Secondary Organic Aerosols: Will they change in the Future?

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*now at PNNL

Winter CESM Working Group meeting; February 8-11, 2016, Boulder, CO

Thanks to Sponsors:

NASA Atmospheric Composition: Modeling and Analysis Program

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One of the strongest feedbacks between climate change and chemistry involves the formation of biogenic Secondary Organic Aerosols

- Biogenic VOC emissions increase with temperature, but also depend on soil moisture, atmospheric composition, vegetation type
  - 20-55% increase based on climate effect alone leads to a 26-150% increase in SOA (*Liao et al.,* 2006; *Tsigaridis and Kanakidou*, 2007; *Heald et al.,* 2008;
- Could act to moderate the overall future warming via aerosol direct and indirect effects

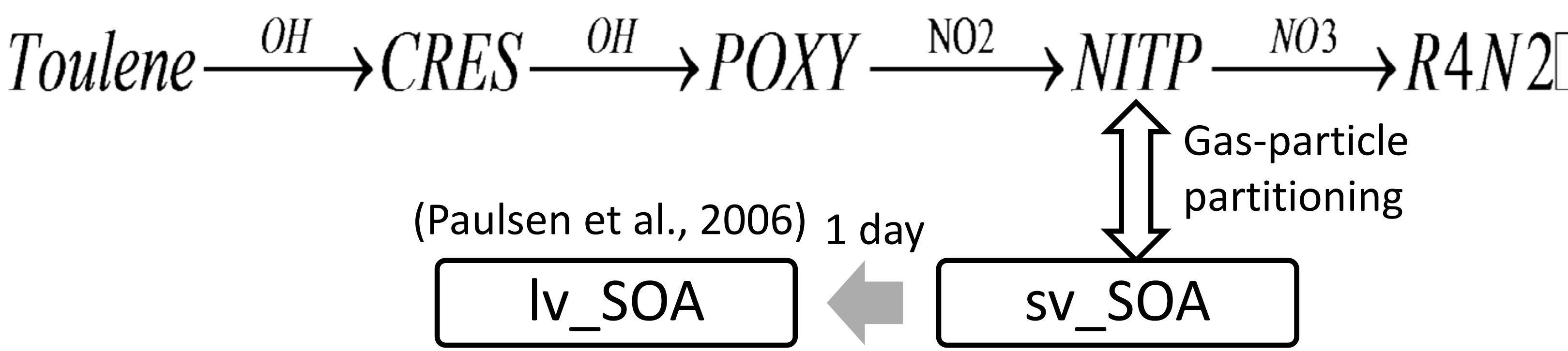
However some feedbacks act to decrease biogenic VOCs and SOA

- Increasing CO_2 inhibits isoprene emissions
- Land use change may decrease emissions
- Decreases in anthropogenic emissions (SO_2) may inhibit formation of SOA

Previous model estimates did not use an explicit mechanism for SOA formation

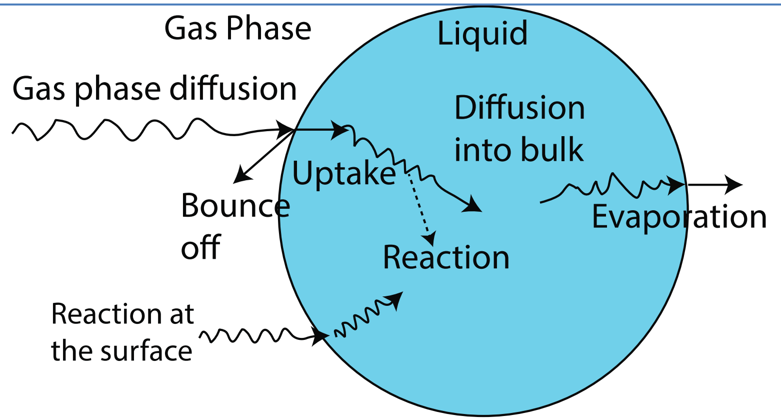
CESM v1.2.2 with CLM4 land model and embedded chemical transport model (IMPACT)

- Explicit gas phase chemistry
  - Basic photochemistry of O_3, OH, NO_x and VOCs (Ito et al., 2007).
  - Epoxide formation from isoprene (Paulot et al., 2009).
  - HO_x regeneration through isoprene oxidation proposed by Peeters et al. (2009) but with a recycled rate reduced by a factor of 10 (Karl et al. 2009).
- SOA formed from gas-particle partitioning of semi-volatiles (Pankow 1994). For example,

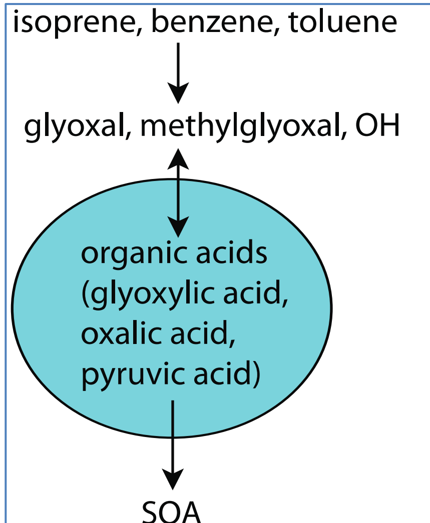


SOA formation mechanisms

- SOA formed from the cloud processing of gloyxal and methylglyoxal



Schematic diagram of uptake and reaction of gases in liquid. Based on Finlayson-Pitts and Pitts, (2000)



Lin et al. 2014: Global modeling of SOA: The use of different mechanisms for aqueous phase formation, Atmos. Chem. Phys.

