Measurement-Based Constraints on the Regional and Global Secondary Organic Aerosol Budgets

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Based on two papers:

- *D.V. Spracklen, J.L. Jimenez, K.S. Carslaw, D.R. Worsnop, M.J. Evans, G.W. Mann, Q. Zhang, M.R. Canagaratna, J. Allan, H. Coe, G. McFiggans, A. Rap and P. Forster. Aerosol mass spectrometer constraint on the global secondary organic aerosol budget. Atmos. Chem. Phys. Discuss., 11, 5699-5755, 2011.*
- *A. Hodzic and J.L. Jimenez. Modeling anthropogenically-controled secondary organic aerosols in a megacity: a simplified framework for global and climate models. Geosci. Mod. Dev. Discuss., in press, 2011.*

DOE ASR Program Meeting

San Antonio, TX, March 29 2011

Funding Acknowledgements:

Photo by C. McNaughton – U Hawaii during MILAGRO 2006

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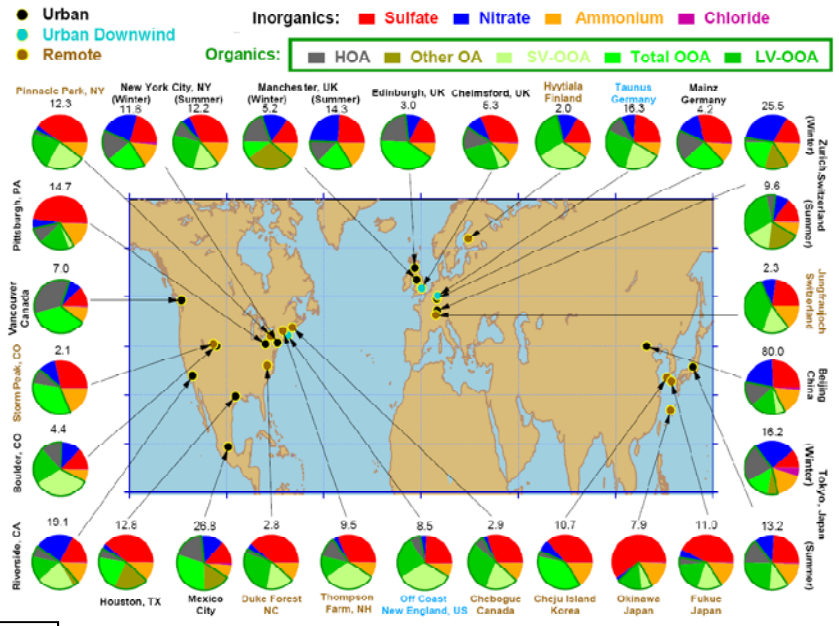
Worldwide OOA ~ SOA

