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

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Based on two papers:

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Worldwide OOA ~ SOA

Jimenez et al. Science 2009

Intro Regional Global Future
Organic Aerosol (OA): Continental Sources

Emitted Gas Phase Compounds

REACTIONS (Secondary)

Organic Aerosol

EMISSIONS (Primary)

"Pollution" "Biomass Burning" "Biogenic" "Pollution" "Biomass Burning"

Observed OA/ΔCO in Polluted Regions

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

DeCarlo et al., ACP, 2010

Primary Urban ER (de Gouw and Jimenez 2009)
C-130 Urban Fires over Crete 3/23/06 (high CO)
C-130 Urban Fires (early outflow) 3/23/06
C-130 (low CO) Range (this study)
Mexican City 5-4 April 2003 (Kreisberg et al. 2009)
Mexican City Outflow (Kreisberg et al. 2007)
NE US outflow FTCT-2002 (de Gouw et al. 2003)
NE US outflow FTCT-2004 (Park et al., 2007)
NE US outflow FTCT-2002 (Skidmore et al., 2007)
Observed Range of urban POA + SOA (de Gouw and Jimenez, 2009)
Parameterizing SOA/ΔCO

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

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!
Global Model Optimization of SOA Sources

OOA: Model Base case vs. AMS

Increase Biogenic SOA

Increase Anthrop. SOA

OC Model Base case vs. IMPROVE

Summer

Winter

R²=0

R²=0

R²=0.3

Spracklen, Jimenez, et al., ACPD, 2011

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
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
  $\Rightarrow$ Higher climate sensitivity

Spracklen, Jimenez, et al., ACPD, 2011
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