SOA formation mechanisms

- SOA formed from the reactive uptake of glyoxal, methylglyoxal and epoxide onto sulfate aerosol

$$\frac{dC_{SOA}}{dt} = \frac{-1}{4} \cdot \gamma \cdot A \cdot < \nu > \cdot C_{gas}$$

γ: reactive uptake parameter.
A: surface area of aqueous sulfate aerosols
Major products: oligomers and organosulfate

Lin et al. 2012: Global modeling of SOA formation from dicarbonyls, epoxides, organic nitrates and peroxides, Atmos. Chem. Phys.

Scenarios

- Biogenic emissions based on MEGAN 2.1

Species	Present day (year 2000)	Future (year 2100)
Isoprene	440	534
$C_{10}H_{16}$	131.9	246.5
PRPE(>=C4 alkenes)	14.0	24.3
Methanol	85.0	159.4
Acetone	38.6	67.2
C_2H_4	23.4	46.0
CO	64.8	127.4
HCHO	4.1	8.7

Scenarios: anthropogenic 2100 emissions based on RCP 8.5

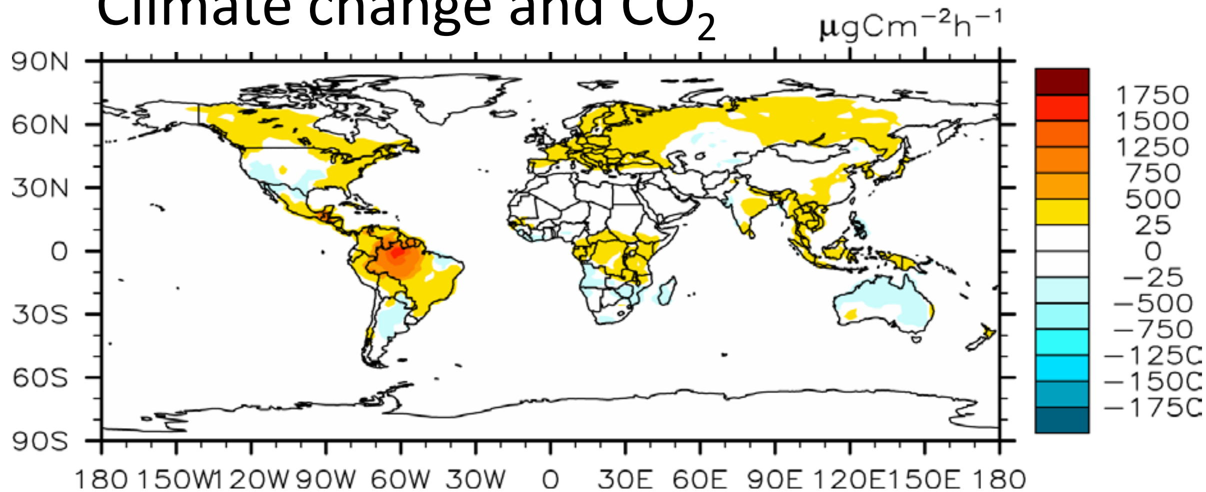
Species	Present day (year 2000)	Future (year 2100)
SO_2	129.2	49.5
NO	88.6	65.5
CO	956.7	711.0
NH_3	64.8	92.1
ALK4(>=C4 alkanes)	35.2	15.1
HCHO	3.2	5.5
ALK7(C6-C8 alkanes)	38.5	16.5
Aromatics	31.7	37.4
HCOOH	7.0	8.7
Acetic acid	8.2	11.4
Acetone	2.5	4.2
POA	49.8	33.2

Experiment design: Separate effects of climate, anthropogenic emissions, land use