Jimenez et al. Science 2009



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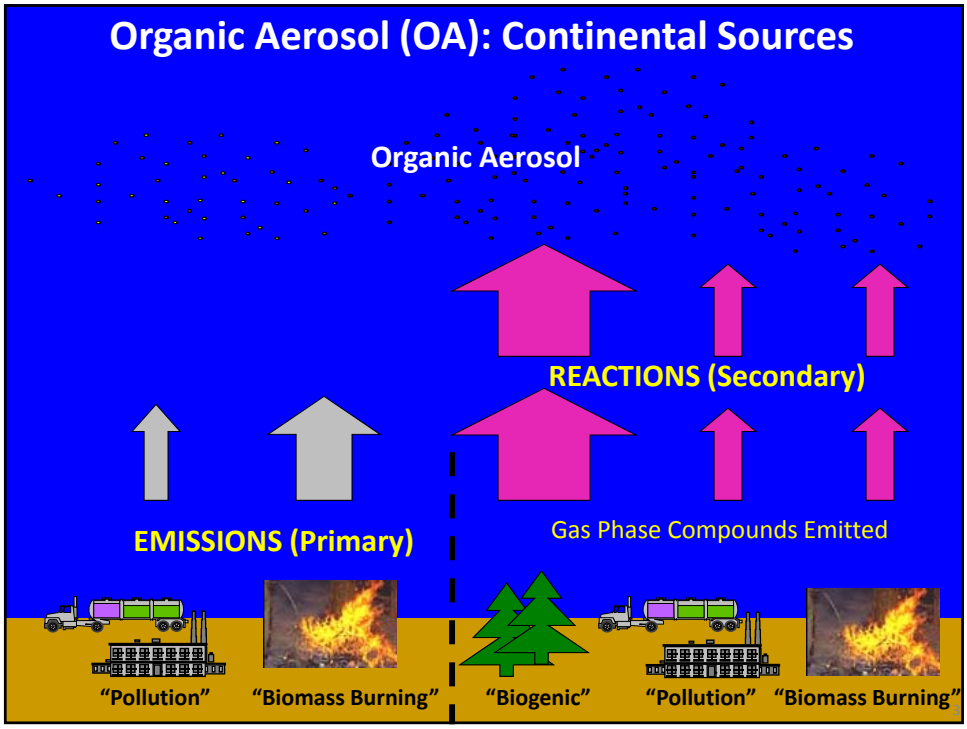
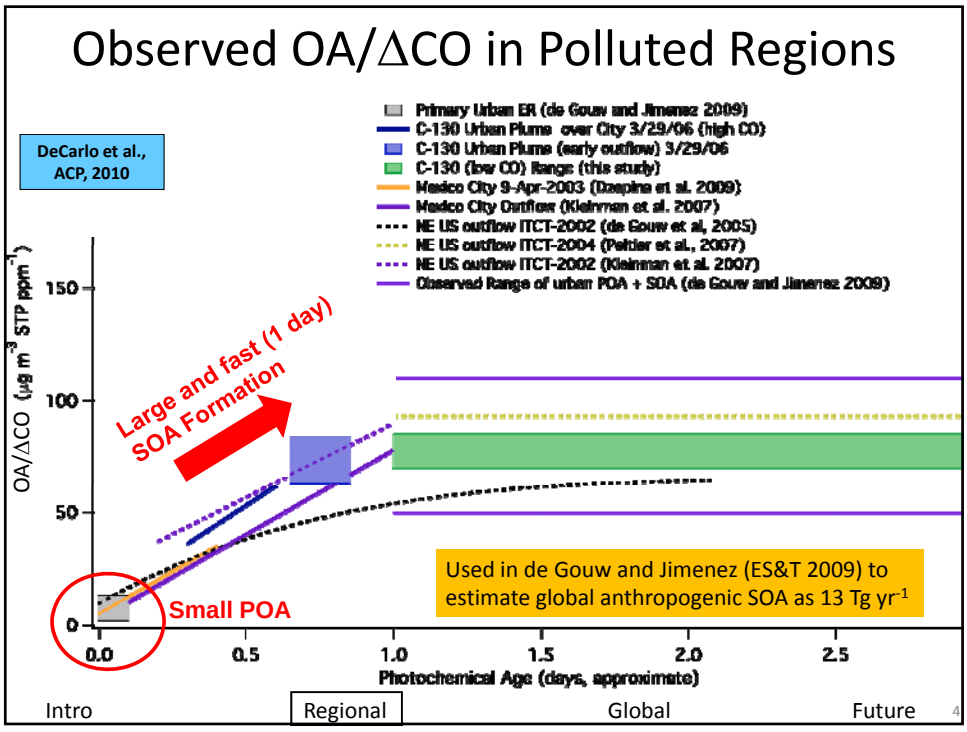

Organic Aerosol (OA): Continental Sources
Organic Aerosol
REACTIONS (Secondary)
EMISSIONS (Primary)
Gas Phase Compounds Emitted
"Pollution"
"Biomass Burning"
"Biogenic"
"Pollution"
"Biomass Burning"


