on climate?

Global Aerosol Forcing (RCP4.5)

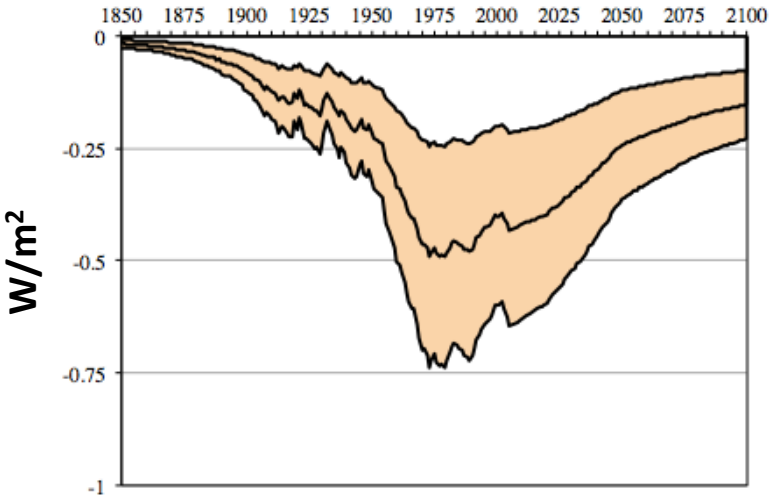


Organic Carbon Direct Forcing



No SOA in these runs!!

Sulfate Direct Forcing



[*Smith and Bond*, 2013]

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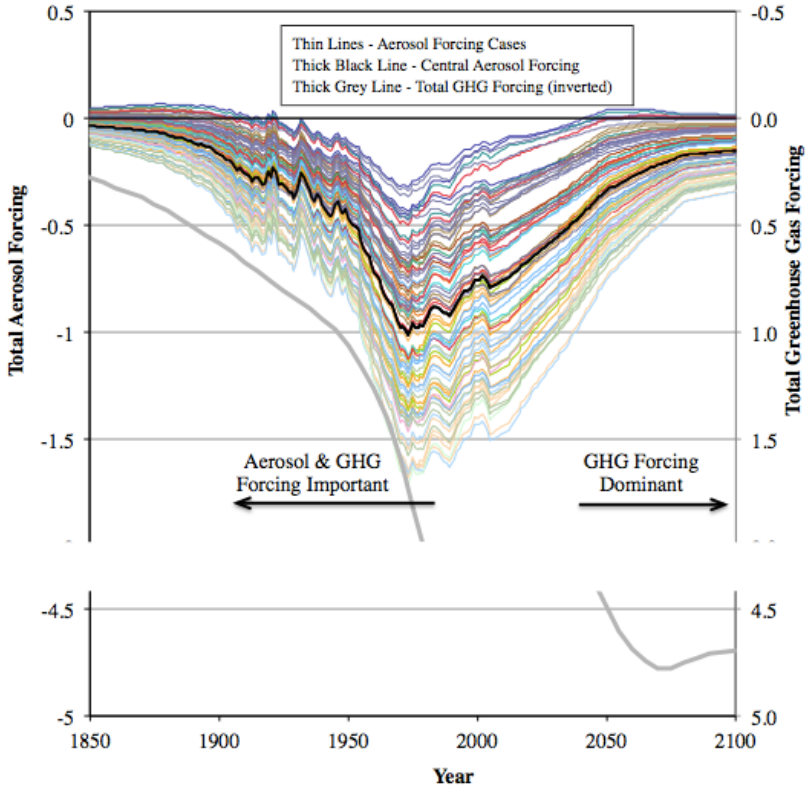
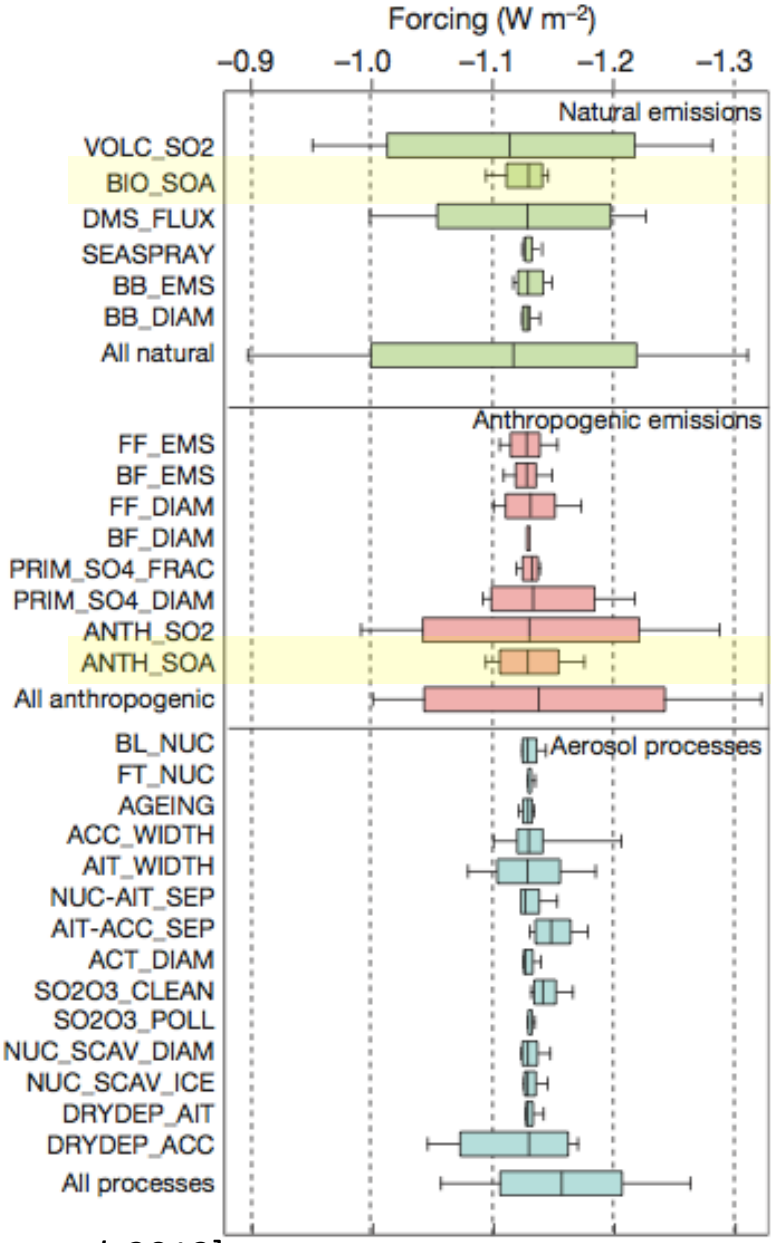