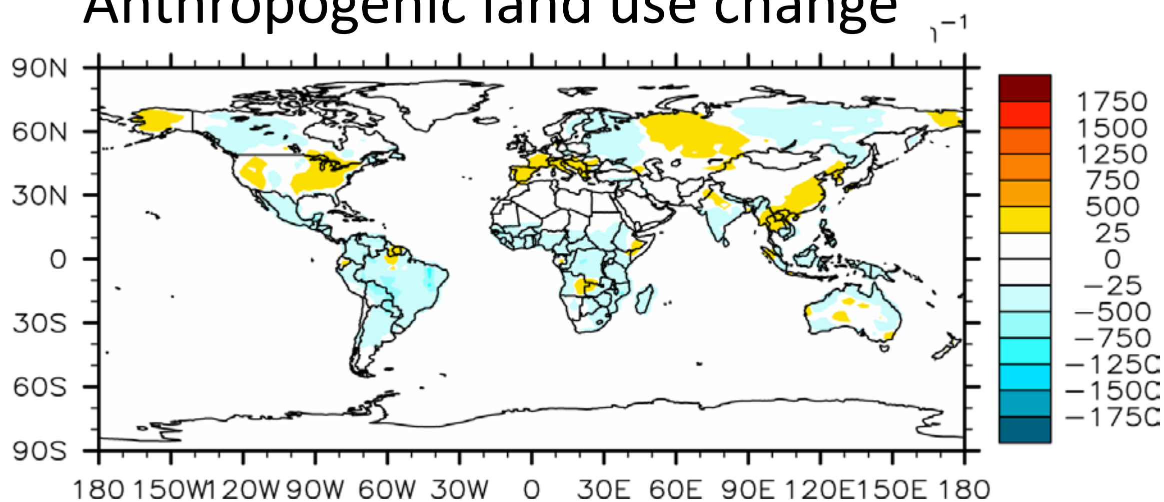
- Change in anthropogenic emissions
- Change in climate and CO_2 concentrations
- Change in anthropogenic land use
- All effects acting in combination
- Examine effects of changing particle acidity, which lowers uptake of IEPOX