Observed OA/ΔCO in Polluted Regions
DeCarlo et al., ACP, 2010
Primary Urban ER (de Gouw and Jimenez 2009)
C-130 Urban Plume over City 3/29/06 (high CO)
C-130 Urban Plume (early outflow) 3/29/06
C-130 (low CO) Range (this study)
Mexico City 9-Apr-2003 (Dzepina et al. 2009)
Mexico City Outflow (Kleinman et al. 2007)
NE US outflow ITCT-2002 (de Gouw et al, 2005)
NE US outflow ITCT-2004 (Peltier et al., 2007)
NE US outflow ITCT-2002 (Kleinman et al. 2007)
Observed Range of urban POA + SOA (de Gouw and Jimenez 2009)
OA/ΔCO (μg m-3 STP ppm-1)
150
100
50
0
Large and fast (1 day) SOA Formation
Small POA
Used in de Gouw and Jimenez (ES&T 2009) to estimate global anthropogenic SOA as 13 Tg yr-1
0.0
0.5
1.0
1.5
2.0
2.5
Photochemical Age (days, approximate)
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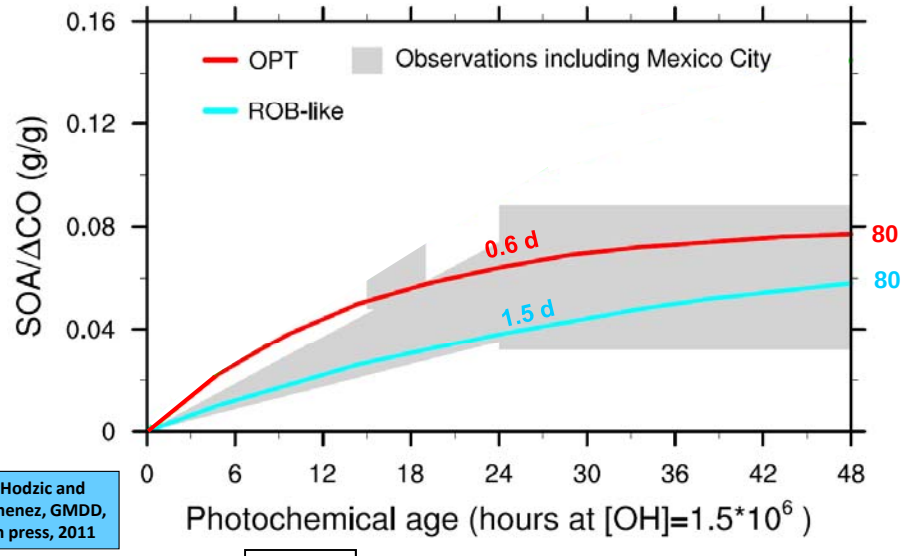
Parameterizing SOA/ΔCO

- Using only amount and timescale (k_{OH})



Hodzic and Jimenez, GMDD, in press, 2011

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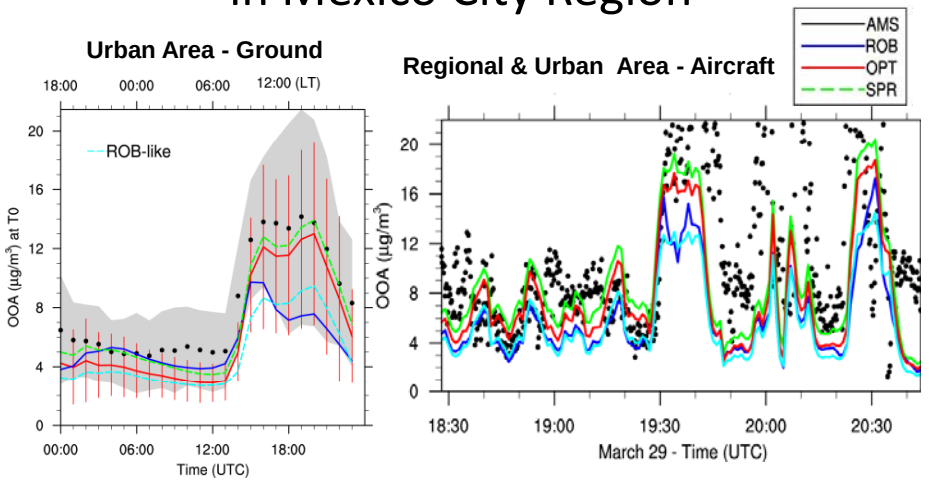
Performance of Parameterized SOA Model in Mexico City Region



- Performance is as good or better than for several more detailed recent models tested for Mexico!