[*Carslaw et al.* 2013]

How large is the anthropogenically controlled SOA?

Simple estimate from SOA - Ox Correlations

Slopes from 30 to 160 μg m^{-3}/ppm

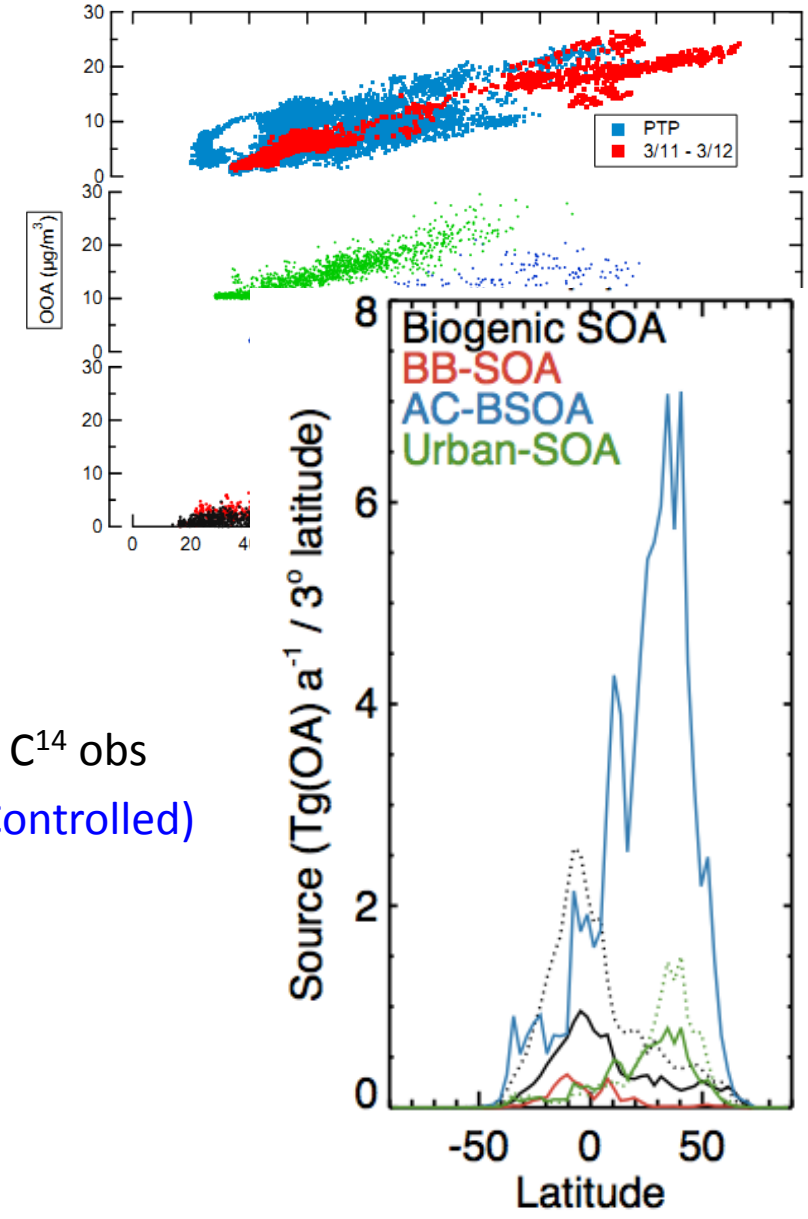
~25 ppb O_3 change (NH preindustrial to present):