Effects on isoprene emissions

Climate change and CO_2

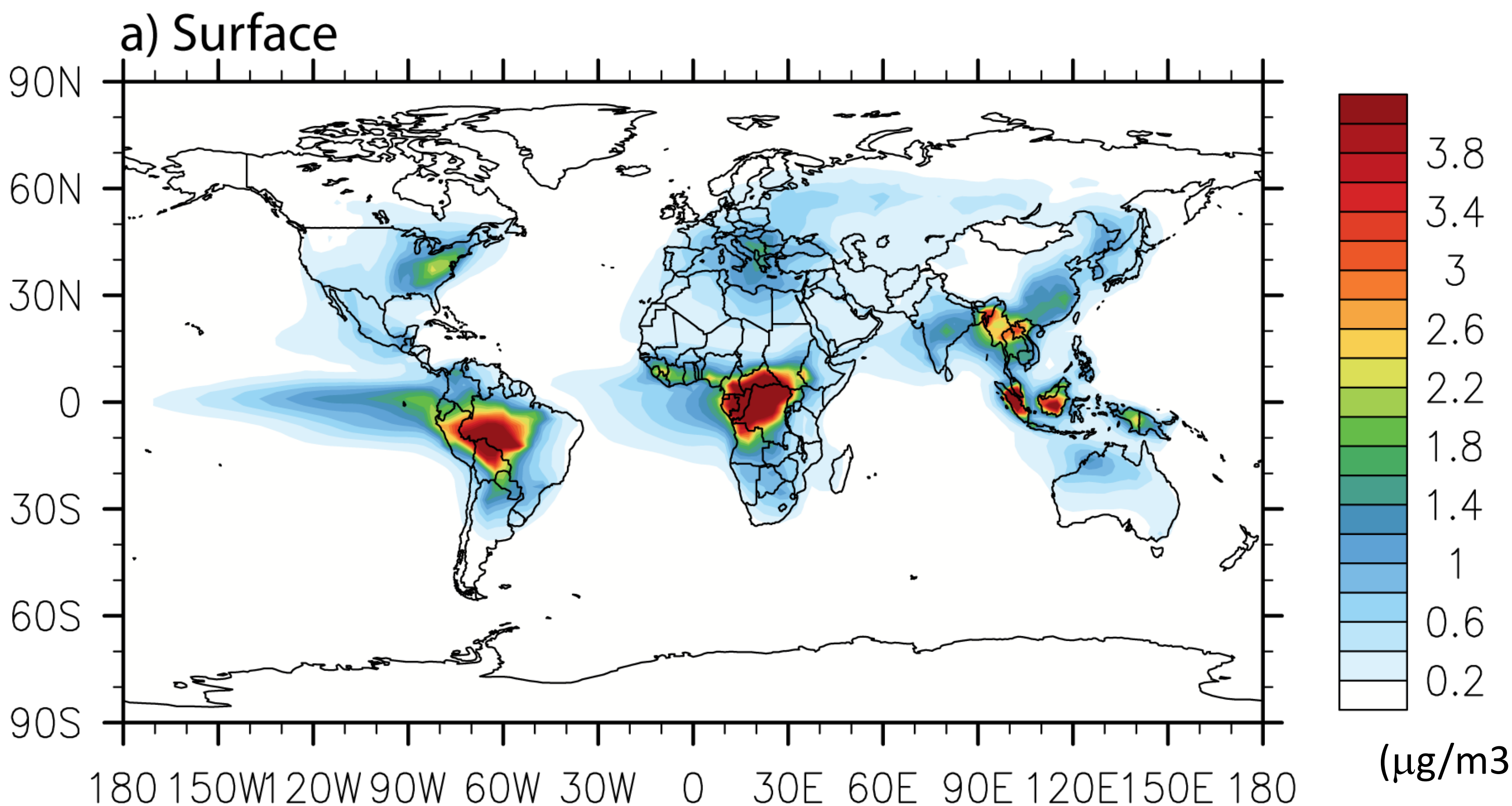


Anthropogenic land use change

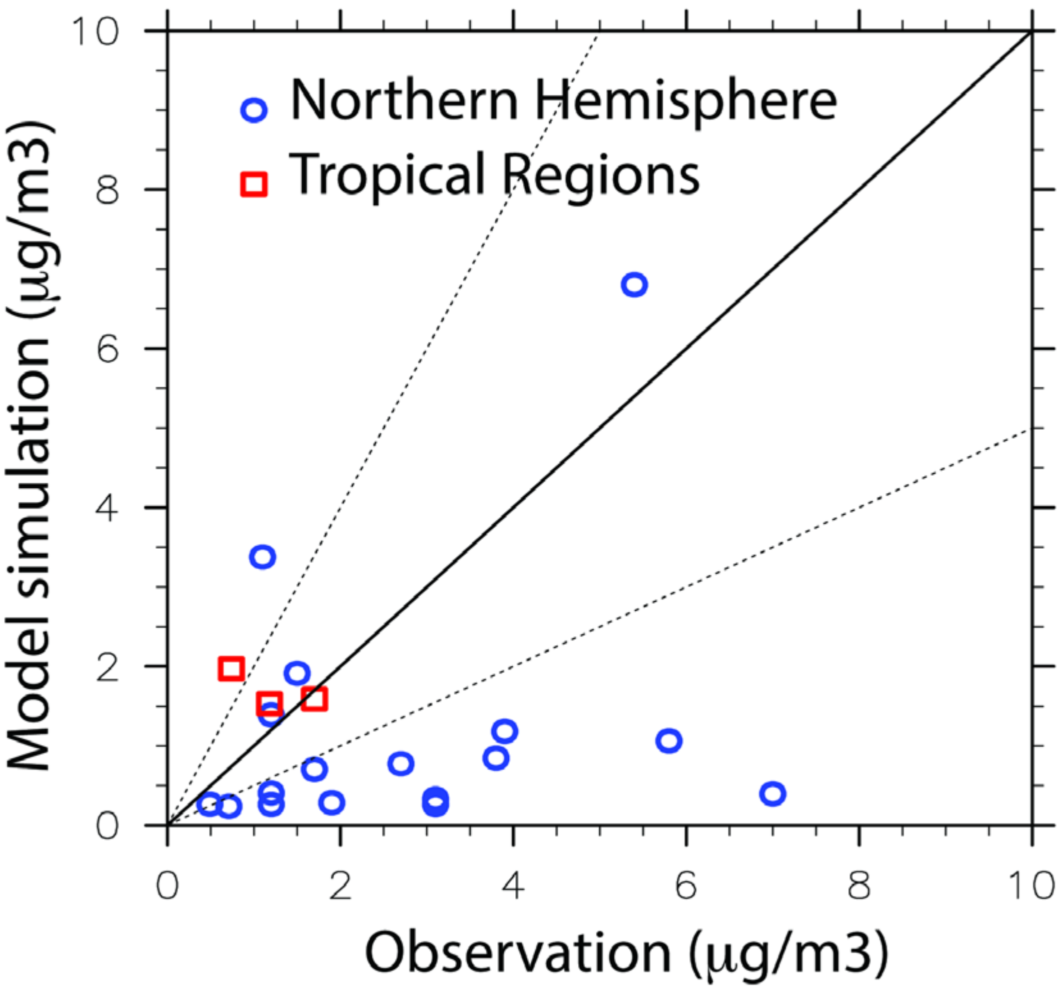


Today's SOA burden is 1.06 Tg, 80% from oxidation of isoprene; 11% from anthropogenic emissions