Hodzic and Jimenez, GMDD, in press, 2011

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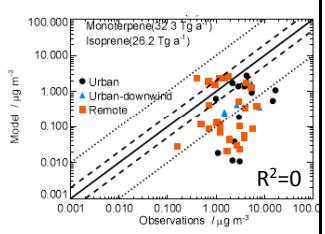
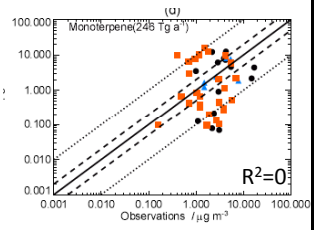
Global Model Optimization of SOA Sources

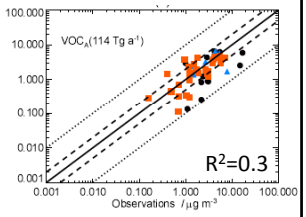
OOA: Model Base case vs. AMS



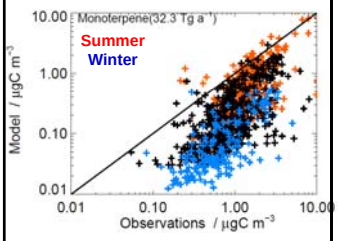
Increase Biogenic SOA

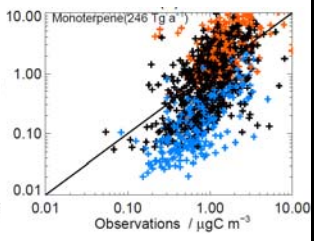


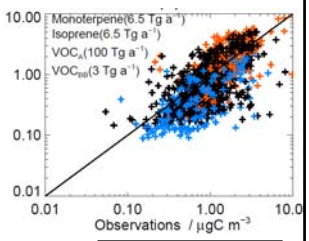
Increase Anthrop. SOA



OC Model Base case vs. IMPROVE







Spracklen, Jimenez, et al., ACPD, 2011

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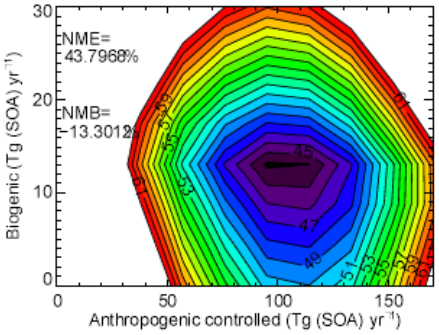
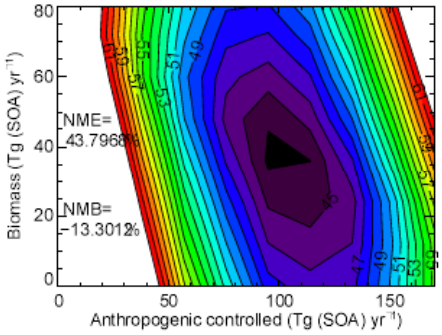
Global Model Optimization of Sources II





- Tune the sources in the model to get the best agreement with the observations
- Observation weights are tuned with VOC distributions
  - More weight to unpolluted locations

Spracklen, Jimenez, et al., ACPD, 2011

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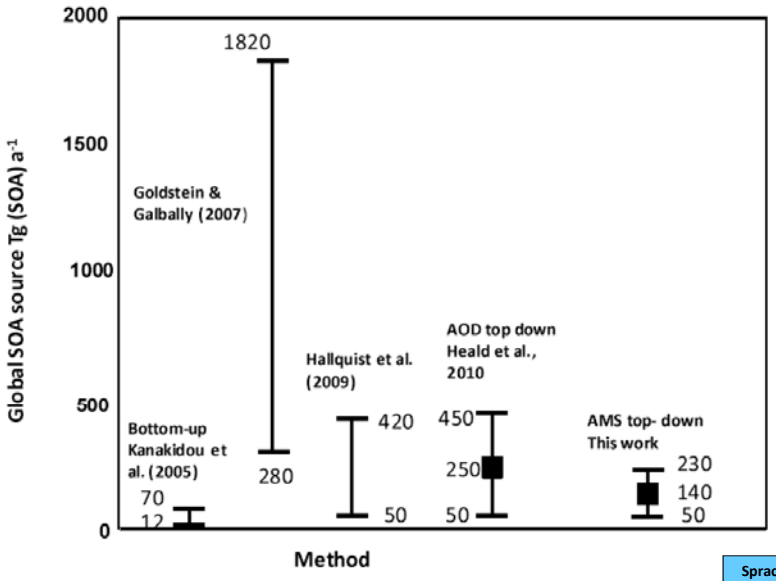
Summary of Global OA Budget