- Slopes imply 0.75 to 4 μg m^{-3}

  ~ 0.3 - 2.0 Tg (2 km PBL)

  ~ 20-145 Tg yr^{-1}

[*Wood et al.* 2010]



Other estimates:

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  - Production: 88 Tg yr^{-1}
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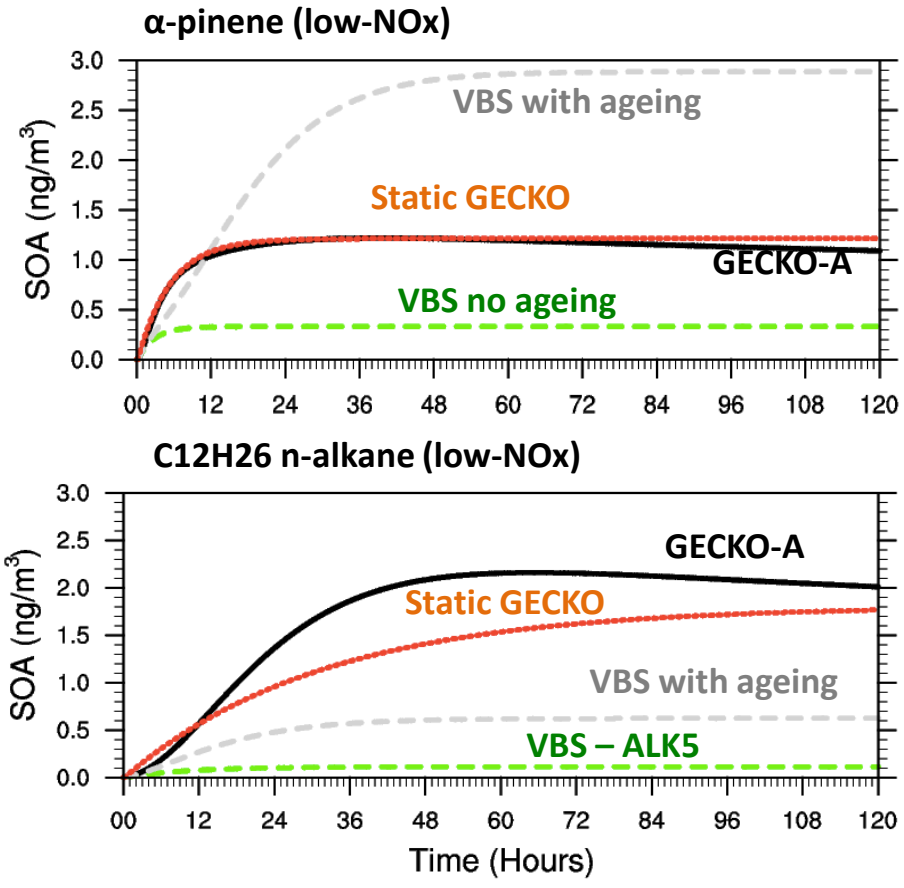
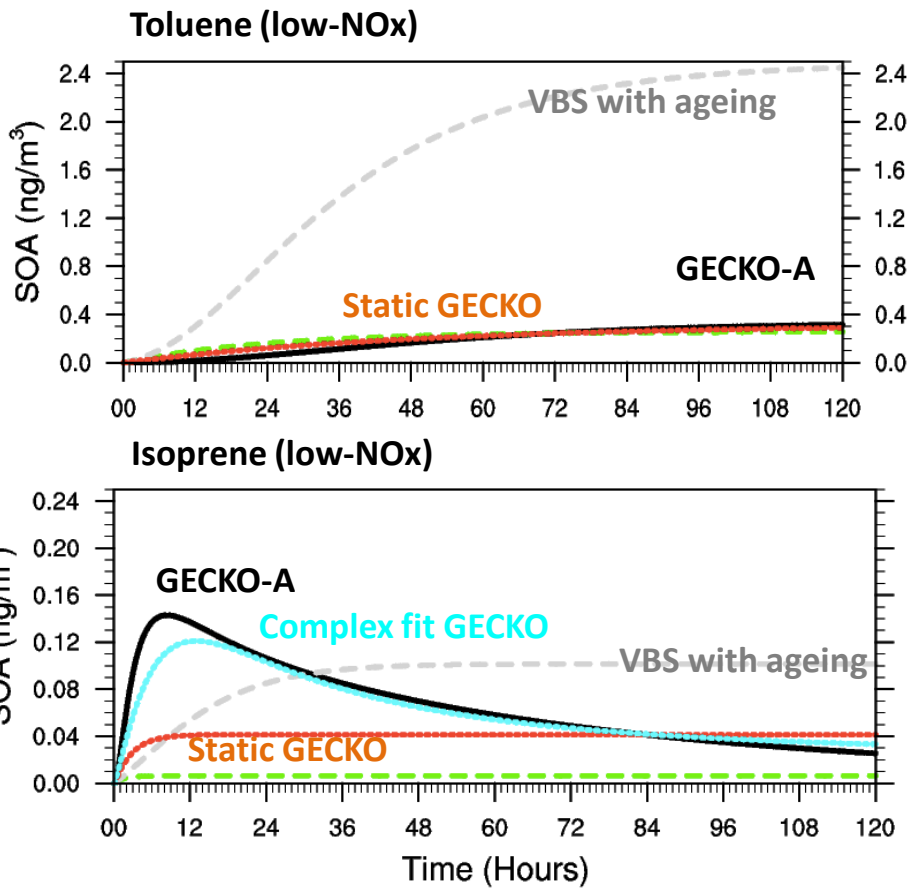
Static parameterization based on GECKO-A