Present-day SOA concentration

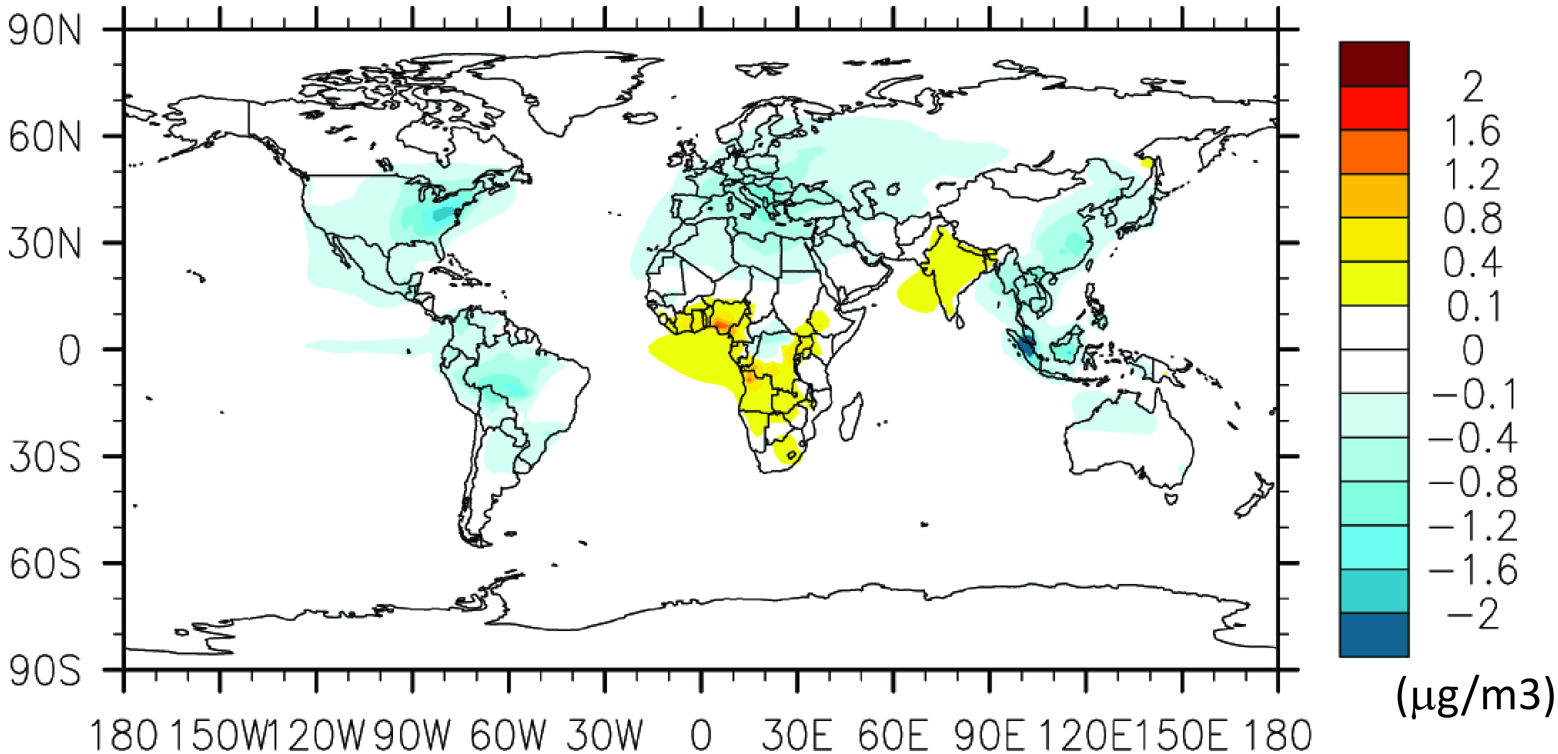


Predicted SOA is lower than AMS observations by 57% in NH and higher by 41% in tropical regions



Only small changes when acidity of aerosols for IEPOX uptake: Decrease of 2% near surface Increase of 2.5% above surface.