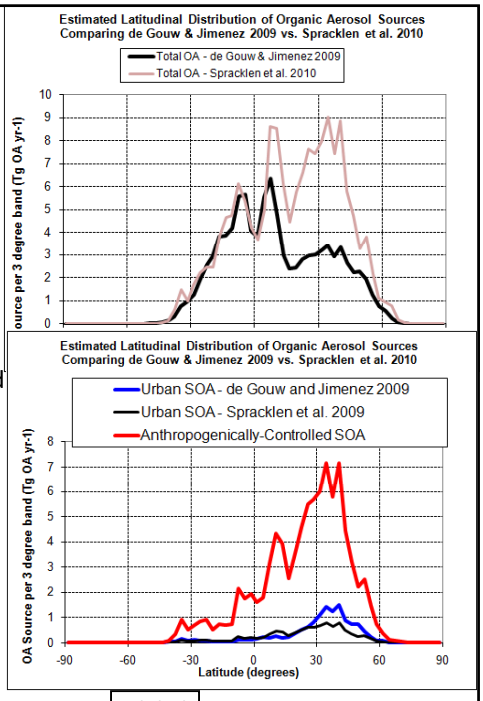


Spracklen, Jimenez, et al., ACPD, 2011

Estimates of Global OA Sources vs. Latitude

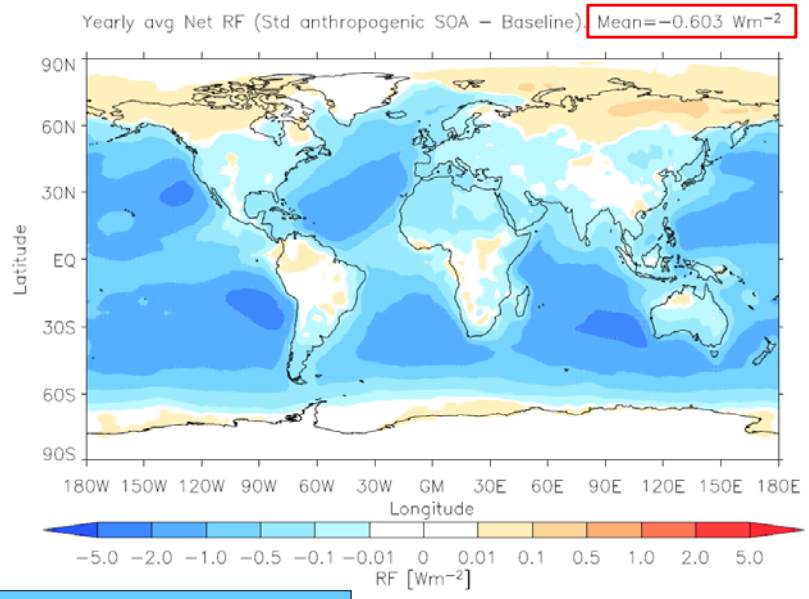
- Most sources are consistent with estimates in de Gouw & Jimenez (2009)
- New very large "anthropogenically-controlled" SOA in N hemisphere
  - Best modeled with VOC tracer based on anthropogenic CO
  - Mostly modern C
  - Implies very strong enhancement of biogenic SOA by pollution
    - x10 higher than in current models
- Extra forcing: -0.86 W m^{-2}

⇒ Higher climate sensitivity



Spracklen, Jimenez et al. ACPD, 2011

Indirect Forcing from Antrop.-Controlled SOA



Spracklen, Jimenez et al. ACPD, 2011

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Conclusions & Challenges

- OA sources
  - SOA/ΔCO approach captures observations and allows model tuning
  - Pollution SOA much larger than in older models
    - Progress in modeling it, but mechanisms unclear
  - Anthropogenically-controlled biogenic SOA may be dominant
    - Large implications for preindustrial vs present and future forcing
- This is a problem where ASR can make a difference
  - Well-designed experiments should shed light in ~3-5 yrs
  - Amazon campaign in 2013 (Martin & Wang)
  - Community move towards SE US campaign in 2013-14, interest in DOE participation
  - Possible “focus group” discussed on Thu

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