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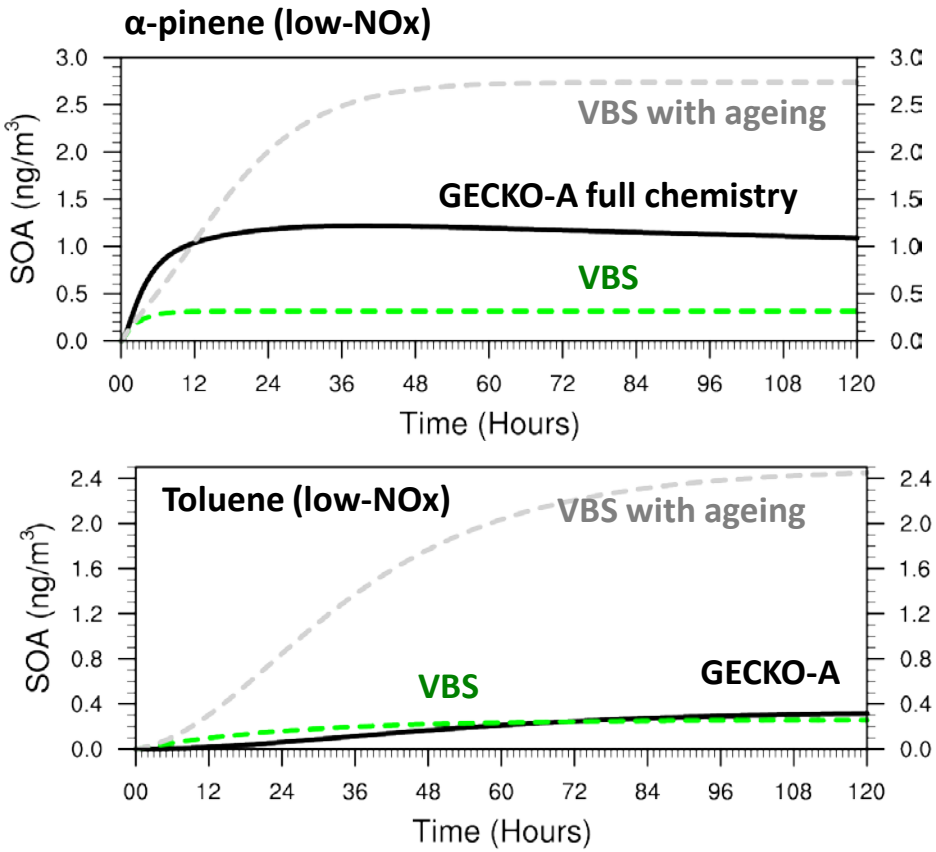

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