Effect of anthropogenic emissions changes: -2%

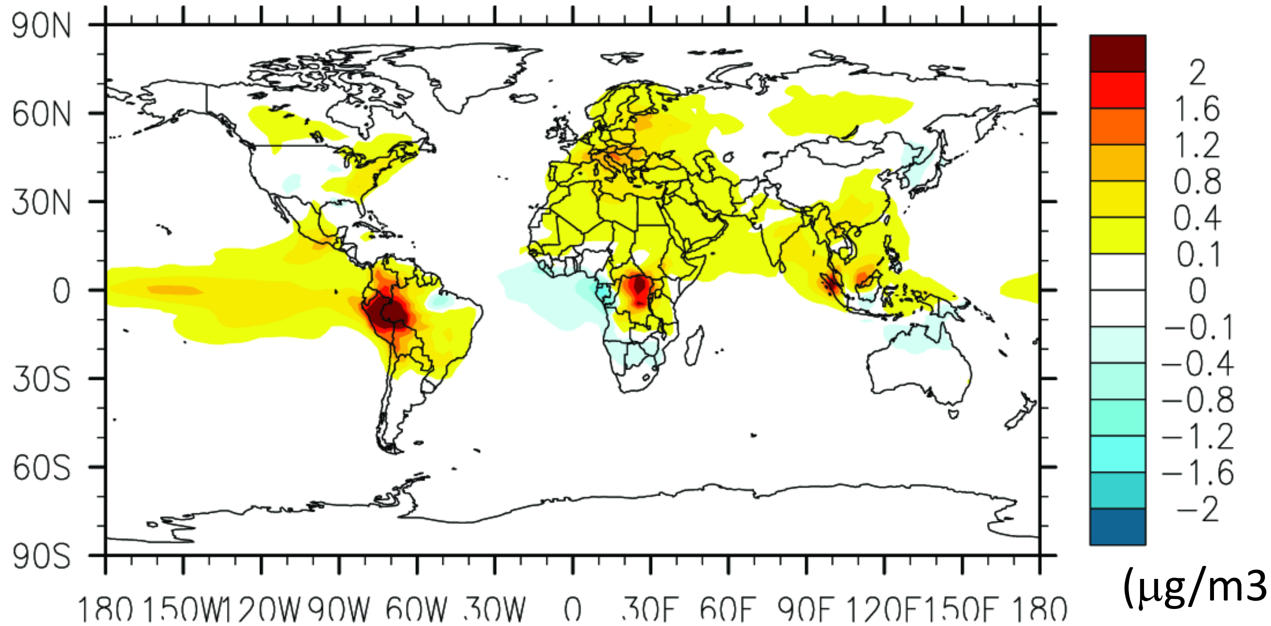


Net effect:

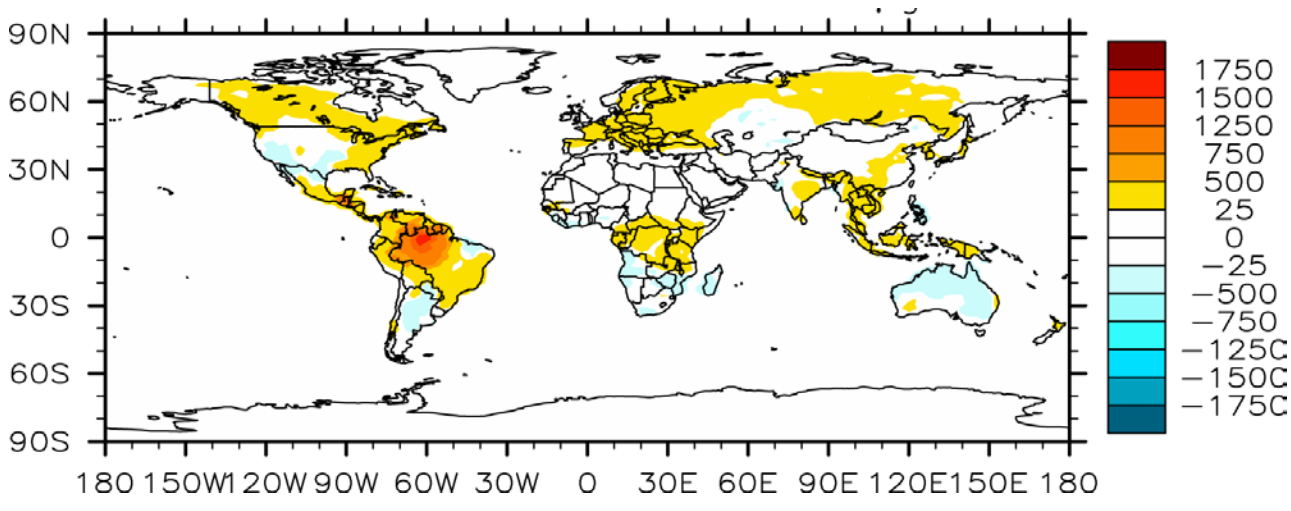
Increases from gas-particle partitioning (0.06 Tg)

Decrease from uptake of glyoxal, methylglyoxal and IEPOX (-0.1 Tg)

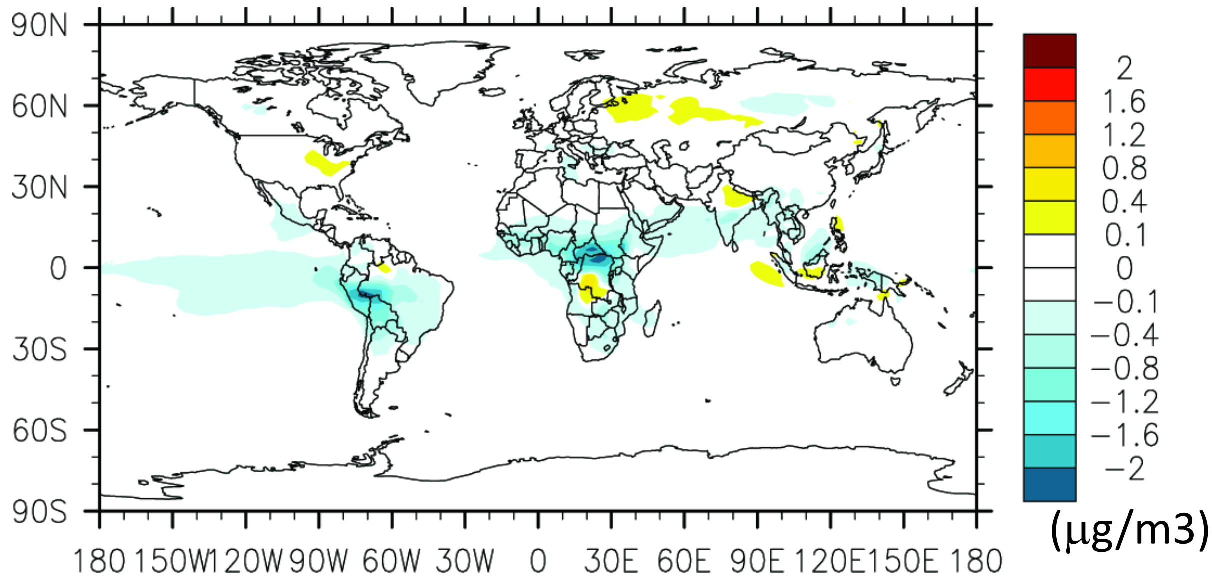
Effect of climate and CO_2 change: +25%



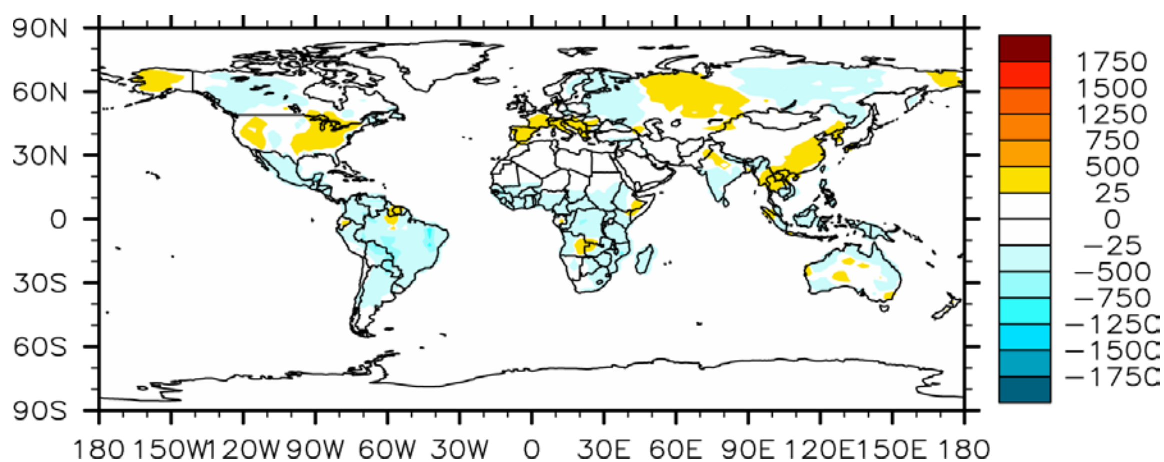
Effect of climate change and CO_2 on isoprene emissions (μgC/m2/h)



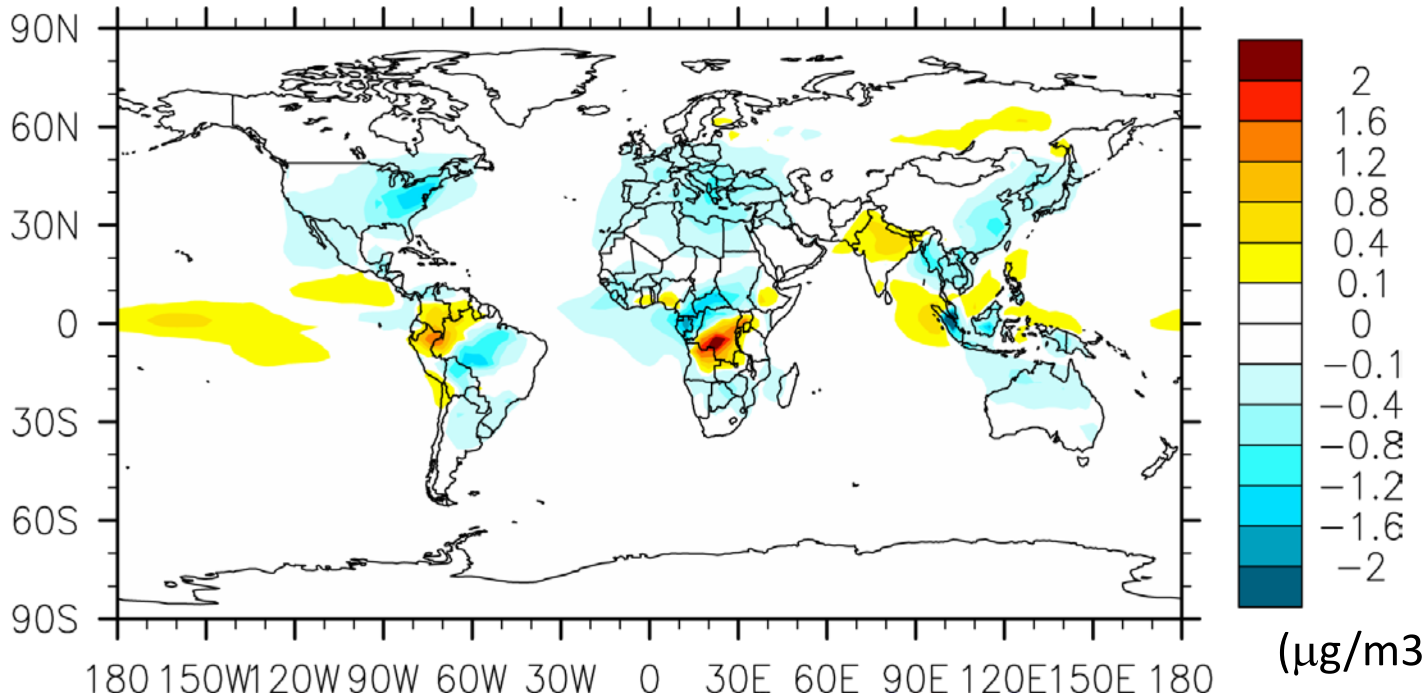
Effect of anthropogenic land use change: -14%



Isoprene emissions change due to anthropogenic land use change



Combined effects: 2%



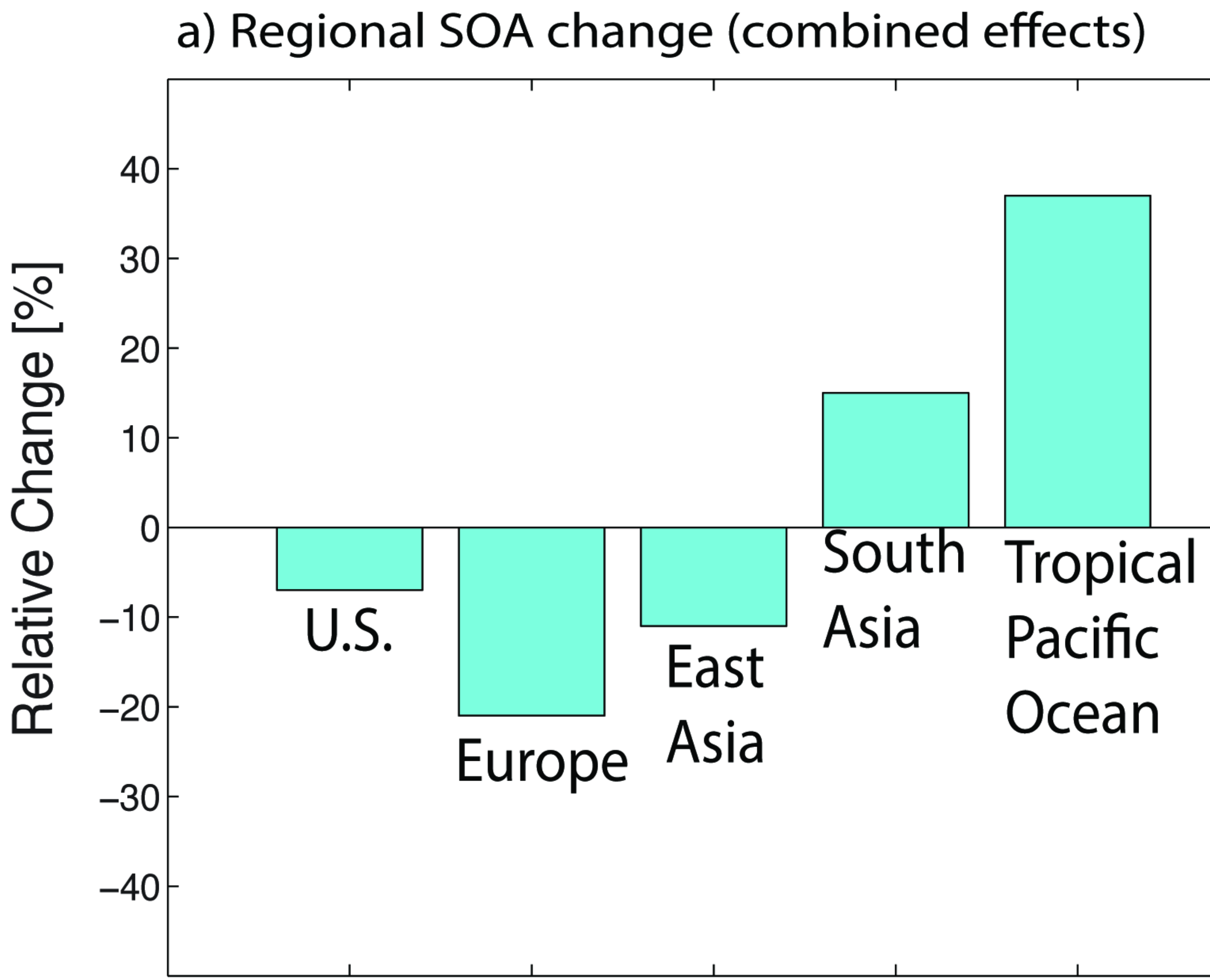
Large increase anticipated from change in climate offset due to:

Decrease in isoprene from CO_2 inhibition

Decrease in anthropogenic emissions of SO_2

Decrease in isoprene from anthropogenic land use change

Regional variations in SOA can be large:



Thank you!